

Electron dynamics for atoms driven by intense and elliptically polarized laser pulses

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École doctorale: Physique et Sciences de la Matière Discipline: Physique Théorique et Mathématiques

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Dynamique d'électrons pour des atomes soumis à des impulsions lasers intenses polarisées elliptiquement

Soutenue le 25 September 2019 devant le jury:

Cristel Chandre	Centre National de la Recherche Scientifique	Directeur de thèse
Éric Charron	Université Paris-Sud	Rapporteur
Éléna Floriani	Aix-Marseille Université	Examinateur
Stéphane Guérin	Université de Bourgogne	Président du jury
Àngel Jorba	Universitat de Barcelona	Examinateur
Sergei Popruzhenko	National Research Nuclear University MEPhI	Examinateur
Jan-Michael Rost	Max Planck Institute, Dresden	Rapporteur





Aix-Marseille Université École doctorale ED352: Physique et Sciences de la Matière UFR Sciences Institut de Mathématiques de Marseille UMR 7373

> Thèse présentée pour obtenir le grade universitaire de docteur Discipline: Physique Théorique et Mathématiques

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À ma sœur, Amandy.

Preface

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Despite the rather long list of names cited above, I know some are missing, and have also contributed significantly to make this thesis manuscript a reality.

Résumé en français

Assujettir des atomes ou des molécules à des impulsions laser de fortes intensités donne lieu à une variété de phénomènes hautement non-linéaires, tels que l'ionisation d'électrons et la radiation de photons de fréquences élevées. Les distributions des vitesses des électrons ionisés ou les distributions des fréquences des photons radiés encodent des informations pertinentes sur les atomes ou les molécules ciblés à l'échelle temporelle naturelle des électrons, l'attoseconde-qui est un millionième, d'un millionième, d'un millionième d'une seconde. Comprendre la dynamique des électrons ionisés ainsi qu'identifier les mécanismes de radiation de fréquences élevées sont des étapes essentielles afin d'interpréter et décoder les informations cryptées dans les mesures expérimentales.

Dans cette thèse, des atomes sujets à des impulsions laser de fortes intensités polarisées elliptiquement dans le régime infra-rouge sont étudiés théoriquement. Malgré leur nature fondamentalement quantique dans les atomes, les électrons manifestent certains comportements classiques lorsqu'ils sont sujets à des impulsions laser de fortes intensités. Nous exploitons ces traits classiques pour comprendre et illustrer, à l'aide des trajectoires, les mécanismes physiques en jeu afin d'interpréter les résultats expérimentaux. Nous montrons le rôle interdépendant de l'ionisation quantique par effet tunnel de l'électron et ultérieurement de son mouvement classique pour interpréter les mesures en science attoseconde.

Après l'ionisation par effet tunnel des électrons, la conjugaison entre leurs interactions avec le laser et leurs interactions avec leur ion parent, en rendant leurs dynamique hautement non-linéaire, donne lieu à de riches et variés canaux d'ionisation. Changer l'ellipticité du laser, qui agit comme un simple bouton de contrôle en expérience, change les canaux d'ionisation prioritairement empruntés par les électrons. De cette façon, par exemple, les électrons peuvent sonder différentes caractéristiques des atomes ciblés. Le mouvement des électrons ionisés est analysé en utilisant des techniques perturbatives et non-perturbatives issues de la dynamique non-linéaire et des systèmes hamiltoniens. Ce travail de thèse démontre la complémentarité de la mécanique quantique et de la dynamique non-linéaire pour comprendre et illustrer des mécanismes impliqués lorsque des atomes sont sujets à des impulsions laser de fortes intensités polarisées elliptiquement.

Introduction

Imager des orbitales atomiques et moléculaires [115, 59, 135], traquer le mouvement des électrons en temps réel [25, 64, 126] et mesurer leurs corrélations dans les atomes, les molécules et les solides [101, 72, 110, 141, 18] a été au cœur de l'activité de recherche en *science attoseconde*, avec pour ambition de réaliser des films de la dynamique structurelle des molécules pendant les réactions chimiques qui ont lieu dans la nature [41]. Ces images instantanées révèleraient des mécanismes sous-jacents à l'échelle temporelle et spatiale de l'électron (inférieure à la femtoseconde 1 fs = 10^{-15} s et inférieure au nanomètre 1 nm = 10^{-9} m, respectivement, voir Fig. 1), le liant de la matière, et permettrait ainsi de mieux comprendre et contrôler ces processus ultra-rapides hors équilibre. De nos jours, l'imagerie de la matière à l'échelle temporelle et spatiale des atomes (1 ps = 10^{-12} s et 1 Å = 10^{-10} m, respectivement, voir Fig. 1) est couramment réalisée en laboratoire. Zoomer sur ces images, à des échelles de temps inférieures à la femtoseconde et à des échelles spatiales inférieures au nanomètre, demeure un défi majeur pour les scientifiques tant sur le plan technique que théorique depuis une trentaine d'années.

Par exemple, des images de structures atomiques dans des surfaces solides, à l'échelle spatiale des atomes, peuvent être obtenues en déplaçant une pointe métallique de quelques angströms au-dessus de la surface ciblée. C'est la base de la microscopie à effet tunnel [scanning tunneling microscopy (STM)]. Les premières images ont été observées en 1982 [23] et ont mené à une nouvelle branche de la microscopie: la microscopie de sonde à balayage [scanning probe microscopy (SPM)]. Malgré le succès du STM pour la localisation d'atomes dans des surfaces solides à l'équilibre, les processus ultra-rapides hors équilibres dans la matière ne sont pas mesurables par la pointe métallique du STM. Ceci illustre clairement les difficultés de mesure en temps réel du mouvement des atomes et des électrons. *Grosso modo*, comme l'illustre la Fig. 2, seuls les appareils sondant la matière à une échelle de temps beaucoup plus rapide que l'échelle de temps naturelle des atomes et des électrons peuvent capturer des images instantanées claires de leurs configurations. Ainsi, la pointe métallique devrait se déplacer sur des échelles de temps inférieures à la femtoseconde pour capturer le mouvement des électrons en temps réel, ce qui est impossible.

Entre-temps, la *femtochimie* en était à ses débuts. En femtochimie, de courtes impulsions laser infrarouges (IR), de l'ordre de la femtoseconde, sont utilisées en tant que caméras ultra-rapides pour photographier les atomes en mouvement pendant des réactions moléculaires, une avancée décisive pour la technologie d'imagerie ultra-rapide. En 1987, lorsque les impulsions laser deviennent suffisamment courtes pour obtenir des images

instantanées claires (quelques femtosecondes), le mouvement d'atomes pendant une réaction chimique est mesuré pour la première fois en temps réel par spectroscopie pompe-sonde [174] (pump-probe spectroscopy). Néanmoins, les électrons dans les atomes et les molécules se déplacent sur des échelles de temps inférieures à la femtoseconde, qui sont plus de mille fois plus rapides que celles des atomes dans les molécules et les solides. Pour mesurer en temps réel le mouvement des électrons pendant des réactions moléculaires, les impulsions laser de la sonde doivent être plus de mille fois plus courtes que celles utilisées en femtochimie, c'est-à-dire qu'elles doivent être des sources lumineuses d'une durée de l'ordre de l'attoseconde (1 as = 10^{-18} s). À la fin des années 90 démarre ainsi la ruée vers les impulsions laser ultra-courtes: un effort théorique et technologique.

Les lasers à électrons libres [69] [free-electron lasers (FELs)] constituent un premier type de sources d'impulsions laser ultra-courtes. Ils nécessitent des ressources de très haute énergie et des infrastructures à grande échelle. À l'opposé, l'autre type de sources d'impulsions laser ultra-courtes s'installe sur des paillasses de taille relativement petites, et peut ainsi être facilement exploité en laboratoire. La première étape menant à la production de ces impulsions laser ultra-courtes a été découverte en 1987, lorsque la génération de photons d'harmoniques élevées [high harmonic generation (HHG)] a été observée pour la première fois en propageant des impulsions laser IR femtosecondes à travers un gaz d'atomes [168]. Lorsqu'une impulsion laser IR se propage dans un milieu constitué d'atomes ou de molécules, ce dernier émet des photons dans le régime des ultraviolets extrêmes [extreme ultraviolet (XUV)]. Le spectre d'intensité HHG correspond à la distribution des fréquences (ou des harmoniques) des photons radiés. Des spectres d'intensité HHG typiques pour un gaz atomique sont représentés dans la Fig. 3 pour différentes intensités du laser. Pour les gaz isotropes, tels que les gaz d'atomes, la fréquence des photons rayonnés est un multiple impair de la fréquence du laser IR [102]. Le spectre d'intensité HHG typique est composé de trois régions distinctes. Quelle que soit l'intensité du laser, l'intensité des harmoniques décroit de manière exponentielle jusqu'à la cinquième harmonique. C'est ce qu'on appelle la région des harmoniques inférieures au seuil [below threshold harmonics (BTH)]. Pour des harmoniques plus élevées, l'intensité des harmoniques est plutôt constante, on parle de la région du plateau. Une coupure abrupte suit la région du plateau, à l'harmonique de coupure, après quoi l'intensité des harmoniques diminue soudainement. Ensuite, l'intensité des harmoniques est extrêmement faible. Sur la Fig. 3, nous observons que l'harmonique de coupure augmente lorsque l'intensité du laser IR augmente.

Les premières observations de la région du plateau dans les spectres d'intensité HHG [54] ont été très prometteuses pour la production d'impulsions laser ultra-courtes. En effet, un peigne de fréquence peut être obtenu après avoir isolé la région du plateau en filtrant le spectre d'intensité HHG. Après traitement, le faisceau de photons résultant est un train d'impulsions laser XUV ultra-courtes. Plus l'harmonique de coupure est grande et plus l'impulsion laser est courte. En 2001, une unique impulsion attoseconde [70] et un train d'impulsions attosecondes [132] ont été observés pour la première fois expérimentalement. De nos jours, les sources de lumière ultra-courtes sur paillasse [153, 35] sont couramment utilisées en laboratoires. Les impulsions laser XUV ultra-courtes ont ouvert l'accès aux mesures de la dynamique électronique avec une résolution de l'ordre de l'attoseconde [56, 67, 64, 155, 87, 135].

État de l'art

La productivité et les technologies de la science attoseconde ont nettement progressées depuis 1993, après la découverte du mécanisme sous-jacent à la région du plateau dans le spectre d'intensité HHG: les recollisions [92, 39, 154]. Une recollision se produit lorsqu'un électron ionisé retourne dans son ion parent. De façon remarquable, alors que le HHG est produit en propageant une impulsion laser IR à travers un gaz atomique ou moléculaire, le mécanisme clé du HHG peut être compris à partir de la réponse d'un unique atome avec un seul électron actif [100, 146]. En plus du HHG, les recollisions donnent lieu à une variété de phénomènes non linéaires et sont désormais considérées comme la clé de voûte de la science attoseconde. Dans la Fig. 4, les courbes rouges représentent le taux de publications (panneau de gauche) et le taux de citations (panneau de droite) depuis la base de données de Web of Science avec "recollision" comme sujet. Nous observons que, depuis lors, l'intérêt suscité par les recollisions n'a cessé d'augmenter.

Recollisions: clé de voûte de la science attoseconde La physique du HHG est bâtie sur un scénario de recollision en trois étapes [39, 154], qui est illustré dans la Fig. 5. Nous considérons un atome à un seul électron actif [single-active electron (SAE)] sujet à un champs laser $\mathbf{E}(t) = \mathbf{e}_x E_0 \cos(\omega t)$ polarisé linéairement [linearly polarized (LP)]. Le champ laser oscille en temps le long de l'axe de polarisation \mathbf{e}_x à une fréquence ω . Si l'amplitude du laser E_0 est suffisamment grande, un électron peut être arraché du noyau par le laser: il s'agit de l'étape (i) du scénario de recollision. Au cours de l'étape (i), un paquet d'onde électronique ionise par effet tunnel à travers la barrière de potentiel induite par le champ laser.

À l'étape (ii) du scénario de recollision [39, 154], le mouvement de l'électron est traité de manière classique. L'électron est initié sur la barrière de potentiel dans la direction opposée à celle du champ laser. Son mouvement est le long de l'axe de polarisation. À partir du principe fondamental de la dynamique $\ddot{x} = -E_0 \cos(\omega t)$, la position de l'électron en fonction du temps est donnée par l'Éq. (1), où $x(t_0) = x_0$ et $\dot{x}(t_0) = 0$ sont respectivement la position et la vitesse de l'électron initié sur la barrière de potentiel au temps d'ionisation t_0 . Les unités atomiques sont utilisées dans tout le manuscrit, sauf indication contraire. Nous observons que le mouvement de l'électron est composé principalement de deux composantes: une dérive dans le terme $\sin(\omega t_0)\omega t$ et des oscillations dans le terme $\cos(\omega t)$. L'électron ionise relativement près du noyau, à $x_0 \sim 1$ a.u. En revanche, l'ordre de grandeur de l'amplitude des oscillations de l'électron dans le champ laser est $E_0/\omega^2 \sim 10$ a.u. pour les paramètres du laser utilisés dans la Fig. 3. Lors de l'excursion de l'électron en dehors de l'ion parent, il est redirigé vers son origine après que le champ laser ait atteint son amplitude maximale et retourne dans l'atome: ceci est une recollision.

Lors de l'étape (iii), l'électron interfère et recombine avec la partie de la fonction d'onde qui est restée dans l'état fondamental de l'atome. Le temps de la recombinaison de l'électron de recollision, noté $t_0 + \Delta t$, est tel que $x(t_0 + \Delta t) \approx 0$, avec Δt le temps d'excursion hors de l'ion parent. Lors de la recombinaison, un photon de fréquence Ω est émis selon la loi décrite par l'Éq. (2), où I_p est le potentiel d'ionisation de l'atome. La partie droite de l'Éq. (2) est l'énergie de l'électron lors de la recollision en fonction du temps d'ionisation t_0 , du temps de recombinaison $t_0 + \Delta t$ et de l'énergie pondéromotrice $U_p = E_0^2/4\omega^2$. Dans le spectre d'intensité HHG, le BTH est constitué de photons émis à une fréquence $\Omega < I_p$. La région du plateau est constituée de photons émis par des processus de recollision [39, 100, 146, 166, 20]. Dans l'Éq. (2), nous observons que la fréquence des photons rayonnés dépend du temps auquel les électrons de recollision ionisent et recombinent. Nous supposons que $x_0 \approx x(t_0 + \Delta t) \approx 0$. En utilisant l'Éq. (1), cela implique que $\sin(\omega t_0) = [\cos(\omega t_0) - \cos(\omega t_0 + \omega \Delta t)]/\omega \Delta t$. Cette condition fixe le temps d'excursion des électrons de recollision en fonction de leur temps d'ionisation. Nous substituons cette condition dans l'Éq. (2). Nous maximisons la fréquence du photon radié en fonction du temps d'ionisation de l'électron ionisé t_0 et de leur temps d'excursion Δt , et nous obtenons la fréquence maximale du photon radié par le processus de recollision [39, 100], donnée par l'Éq. (3) pour $\omega \Delta t \approx 4.09$ et $\omega t_0 \approx 0.31$. Par conséquent, les trajectoires qui ramènent le plus d'énergie à l'ion parent, de l'ordre de $3.17 U_p$, ionisent peu après que le laser ait atteint son amplitude maximale, et recollisionnent à peu près un demi cycle laser plus tard. Pour un laser de longueur d'onde $\lambda = 1064$ nm et des lasers d'intensité $I = 3 \times 10^{13}$ W · cm⁻², $I = 1.3 \times 10^{13}$ W · cm⁻², $I = 9 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}, I = 7 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}, I = 5 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}$ (intensités de laser utilisées dans la Fig. 3), les harmoniques de coupure prédite par l'Éq. (3) sont (\circ) $\Omega_c/\omega \approx 19$, (\bullet) $\Omega_c/\omega \approx 14$, (\Box) $\Omega_c/\omega \approx 13$, (I) $\Omega_c/\omega \approx 12$ et (Δ) $\Omega_c/\omega \approx 12$, respectivement. La loi de coupure prédite par l'Éq. (3) est en accord avec les mesures expérimentales de la Fig. 3, en particulier pour des intensités $I > 7 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}$. Le mécanisme de recollision et le processus physique donnant lieu au HHG sont résumés dans la Fig. 6. La Fig. 6a montre l'installation expérimentale dans laquelle un gaz d'atomes est sujet à une impulsion laser IR intense. La Fig. 6b représente schématiquement le mouvement de l'électron dans un atome sujet au champ laser. La Fig. 6c représente l'harmonique correspondant à l'énergie de retour de l'électron en fonction de son temps d'ionisation (panneau du haut) et un spectre d'intensité HHG en fonction de l'harmonique des photons radiés. A chaque fréquence de photons radiés dans la région du plateau est associée une énergie de retour d'un électron de recollision.

En résumé, la mécanique classique a été capable de dévoiler le mécanisme physique sous-jacent HHG, produit lorsque des atomes sont sujets à des impulsions laser intenses. En particulier, la loi de l'harmonique de coupure prédite par le modèle de recollision (3) est en accord avec les mesures expérimentales [102]. La durée du champ laser après traitement du signal est inversement proportionnelle à la valeur de l'harmonique de coupure. Pour augmenter l'harmonique de coupure de manière significative, la fréquence du laser doit être abaissée ou son amplitude doit être augmentée. Néanmoins, l'intensité et la fréquence du champ laser sont limitées par les outils technologiques disponibles. Augmenter la fréquence de coupure n'est pas tâche facile, et est au cœur de nombreuses études (voir Réfs. [7, 44], e.g.).

Le succès du scénario de récollision pour décrire la production de HHG démontre l'existence de caractéristiques classiques dans ces systèmes fondamentalement quantiques. Étant donné qu'il s'agit ici d'un point de rencontre entre la mécanique classique et la mécanique quantique, un autre rôle important de la science attoseconde est de tester et de remettre en question des concepts, méthodes et interprétations issus de cette mixture de mécanique classique et quantique. Par exemple, le "temps que l'électron passe sous la barrière de potentiel" pour ioniser pendant l'étape (i), appelé "temps de tunnel" dans les Réfs. [95, 94], demeure un débat dans la communauté [30, 155, 10, 134, 95, 94, 124, 60, 152, 149]. Dans cette thèse, nous exploitons les caractéristiques classiques de la dynamique électronique après ionisation afin de comprendre et d'interpréter, à l'aide des trajectoires, les phénomènes non linéaires observés dans des atomes sujets à des impulsions laser intenses et polarisées elliptiquement.

Phénomènes non-linéaires Au vu du succès de la prédiction de l'harmonique de coupure par le modèle en trois étapes, obtenue en ignorant l'interaction entre l'électron et son ion parent, nous pourrions penser que cette dernière joue un rôle mineur dans le mécanisme de production de sources de lumière ultra-courtes. Il est pourtant dominant lorsque l'électron revient dans l'atome. En expérimentant de fortes interactions avec l'ion parent, l'électron sonde ses structures et sa dynamique électronique interne. Si l'électron émet des photons, ces informations se retrouvent dans le spectre d'intensité HHG. La spectroscopie des harmoniques élevées est une technique permettant d'extraire ces informations depuis les spectres d'intensité HHG. Par exemple, la spectroscopie à hautes harmoniques est utilisée pour mesurer la migration de charge dans les molécules [90].

Un premier effet évident de l'interaction ion-électron à l'intérieur des atomes est la déviation de l'électron de recollision au moment du retour (voir le panneau central de la Fig. 7). La combinaison des fortes interactions de l'électron avec le laser et l'ion parent peut produire des électrons ionisés de hautes énergies [39, 133]. Il s'agit d'une ionisation au-dessus du seuil [above-threshold ionization (ATI)]. La déviation des électrons de recollision par l'ion parent est appelée la rediffusion de l'électron. Au cours de la rediffusion, les électrons sondent la structure interne et la dynamique à l'intérieur de l'atome et des molécules ciblées [99]. Les distributions des électrons ionisés au niveau du détecteur, appelées distributions de moments photoélectroniques [photoelectron moementum distributions (PMDs)], encodent ces informations [115, 76]. C'est la base d'une majorité de techniques d'imagerie, par exemple en diffraction électronique induite par le laser [176, 115, 139] [laser induced electron diffraction (LIED)] pour l'imagerie moléculaire [25] et en holographie photoélectronique [76, 22]. Ces techniques sont souvent perçues comme des expériences de "demi-diffusion", en analogie avec les expériences de "diffusion" de Rutherford [151] en 1911, dans lesquelles des particules sont envoyées depuis l'infini afin de sonder des atomes ciblés. Ici, les particules sondant la cible proviennent des cibles elles-mêmes.

Un autre effet évident des interactions ion-électron est de contenir les électrons au voisinage du noyau. En particulier, les interactions ion-électron peuvent contenir des électrons au voisinage du noyau pendant que des électrons de recollision font une excursion hors de l'ion parent. Lorsqu'un électron de recollision retourne dans l'atome il échange de l'énergie avec un électron lié et les deux peuvent ioniser [39, 110] (voir le panneau inférieur dans la Fig. 7). Il s'agit d'une double ionisation non séquentielle [nonsequential double ionization (NSDI)]. La NSDI est l'une des manifestations les plus dramatiques de la corrélation électron-électron dans la nature [17]. Les PMDs des atomes doublement ionisés encodent des informations sur les corrélations électron-électron [141, 18].

En résumé, les recollisions, parce qu'elles impliquent la combinaison de fortes interactions des électrons avec le champ laser et les ions parents, donnent lieu à une variété de phénomènes hautement non linéaires [39, 16, 41, 5, 91, 17] résumés dans la Fig. 7. Des techniques d'imagerie basées sur ces phénomènes hautement non linéaires offrent des perspectives très prometteuses. Cependant, en raison de leur inhérente technicité et de la difficulté fondamentale à évaluer le mouvement des électrons en interaction avec un laser et leur ion parent, elles ne sont pas encore des procédures couramment utilisées dans les laboratoires pour mesurer en temps réel le mouvement des électrons dans la matière. En particulier, des films de dynamique électronique dans de grandes molécules biologiques polyatomiques n'ont pas été observés jusqu'alors. La science attoseconde en est encore à un stade relativement précoce.

Ellipticité du laser et potentiel de Coulomb La force de l'interaction ion-électron la plus fondamentale est quantifiée par le potentiel de Coulomb. Alors que le rôle du potentiel de Coulomb est évident dans les processus de rediffusion (en ATI) et dans le piégeage des électrons proches de leur ion parent (en NSDI), il n'en va pas de même pour ses manifestations dans la dynamique des électrons ionisés. L'ellipticité du champ laser qui, expérimentalement, peut être utilisé comme un simple bouton de commande, met en évidence différents phénomènes issus de la dynamique électronique [9, 63, 156, 96, 75, 47, 68, 36, 43, 114]. La Fig. 4 montre le taux de citations et de publications ayant pour thème "Impulsions laser à polarisation elliptique" et "Coulomb dans les impulsions laser intenses". Depuis 1990, l'intérêt porté à ces sujets s'est considérablement accru. En particulier, puisqu'il a été constaté que le changement d'ellipticité du laser IR met en évidence l'impact du potentiel de Coulomb sur le mouvement des électrons ionisés, appelés *effets de Coulomb* [28, 11, 62, 38, 93]. En d'autres termes, en modifiant l'ellipticité du laser, il a été constaté que le potentiel de Coulomb est en réalité important, même après l'ionisation de l'électron.

La Fig. 8 montre les PMDs et les distributions angulaires photoélectroniques [photoelectron angular distributions (PADs)] des atomes de He pour différentes ellipticités du laser. Les PMDs sont les distributions

de la quantité de mouvement des électrons ionisés au niveau du détecteur, et les PADs sont les distributions de l'angle de la quantité de mouvement des électrons ionisés par rapport à l'axe majeur de polarisation. Le champ laser est de la forme $\mathbf{E}(t) = E_0 f(t) [\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \xi \sin(\omega t)] / \sqrt{\xi^2 + 1}$, où \mathbf{e}_x et \mathbf{e}_y sont respectivement l'axe majeur et mineur de polarisation. L'ellipticité du laser est ξ et son enveloppe est f. La durée du laser est relativement longue, de sorte que l'enveloppe du laser est $f \approx 1$ pour plusieurs cycles laser. Dans la Fig. 8, nous observons que lorsque l'ellipticité du laser est modifiée, la forme des PMDs subit des modifications drastiques.

Dans la Fig. 8, pour $\xi = 0$ (champs LP), la PMD est un lobe centré autour de l'origine de l'espace des moments. Dans la PAD, nous observons que ce nuage est légèrement asymétrique par rapport à l'axe mineur de polarisation. Pour $\xi = 0.15$, les PMDs se divisent en deux lobes symétriques par rapport à l'origine de l'espace des moments. Pour augmenter l'ellipticité, les deux lobes s'éloignent davantage. Le cadre théorique naturel afin d'interpréter ces mesures consiste à appliquer la même procédure que celle utilisée pour décrire le scénario de recollision, à savoir: (i) l'électron ionise par effet tunnel, (ii) le mouvement de l'électron est traité classiquement sans tenir compte du potentiel de Coulomb. L'électron ionise en dehors de la barrière de potentiel induite par le champ laser, son impulsion initiale est donc $\mathbf{p}_0 = \mathbf{0}$. D'après le principe fondamental de la dynamique, $\dot{\mathbf{p}} = -\mathbf{E}(t)$, l'impulsion de l'électron est donnée par $\mathbf{p} = \mathbf{A}(t) - \mathbf{A}(t_0)$, où t_0 est le temps d'ionisation. Le potentiel vecteur est $\mathbf{A}(t)$ tel que $\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$. Asymptotiquement, au niveau du détecteur, le potentiel vecteur du champ laser est nul et donc $\mathbf{p}(t) = -\mathbf{A}(t_0)$. Le temps d'ionisation le plus probable de l'électron est tel que l'amplitude du champ laser est la plus grande, c'est-à-dire que $\omega t_0 = k\pi$ avec $k \in \mathbb{Z}$. Par conséquent, en l'absence du potentiel de Coulomb, le moment asymptotique de l'électron le plus probable est donné par l'Éq. (4). Pour $\xi = 0$ (champs LP), le moment asymptotique le plus probable de l'électron est égal à zéro, conformément aux mesures expérimentales de la Fig. 8. Pour des ellipticités plus grandes, le moment asymptotique le plus probable de l'électron s'éloigne à partir de l'origine le long de l'axe mineur de polarisation, en accord qualitatif avec les mesures expérimentales des panneaux supérieurs de la Fig. 8. Pour des ellipticités croissantes, l'énergie de dérive de l'électron augmente, et par conséquent, l'électron s'éloigne de plus en plus vite de l'ion parent. L'électron ionise sans recollision.

Afin d'évaluer ces changements qualitatifs des PMDs, dans la Réf. [93], la localisation du pic du lobe supérieur des PMDs est suivie en fonction de l'ellipticité du laser (panneau inférieur de la Fig. 8), notée $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$. Dans le panneau inférieur de la Fig. 8, les cercles bleus et rouges sont les coordonnées des moments du pic du lobe supérieur dans les PMDs, P_x et P_y , respectivement. La courbe verte continue est la composante positive de P donnée par l'Éq. (4) le long de l'axe mineur de polarisation \mathbf{e}_y . La courbe noire continue est la composante de **P** donnée par l'Éq. (4) le long de l'axe majeur de polarisation \mathbf{e}_x . Pour $\xi \lesssim 0.1$, nous observons que $\mathbf{P} \approx \mathbf{0}$, par conséquent, la PMD est principalement constituée d'un lobe centré à l'origine. En revanche, la courbe verte augmente linéairement dans cette plage d'ellipticités. La persistance du lobe unique dans les PMDs, révélée par le panneau inférieur de la Fig. 8, est appelée la focalisation de Coulomb [28, 38]. Pour $\xi \gtrsim 0.1$, nous observons que lorsque P_y devient positif, P_x devient négatif. Dans cette plage d'ellipticités, les cercles rouges correspondent bien à la courbe verte. Cependant, il y a une différence notable entre la courbe noire horizontale et les cercles bleus. En effet, P_x est négatif et non nul. Dans les PMDs, nous observons une asymétrie par rapport à l'axe mineur de polarisation. Cette asymétrie, révélée par le panneau inférieur de la Fig. 8, est appelée l'asymétrie de Coulomb [11, 62]. Dans les mesures expérimentales reportées dans la Réf. [104], l'impulsion finale de l'électron présente les mêmes caractéristiques que dans les expériences [140, 93] (voir Fig. 3.13). L'hypothèse faite dans la Réf. [93] est qu'il existe une *bifurcation* lorsque varie l'ellipticité du champ laser.

En résumé, le changement d'ellipticité révèle les effets du potentiel de Coulomb et modifie le canal d'ionisation prioritaire emprunté par les électrons. Ainsi, par exemple, les électrons peuvent sonder différentes caractéristiques des atomes ciblés. Cependant, il y a toujours eu des obstacles fondamentaux à la compréhension et à l'évaluation de l'impact du potentiel de Coulomb sur les trajectoires des électrons ionisés. En particulier, l'entrelacement entre l'interaction des électrons ionisés avec le laser et l'interaction des électrons ionisés avec leur ion parent donne lieu à de multiples échelles temporelles et spatiales, ce qui confère à la dynamique des électrons ionisés une très haute non linéarité.

Travail de thèse

Dans cette thèse, nous considérons des atomes d'électrons actifs simples (à l'exception de la Sec. 4.3 où une extension de l'étude est réalisée pour les atomes à deux électrons actifs) sujets à des impulsions laser intenses et polarisées elliptiquement. La dynamique de l'électron est étudiée dans le cadre quantique et classique. Dans le cadre classique, des outils issus de la dynamique non linéaire et du formalisme hamiltonien sont utilisés pour étudier le mouvement de l'électron.

Objectifs de la thèse L'objectif général de cette thèse est de comprendre les changements de la dynamique des électrons dans les atomes sujets à des impulsions laser intenses pour différentes ellipticités, ainsi que leur impact sur les phénomènes hautement non linéaires observés en expériences. L'un des objectifs est de décrire le mouvement des électrons dans le cadre classique, afin d'exploiter des outils issus de la dynamique non linéaire, tout en tenant compte de la nature quantique des électrons dans les atomes. Un autre but consiste ensuite à évaluer le rôle du potentiel de Coulomb dans la dynamique des électrons ionisés et dans l'excursion des électrons de recollision. Cela consiste à identifier les manifestations du potentiel de Coulomb dans des mesures expérimentales ou numériques et à déterminer les mécanismes sous-jacents en termes de trajectoires d'électrons. Pour se faire, une autre visée est d'isoler la contribution du champ laser et de l'interaction coulombienne, afin d'identifier et de décrire le rôle du potentiel de Coulomb dans les phénomènes non linéaires observés en science attoseconde.

Méthodes Dans ce manuscrit de thèse, nous détaillons les investigations numériques et analytiques qui ont été effectuées. Nous montrons:

- La dérivation d'une hiérarchie de modèles réduits pour décrire la dynamique de l'électron dans un champ laser intense en termes de la dynamique de son centre guide.
- Le calcul de structures invariantes de haute dimension dans l'espace des phases (variétés invariantes d'une famille de tores invariants unidimensionnels).
- La représentation de structures invariantes de grande dimension en deux dimensions.
- Le calcul des simulations de Monte Carlo à trajectoires classiques [classical trajectory Monte Carlo (CTMC)].
- Les résultats des statistiques de haute résolution calculées à l'aide de cartes graphiques [graphics processing units (GPU)].

Chapitre 1. Modèle Hamiltonien pour des atomes sujets à des impulsions laser intenses polarisées elliptiquement

Dans ce chapitre, nous dérivons les modèles hamiltoniens pour des atomes à un électron actif sujets à des impulsions laser intenses et polarisées elliptiquement dans le cadre de la mécanique quantique et classique. En comparant la distribution de quasi-probabilité de Wigner [167] de l'électron et sa distribution classique dans l'espace des phases, nous montrons que la dynamique quantique de l'électron présente des caractéristiques classiques claires après son ionisation. Nous décrivons un modèle en deux étapes dans lequel l'électron (i) ionise par effet tunnel à travers la barrière de potentiel induite par le champ laser et (ii) se déplace classiquement en dehors de la région de l'ion parent. Ce modèle en deux étapes nous permet d'exploiter des outils issus de la dynamique non linéaire tout en tenant compte de la nature fondamentalement quantique des électrons dans les atomes.

Premièrement, le traitement classique de la dynamique des électrons après ionisation nous permet de décrire les mécanismes sous-jacents des phénomènes non-linéaires observés expérimentalement en termes de trajectoires électroniques. Les trajectoires, qui sont des représentations locales dans l'espace, nous permettent de visualiser et d'interpréter plus clairement le mouvement et le comportement des électrons en fonction du temps. Deuxièmement, le modèle en deux étapes nous permet de distinguer entre les phénomènes dus à la fois à la population d'états initiaux de l'ionisation quantique des électrons et à leur mouvement classique ultérieur, et les phénomènes uniquement dus au mouvement classique ultérieur à l'ionisation par effet tunnel. En effet, dans la description classique de la dynamique, les conditions initiales de l'électron ont une importance primordiale. C'est aussi le cas dans des phénomènes comme la bifurcation dans les PMDs (étudiée au Chap. 3) ou l'absence de recollisions dans les champs polarisés circulairement [circularly polarized CP] pour des atomes spécifiques (étudié au Chap. 4), qui sont dues à la fois à la population de conditions initiales par ionisation quantique et au mouvement classique ultérieur de l'électron. En revanche, des phénomènes, tels que la création d'états de Rydberg et les recollisions induites par l'interaction coulombienne (également étudiées au Chap. 3), sont principalement dus au mouvement classique ultérieur de l'électron après son ionisation.

Dans le cadre de la description classique, la dynamique de l'électron est régie par le hamiltonien (1.14). Dans ce manuscrit, deux méthodes issues de la dynamique non-linéaire sont principalement utilisées pour étudier le hamiltonien (1.14). Au Chap. 2, nous utilisons des méthodes perturbatives pour déterminer des modèles réduits permettant de décrire le mouvement de l'électron dans l'espace des phases. Au Chap. 5, nous utilisons des méthodes non perturbatives pour analyser la dynamique de l'électron dans l'espace des phases. En particulier, nous identifions des objets invariants qui structurent l'espace des phases et guident les recollisions.

Chapitre 2. Dynamiques réduites pour des atomes sujets à des impulsions laser intenses polarisées elliptiquement: Le modèle du centre guide

Dans ce chapitre, nous dérivons des modèles réduits d'atomes soumis à des impulsions laser intenses et polarisées elliptiquement, à savoir: l'approximation à champ fort [strong field approximation (SFA)], l'approximation à champ fort perturbé par le potentiel de Coulomb [Coulomb-corrected strong field approximation (CCSFA)] et les modèles du centre guide [guiding center (GC)]. Le SFA néglige le potentiel de Coulomb et le CCSFA le traite comme une perturbation de l'interaction laser. Concernant les modèles du GC, notés G_n , nous établissons une hiérarchie de modèles pour la dynamique du GC. Le mouvement du GC correspond au mouvement moyen de l'électron dans le champs laser et l'interaction coulombienne. La hiérarchie de modèles G_n est dérivée en utilisant les transformées de Lie canoniques et prend en compte le potentiel de Coulomb.

Les modèles réduits G_n sont composés d'un hamiltonien moyen H_m régissant la dynamique du GC [Éqs. (2.17)] et une transformation Φ_n qui permet de passer des coordonnées de l'électron dans l'espace des phases à celles du GC [Éqs. (2.19)]. Grâce à la moyennisation, les hamiltoniens du GC ne dépendent pas explicitement du temps. On peut ainsi définir une énergie pour les électrons à partir de l'énergie de leur GC. De manière générale, ces modèles sont pertinents lorsque l'électron est relativement éloigné de l'ion parent (lorsque la distance entre l'ion parent et l'électron est supérieure à la distance caractéristique de l'électron dans le laser). Ces modèles ne décrivent pas les événements brefs où l'électron est diffusé par l'ion parent lors de la recollision. Cependant, ces modèles capturent la dynamique de l'électron sur des échelles de temps longues, et peuvent ainsi décrire les recollisions qui ont lieu sur plusieurs cycles laser ou la capture de l'électron dans un état de Rydberg.

Nous avons distingué deux modèles G_2 et G_5 : le premier modèle fournit le comportement principal des électrons, il est le plus maniable en raison de sa simplicité et le plus robuste près de l'ion parent. Afin d'améliorer l'accord quantitatif, un modèle d'ordre supérieur tel que G_5 doit être utilisé. Tous ces modèles permettent de faire la distinction entre ionisations directes et rediffusées par l'ion parent grâce au critère de conservation d'énergie de la trajectoire du GC. Ceci peut-être particulièrement utile pour l'analyse des PMDs pour l'imagerie atomique ou moléculaire. Les événements de rediffusion peuvent être vus comme des sauts d'énergie du GC résultant du transfert d'énergie de l'ion parent à l'électron.

Chapitre 3. Impact du potentiel de Coulomb en ATI: Les recollisions engendrées par l'interaction coulombienne et la création d'états de Rydberg

Dans ce chapitre, nous étudions le rôle du potentiel de Coulomb durant l'étape (ii) du processus d'ionisation en utilisant quatre modèles, à savoir: le modèle hamiltonien de référence (1.14), le SFA [Éqs. (2.2)], le CCSFA [Éqs. (2.3)] et le modèle GC au second ordre [$G_2 = (H_2, \Phi_2)$, voir Tab. 2.1]. L'analyse de ces trois modèles réduits nous permet de mettre en lumière les manifestations du potentiel de Coulomb dans divers processus d'ionisation. Le SFA est utilisée pour cibler les contributions du potentiel de Coulomb, comme nous l'avons fait avec l'Éq. (4) et la Fig. 8. Le CCSFA et le modèle GC de second ordre sont utilisés pour découpler les contributions du laser et du potentiel de Coulomb.

Dans le SFA, il existe principalement deux types de trajectoires: les recollisions plus courtes qu'un cycle laser et les ionisations directes. Cependant, même lorsque l'intensité est très grande, c'est-à-dire lorsque les conditions du SFA sont réunies, l'interaction coulombienne se fait toujours ressentir lors des phénomènes à longue portée. En particulier, même à de très hautes intensités, l'asymétrie de Coulomb persiste, comme le montre la Fig. 3.3 et est discutée dans la Sec. 3.1.1. L'interaction coulombienne apporte une variété de différents types de trajectoires supplémentaires, tels que les recollisions engendrées par l'interaction coulombienne et les états de Rydberg. Nous montrons dans la Sec. 3.2 que ces deux processus sont intimement liés et peuvent être interprétés et prédits par le modèle de GC.

Au cours de l'étape (ii) du scénario de recollision, nous avons montré que l'électron oscille autour de la trajectoire du GC. Dans l'espace des phases, la trajectoire du GC repose sur une variété d'énergie constante $\mathcal{E} = H_2(\bar{\mathbf{r}}, \bar{\mathbf{p}})$. Si $\mathcal{E} > 0$, le mouvement du GC est non lié. Dans ce cas, il est probable que l'électron recollisionne si le moment angulaire de son GC est proche de zéro et si son moment radial initial est négatif (comme sur la Fig. 3.7b). Sinon, l'électron ionise directement sans recollisionner (comme sur la Fig. 3.7c). Si $\mathcal{E} < 0$, le mouvement du GC est lié. Dans ce cas, il existe au moins un instant lors duquel l'électron

revient dans la direction de l'ion parent, lorsque son moment radial s'annule. Ensuite, l'électron peut ou non retourner dans l'atome (recollisionner) avant que le champ laser ne soit éteint. Si l'électron recollisionne avant que le champ laser soit éteint (comme sur les Figs. 3.7d et 3.8d), l'énergie du GC passe à un nouveau niveau d'énergie. Si le champ laser est désactivé avant que l'électron recollisionne (comme sur les Figs. 3.7e et 3.8e), l'électron finit sa route sur la trajectoire du GC et est capturé dans un état de Rydberg.

Le modèle GC ne prend pas en compte les effets de rediffusion proches de l'ion parent, mais le CCSFA peut, car il s'agit d'un phénomène sur une échelle temporelle relativement courte [82, 84, 85, 114]. Comme observé sur la Fig. 3.15, les variations d'énergie du modèle de référence (1.14) peuvent être bien décrites par le CCSFA sur des échelles de temps courtes. Après rediffusion, l'électron ionise potentiellement si l'énergie de son GC devient positive (comme dans la Fig. 3.7d). Par conséquent, les modèles CCSFA et GC sont clairement complémentaires. Le CCSFA est adapté pour la description de processus à temps courts, tels que la rediffusion, tandis que les modèles GC conviennent davantage à la description de processus à temps long, tels que les recollisions induites par l'interaction coulombienne et la création d'états de Rydberg.

Enfin, nous montrons que le changement d'ellipticité modifie le rendement des différents types de trajectoires après l'ionisation par effet tunnel. En particulier, pour des ellipticités croissantes, le domaine de rediffusion pour lequel $\mathcal{E} < 0$, dans lequel les recollisions induites par l'interaction coulombienne et les états de Rydberg sont créés, se déplace vers des régions des conditions initiales à faible taux d'ionisation. Par conséquent, le rendement de la création d'états de Rydberg diminue considérablement avec l'augmentation de l'ellipticité, comme observé dans le panneau de gauche de la Fig 3.1. De plus, nous montrons que le comportement de la trajectoire-T change radicalement avec l'augmentation de l'ellipticité. De plus, le moment asymptotique de la trajectoire-T subit une bifurcation en fonction de l'ellipticité du laser et permet ainsi d'interpréter la bifurcation observée expérimentalement (voir le panneau inférieur de la Fig. 8). Près des champs LP ($\xi = 0$), l'énergie du GC de la trajectoire-T est négative et l'électron recollisionne avec une forte probabilité. À l'ellipticité critique ξ_c , l'énergie du GC de la trajectoire-T change de signe et le mouvement de son GC devient non lié. L'électron subit probablement une ionisation directe pour $\xi > \xi_c$. Par conséquent, à $\xi = \xi_c$, le rendement des états de Rydberg est beaucoup plus faible que celui à $\xi = 0$. La diminution drastique du rendement des états de Rydberg et la bifurcation dans les PMDs sont dues à la combinaison entre la population d'états classiques par l'ionisation par effet tunnel quantique, et le mouvement classique ultérieur de l'électron impacté par le potentiel de Coulomb.

Chapitre 4. Mécanisme de recollision à hautes ellipticités et leur importance en HHG et NSDI: Rôle de l'enveloppe du champ laser

Dans ce chapitre, nous identifions un canal de recollisions, dans lequel les électrons retournent dans l'atome avec une grande énergie et une forte probabilité, quelque-soit l'ellipticité du champ laser. Ce canal de recollision est dû aux effets de l'enveloppe du las f(t). La compétition entre l'interaction coulombienne et le champ laser crée un canal d'ionisation dès que le champ laser est activé. Une fois que l'électron est en dehors de l'atome, l'amplitude du potentiel vecteur est faible et la dérive latérale de l'électron peut donc être compensée par son moment initial. L'électron ne dérive pas, même pour de hautes ellipticités, et peut par conséquent retourner dans son atome. Ces recollisions sont appelées les recollisions induites par l'enveloppe. Le modèle GC dérivé dans le Chap. 2 nous permet de déterminer les conditions pour lesquelles ces recollisions peuvent être observées dans des champs laser fortement polarisés elliptiquement. Pour des champs laser CP, le potentiel d'ionisation de l'électron doit être tel que $I_p < I_c$, où I_c est le potentiel critique d'ionisation donné par l'Eq. (4.17). Le potentiel critique dépend principalement de la longueur d'onde du laser. Par conséquent, ces recollisions peuvent être observées et se manifester pour des atomes spécifiques et des longueur d'ondes spécifiques. Pour les champs laser fortement polarisés elliptiquement, lors de son excursion dans le continuum, les électrons gagnent de l'énergie principalement à partir des variations de l'enveloppe du laser. Au cours de leur excursion en-dehors de l'ion parent, l'énergie des recollisions induites par l'enveloppe peut augmenter leur énergie de $2U_p$, ce qui permet un retour d'énergie de l'électron \mathcal{E}_r supérieur à $2U_p$. Nous observons également un bon accord entre l'énergie de retour de l'électron dans le SFA et l'énergie de retour du hamiltonien (1.14).

Nous montrons que ce canal de recollision peut être utilisé pour produire du HHG et du NSDI avec des atomes sujets à des champs laser à polarisation elliptique élevée (champs à polarisation proche de CP). Les conditions d'existence de ce canal de recollisions nous permettent de déterminer les conditions pour lesquelles l'augmentation de la double ionisation ou le HHG peuvent être observés dans des atomes sujets à des champs laser CP (voir la Fig. 4.4), et ceux, en accord avec les conditions pour lesquelles les manifestations des recollisions dans des atomes soumis à des champs CP sont expérimentalement mesurées [61, 111] [55, 65, 61]. De plus, nous remarquons que, pour des conditions raisonnables, ces recollisions peuvent avoir lieu dans la

configuration de l'Attoclock, où les recollisions sont toujours supposées inexistantes [53, 152].

Chapitre 5. Structure de l'espace des phases de l'électron de recollision durant l'étape (ii)

Dans ce chapitre, nous étudions l'espace des phases de l'électron lors de l'étape (ii) à l'aide de méthodes non perturbatives. Le potentiel de Coulomb et les interactions laser sont pleinement pris en compte. Les calculs numériques sont détaillés dans l'annexe D. Nous étudions le comportement global des trajectoires à partir de l'analyse des structures invariantes pour d = 1, 2, 3, où d est la dimension de l'espace des configurations. En particulier, nous montrons qu'un ensemble de variétés invariantes d'une famille de tores invariants associés à une orbite périodique issue de la compétition entre les fortes interactions coulombiennes et du laser, guide la dynamique des électrons dans l'espace des phases. Nous montrons que les orbites périodiques de recollisions [recolliding periodic orbits (RPOs)], une famille spécifique d'orbites périodiques introduite dans les Réfs. [81, 80], sont situées au voisinage de cette structure invariante de haute dimension. Nous étudions les symétries des équations du mouvement de l'électron pour différentes ellipticités et leur impact sur les objets invariants. Nous montrons que les structures invariantes clés qui dirigent les recollisions existent indépendamment de l'ellipticité.

Conclusions

La science attoseconde utilise la dynamique connue des électrons ionisés, arrachés de leur atomes ou molécules parents par une impulsion laser intense, pour imager la dynamique structurelle de ces cibles lors de processus ultra-rapides hors équilibre, tels que les migrations de charges et la dynamique électronique lors de réactions chimiques. Dans cette thèse, nous avons montré que, malgré leur nature fondamentalement quantique dans les atomes et les molécules, les électrons manifestent certains comportements classiques lorsqu'ils sont soumis à d'intenses impulsions laser. De manière remarquable, la mécanique classique, à l'aide de trajectoires et d'outils issus de la dynamique non linéaire, s'est avérée très pertinente pour identifier et interpréter les mécanismes sous-jacents des phénomènes non linéaires observés en expériences. Cependant, il existe des obstacles fondamentaux pour interpréter avec précision ces trajectoires électroniques.

La première difficulté consistait à prendre en compte la nature quantique des électrons dans les atomes et les molécules. En effet, dans la nature, les électrons dans les atomes et les molécules sont uniquement décrits avec précision par la mécanique quantique et leur représentation en tant que fonction d'onde. Ce n'est qu'après avoir ionisé, pour des intensités laser suffisamment grandes, que la dynamique quantique des électrons présente clairement des caractéristiques classiques. Dans notre cadre, l'état des électrons avant l'ionisation a été traité de manière quantique. Selon les paramètres, les électrons peuvent ioniser à travers ou au-dessus de la barrière de potentiel induite par le champ laser intense. Alors que l'ionisation à travers la barrière de potentiel est un processus purement quantique, l'ionisation au-dessus de la barrière a également été décrite avec précision par la mécanique classique. Après l'ionisation, le mouvement de l'électron a été traité de manière purement classique en termes de trajectoires. Dans les deux cas, l'ionisation par effet tunnel ou au-dessus de la barrière, les caractéristiques quantiques des électrons étaient contenues dans la distribution des conditions initiales des trajectoires.

La deuxième difficulté était de comprendre et d'analyser les trajectoires des électrons ionisés en tenant compte de l'interaction avec leur ion parent, qui est généralement ignorée mais qui peut manifester sa présence même après l'ionisation. Nous avons montré que l'interaction des électrons ionisés avec le laser et leur ion parent donnent naissance à de multiples échelles temporelles et spatiales, ce qui rend leur dynamique extrêmement non linéaire et donnent naissance à des canaux d'ionisation riches et variés. En modifiant l'ellipticité du laser, qui est utilisé comme un simple bouton de contrôle en expériences, le canal d'ionisation prioritairement emprunté par les électrons est aussi modifié. Afin de comprendre et d'analyser la variété de canaux d'ionisation que peuvent emprunter les électrons ionisés, nous avons utilisé principalement deux méthodes. La première méthode était non perturbative et consistait à comprendre la dynamique des électrons à travers l'analyse de structures invariantes dans l'espace des phases. L'un des avantages de cette technique est de fournir un cadre d'analyse du comportement global des trajectoires plutôt que de leurs comportements individuels. La seconde méthode était perturbative et consistait à dériver des modèles réduits et à les utiliser pour interpréter les trajectoires des électrons. Ces méthodes nous ont permis d'identifier l'impact de l'interaction coulombienne dans les phénomènes non linéaires observés expérimentalement et d'inclure son rôle dans la description des mécanismes sous-jacents.

Considérer la nature quantique des électrons avant l'ionisation et leur mouvement classique après l'ionisation, à la lumière de la dynamique non linéaire, nous ont permis de démêler les mécanismes à l'origine des phénomènes non linéaires observés expérimentalement, que la dynamique linéaire ne pouvait pas prédire. Par exemple, nous avons dévoilé le mécanisme à l'origine de la bifurcation observée dans les PMDs mesurées expérimentalement [93]. Nous avons déterminé, et avons pu décrire avec précision, comment les électrons peuvent retourner dans leur ion parent après plusieurs cycles laser et comment ils peuvent être piégés dans des états de Rydberg. Nous avons déterminé les conditions pour lesquelles des manifestations expérimentales de recollisions dans les champs CP et proches CP peuvent être observées. Ce travail de thèse démontre la complémentarité de la mécanique quantique et de la dynamique non linéaire pour comprendre et illustrer les mécanismes mis en jeux lorsque les atomes sont sujets à des impulsions laser intenses et polarisées elliptiquement.

Summary in English

Subjecting atoms or molecules to intense laser pulses gives rise to a variety of highly nonlinear phenomena, such as for instance the ionization of electrons and the radiation of high-frequency photons. The distributions of the velocity of the ionized electrons or the frequency of the radiated photons measured at the detector encode relevant informations on the target atoms and molecules at the natural time scale of the electrons, the attosecond-that is, million, million, millionths of a second. Understanding the dynamics of the ionized electrons and identifying the mechanisms of high-frequency radiation are essential steps toward interpreting and decoding the informations encrypted in the experimental measurements.

In this thesis, atoms subjected to intense and elliptically polarized laser fields in the infrared regime are theoretically studied. Despite their fundamental quantal nature in atoms, electrons display some classical behaviors when subjected to intense laser pulses. We exploit these classical features to understand and picture, with the help of trajectories, the physical mechanisms at play in order to interpret experimental measurements. We show the interdependent role of the quantum tunnel ionization of the electron and its subsequent classical motion for interpreting measurements in attosecond science.

After tunnel ionization of the electrons, the interplay between their interactions with the laser and their parent ion, by yielding their dynamics highly nonlinear, gives rise to rich and diverse ionization channels. Changing the ellipticity of the driving laser, which acts as a simple control knob in experiments, changes the prioritized ionization channel taken by the electrons. In this way, for instance, the electrons can probe different characteristics of the target atoms. The motion of the ionized electrons is analyzed using perturbative and nonperturbative techniques from nonlinear dynamics and Hamiltonian systems. This thesis work demonstrates the complementarity of quantum mechanics and nonlinear dynamics for understanding and illustrating the mechanisms involved when atoms are subjected to intense and elliptically polarized laser pulses.

Keywords

- Atomic, molecular and optical physics (AMO)
- Laser-matter interaction
- Hamiltonian systems
- Nonlinear dynamics
- Attosecond science
- Ultrafast phenomena
- Electron-electron correlation
- Above-threshold ionization (ATI)
- Nonsequential double ionization (NSDI)
- High harmonic generation (HHG)

Contents

\mathbf{P}	reface	2	vii
	Ackr	nowledgments/Remerciements	vii
	Résu	ımé en français	ix
		Introduction	ix
		Chapitre 1. Modèle Hamiltonien pour des atomes sujets à des impulsions laser intenses polar-	
		isées elliptiquement	xiv
		Chapitre 2. Dynamiques réduites pour des atomes sujets à des impulsions laser intenses po-	
		larisées elliptiquement: Le modèle du centre guide	XV
		Chapitre 3. Impact du potentiel de Coulomb en ATI: Les recollisions engendrées par l'interaction	
		coulombienne et la création d'états de Rydberg	XV
		Chapitre 4. Mécanisme de recollision à hautes ellipticités et leur importance en HHG et NSDI:	
		Rôle de l'enveloppe du champ laser	xvi
Chapitre 5. Structure de l'espace des phases de l'électron de recollision durant l'éta			
		Conclusions	xvii
	Sum	mary in English	xix
	Keyv	words	xix
т	4 a .l.		-
In	troa	uction	1 9
	State		ა ი
		New linear share and attosecond science	37
		Noninear phenomena	(
	T -1	Laser empticity and the Coulomb potential	0 10
	1 nes	SIS WORK	10
		I nesis objectives	10
		Methods	10
		Summary of the manuscript	11
1	Han	niltonian model for atoms subjected to intense and elliptically polarized laser pulses	13
	1.1	Hamiltonian model	13
		1.1.1 Orders of magnitude of an electron driven by intense laser fields	14
		1.1.2 Field-free single-active electron atom	15
		1.1.3 Intense laser field perturbation	17
	1.2	Description of the electronic dynamics in two steps	18
		1.2.1 Comparison between the quantum and the classical description of the dynamics	18
		1.2.2 Step (i): Ionization of the electron from the ground state	21
		1.2.3 Step (ii): Classical motion of the ionized electron	25
	1.3	Qualitative analysis of the T-trajectory in phase space	27
		1.3.1 Dynamics of the electron for $d = 1$: Poincaré section	27
		1.3.2 Dynamics of the electron in higher dimensions	28
	1.4	Conclusions	29
_			
2	Red	luced dynamics for atoms subjected to intense and elliptically polarized laser pulses	:
	'I'he	guiding center model	31
	2.1	The SFA and the CCSFA	33
		2.1.1 Strong field approximation (SFA)	33
		2.1.2 Coulomb-corrected strong field approximation (CCSFA)	34

	2.2	Derivation of the hierarchy of models for the guiding center (GC) $\ldots \ldots \ldots \ldots \ldots$	34
		2.2.1 Canonical Lie transforms	34
		2.2.2 Averaging the electron dynamics	36
	2.3	Analysis of the guiding-center (GC) models	41
		2.3.1 Comparison between Hamiltonian (1.14) and the reduced models	41
		2.3.2 Guiding-center (GC) phase-space dynamics	46
	2.4	Conclusions	49
3	The	e impact of the Coulomb potential in ATI: Coulomb-driven recollisions and Rydberg	5
	stat	te creations	51
	3.1	Photoelectron momentum distributions (PMDs)	52
		3.1.1 Short time-scale dynamics	53
		3.1.2 Long time-scale dynamics	56
	3.2	Coulomb-driven recollisions and Rydberg state creation	60
		3.2.1 Ionization time dependence	60
		3.2.2 Long plateau durations	65
		3.2.3 Rate of Rydberg state creation	67
	3.3	The shape of the rescattering domain and its experimental implications	68
		3.3.1 Analysis of the shape of the rescattering domain	68
		3.3.2 Implication of the shape of the rescattering domain	71
	3.4	Conclusions	77
4	Rec	collision mechanism at high ellipticities and its significance in HHG and NSDI: Role	;
	of t	he laser envelope	79
	4.1	Scenario of envelope-driven recollisions	81
		4.1.1 Field-free atom in the rotating frame (RF)	82
		4.1.2 Over-the-barrier ionization: Role of the target species and the laser wavelength	82
		4.1.3 Return energy of the recolliding electron	88
	4.2	Applications to HHG	91
		4.2.1 Production of HHG with high EP laser fields	91
		4.2.2 Time-frequency analysis: Dominance of the envelope-driven recollisions for highly el-	
		liptic laser polarizations	93
		4.2.3 Influence of the duration of the ramp-up: Control of the HHG cutoff	94
	4.3	Applications to NSDI	95
		4.3.1 Double ionization probability curves	95
		4.3.2 Ionization and return times of the recolliding electron	98
		4.3.3 Breakdown of NSDI as a function of the laser frequency and the target species	99
	4.4	Conclusions	99
5	Str	ucture of the phase space of the recolliding electron in step (ii)	101
	5.1	Linearly polarized fields for $d = 1$	103
		5.1.1 One-dimensional recolliding periodic orbits (RPOs): \mathcal{O}_A	104
		5.1.2 Saddle periodic orbits (SPOs): \mathcal{O}_S	106
		5.1.3 Invariant manifolds structuring the phase space of the recolliding electron	107
	5.2	Linearly polarized fields in higher dimensions	108
		5.2.1 Two-dimensional recolliding periodic orbits (RPOs): \mathcal{O}_F	109
		5.2.2 Recollisions in higher dimensions: A family of invariant tori drives the electron dynam- ics	110
	53	From linearly to circularly polarized fields: Consequences of the symmetry brooking on the	110
	0.0	periodic orbits	112
		5.3.1 Symmetries in phase space	112
		5.3.2 Evolution of recolliding periodic orbits (RPOs) with respect to the lasor ellipticity	11/
		5.3.3 Connection between \mathcal{O}_F and a fixed point in the rotating frame (RF): Case of circularly	114
	E 4	Conclusions	115
	0.4		11(

Conclusions Summary Perspectives	119 119 121			
A Glossary A.1 Acronyms A.2 Notations A.2.1 Numerator layout convention A.2.2 Laser A.2.3 Atomic model A.2.4 Dynamical systems and Hamiltonian formalism	123 123 124 124 125 125			
B Atomic scales B.1 Hydrogen atom: The Bohr semi-classical approach B.2 Atomic units B.2.1 Laser parameters	127 127 128 128			
C Dynamical systems C.1 Continuous flow C.1.1 Invariant objects C.1.2 T-periodic change of coordinates C.2 Hamiltonian systems C.2.1 Change of coordinates C.2.2 Canonical Poisson bracket	131 131 133 134 135 136			
 D Computational methods D.1 Statistical analysis	139 139 141 142 142 143 146 148			
Bibliography				
List of publications 1				

Introduction

Imaging atomic and molecular orbitals [115, 59, 135], tracking the motion of the electrons in real time [25, 64, 126] and measuring their correlations in atoms, molecules and solids [101, 72, 110, 141, 18] have been the focus of *attosecond science* with the long term goal of making movies of structural dynamics in chemical reactions occuring in nature [41]. These snapshots would reveal underlying mechanisms at the temporal and spatial scales of the electrons (subfemtosecond 1 fs = 10^{-15} s and subnanometer 1 nm = 10^{-9} m, respectively, see Fig. 1), the binder of matter, which would allow to gain a deeper understanding and control of these ultrafast nonequilibrium processes. While, nowadays, imaging matter at the temporal and spatial scales of atoms (1 ps = 10^{-12} s and 1 Å = 10^{-10} m, respectively, see Fig. 1) is routinely achieved in laboratories. Zoom in these images, on subfemtosecond time scales and subnanometer spatial scales, keeps challenging scientists in both the technical and theoretical aspects since about thirty years.

For instance, images of atomic structures in solid surfaces, at the spatial scale of atoms, can be obtained by moving a metallic tip of a few angstroms wide above the targeting surface. This is the basis of scanning tunneling microscopy (STM). The first images are observed in 1982 [23], and lead to a new branch of microscopy: the scanning probe microscopy (SPM). Despite the success of STM for locating atoms in solid surfaces at equilibrium, ultrafast nonequilibrium processes inside matter are not tractable by the metallic tip of the STM. This illustrates clearly the difficulties of performing real-time measurements of the motion of atoms and electrons. Roughly speaking, as it is illustrated in Fig. 2, devices probing matter in a time scale much faster than the natural time scale of atoms and electrons can exclusively capture clear snapshots of their configurations. The metallic tip should move on subfemtosecond time scales to capture in real time the



Figure 1: Comparison of the characteristic spatial and temporal scales of electrons and atoms in microscopic structures reproduced from Ref. [91]. In comparison, the duration of the shortest laser pulse generated (June 18, 2019) is 43 as $= 4.3 \times 10^{-17}$ s.

motion of the electrons, which is not feasible.

Meanwhile, femtochemistry were at its early stage. The wisdom of femtochemistry is to use short infrared (IR) laser pulses, of the order of the femtosecond, as fast photographers of atoms in molecular reactions, a breakthrough for ultrafast imaging technology. In 1987, when laser pulses became short enough to obtain clear snapshots (few femtoseconds duration), the first real-time measurements of the motion of atoms during chemical reactions are performed using pump-probe spectroscopy [174]. Nonetheless, electrons in atoms and molecules move on subfemtosecond time scales, more than a thousand times faster than the motion of atoms in molecular reactions, the probe laser pulses must be thousand times shorter than the ones employed in femtochemistry, i.e., they must be attosecond-duration light sources (1 as $= 10^{-18}$ s). In the late nineties started the rush of ultrashort laser pulses: a theoretical and technological endeavor.

Free-electron lasers [69] (FELs) are one type of sources of ultrashort laser pulses which necessitate veryhigh-energy resources and large-scale infrastructures. The other type of sources of ultrashort laser pulses provides table-top ultrashort light sources, which can easily be used in laboratories. The first steps which led



Figure 2: This illustrates the difficulty of capturing clear images of electrons in atoms and molecules at their natural time scale. These are two photographs taken from Jackson bridge in Atlanta using two different shutter speed of the camera (displayed in the upper left corner of each panel). The typical time scale of cars on the highway is 0.2 s (the typical speed of the cars on the highway is 100 km \cdot h⁻¹ and their typical spatial scale is 5 m). In the upper panel, the shutter speed is twice smaller than their typical time scale, and their representation is clear. In contrast, in the lower panel, the shutter speed is roughly ten times faster than their typical time scale, and we observe luminous trails. A clear image of the cars is obtained only if the shutter speed of the camera is faster than their natural time scale. In molecular imaging, ultrashort lasers probe electron dynamics in atoms and molecules faster than the natural time scale of the electrons (i.e., subfemtosecond in molecules) and can capture clear images of their dynamics [59, 115, 64, 25, 135, 126].

to the production of table-top ultrashort laser pulses are discovered in 1987, when high harmonic generation (HHG) has been observed for the first time by propagating femtosecond IR laser pulses through a gas of atoms [168]. When an IR laser pulse propagates through a medium of atoms or molecules, the target medium radiates photons in the extreme ultraviolet (XUV) regime. The HHG intensity spectrum corresponds to the distribution of the frequencies (or harmonics) of the radiated photons. Typical HHG intensity spectra for an atomic gas are shown in Fig. 3 for different laser intensities. For isotropic gases, such as a gases of atoms, the frequency of the radiated photons is an odd multiple of the frequency of the driving laser [102]. The typical HHG intensity spectrum is composed of three distinct regions. Regardless the laser intensity, harmonic intensities exponentially decrease until the fifth harmonic order, this is referred to as the below threshold harmonic (BTH) region. For larger harmonics, the plateau region, at the *harmonic cutoff*, after which the harmonic intensities drop off rather suddenly. Then, the harmonic intensities are extremely small. In Fig. 3, we observe that the harmonic cutoff increases for increasing intensities of the driving laser.

The first observations of the plateau region in HHG intensity spectra [54] were very promising for the production of ultrashort laser pulses. Indeed, a frequency comb can be obtained after isolating the plateau region by filtering the HHG intensity spectrum. The resulting post-processed beam of photons is a train of ultrashort XUV laser pulses. The larger the harmonic cutoff and the shorter the laser pulse. In 2001, a single attosecond pulse [70] and a train of attosecond pulses [132] are observed for the first time in experiments. Nowadays, table-top ultrashort light sources [153, 35] are used routinely in laboratories. Ultrashort XUV laser pulses have opened up access to attosecond-time resolved measurements of electron dynamics [56, 67, 64, 155, 87, 135].

State of the art

The productivity and the technologies in attosecond science have improved significantly from 1993, after the discovery of the mechanism underlying the plateau region in HHG intensity spectra: The *recollisions* [92, 39, 154]. A recollision occurs when an ionized electron returns to its parent ion. Remarkably, while HHG is produced by propagating an IR laser pulse through atomic or molecular gases, the key mechanism can be understood from the response of a single atom with a single-active electron [100, 146]. In addition to HHG, recollisions give rise to a variety of nonlinear phenomena, and are now considered as the keystone of attosecond science. In Fig. 4, the red curves represent the rate of publications (left panel) and the rate of citations (right panel) from the Web of Science database with "recollision" as a topic. We observe that, since then, the interest triggered by the recollisions has kept increasing.

Recollisions: keystone of attosecond science

The physics of HHG is built on the three-step scenario of the recollisions [39, 154], which are illustrated in Fig. 5. We consider a single-active electron (SAE) atom driven by linearly polarized (LP) fields $\mathbf{E}(t) = \mathbf{e}_x E_0 \cos(\omega t)$. The laser field oscillates in time at a frequency ω along the polarization axis \mathbf{e}_x . If the amplitude of the laser E_0 is large enough, an electron can be teared off the core by the laser: this is step (i) of the recollision scenario. During step (i), an electron wave packet tunnel ionizes through the potential barrier induced by the laser field.

In step (ii) of the recollision scenario [39, 154], the motion of the electron is treated classically. The electron is initiated on the potential barrier in the direction opposite of that of the laser field. Its motion is along the polarization axis. Using the fundamental principle of dynamics $\ddot{x} = -E_0 \cos(\omega t)$, the position of the electron as a function of time is given by

$$x(t) = x_0 + \frac{E_0}{\omega^2} \left[\sin(\omega t_0) \omega(t - t_0) + \cos(\omega t) - \cos(\omega t_0) \right],$$
(1)

where $x(t_0) = x_0$ and $\dot{x}(t_0) = 0$ are the position and velocity of the electron, respectively, initiated on the potential barrier at ionization time t_0 . Atomic units are used throughout the manuscript unless stated otherwise. We observe that the motion of the electron is composed of mainly two components: a drift in the term $\sin(\omega t_0)\omega t$, and oscillations in the term $\cos(\omega t)$. The electron ionizes relatively close to the core, at $x_0 \sim 1$ a.u. In contrast, the order of magnitude of the amplitude of the oscillations of the electron in the laser is $E_0/\omega^2 \sim 10$ a.u. for the laser parameters used in Fig. 3. During the excursion of the electron outside the parent ion, after the laser field has reached its peak amplitude, it pushes the electron back towards the core, and the electron collides with the parent ion: this is a recollision.



Figure 3: Number of radiated photons (HHG intensity spectrum) in Xe (ionization potential $I_p = 12.13 \text{ eV}$) as a function of the harmonic of the laser field Ω/ω measured in experiments reproduced from Ref. [102], where Ω is the frequency of the radiated photon and ω is the frequency of the driving field. The intensities are (\circ) $3 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, (\bullet) $1.3 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, (\Box) $9 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}$, (\blacksquare) $7 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}$, and (Δ) $5 \times 10^{12} \text{ W} \cdot \text{cm}^{-2}$. The wavelength is $\lambda = 1064 \text{ nm}$. The typical HHG intensity spectrum is composed of three distinct regions: harmonic intensities exponentially decrease until the fifth harmonic order, this is referred to as the below threshold harmonic (BTH) region. For larger harmonics, the harmonic intensities are rather constant, this is referred to as the plateau region. An abrupt cutoff follows the plateau region, at the harmonic cutoff, after which the harmonic intensities drop off rather suddenly.



Figure 4: Number of (left panel) publications and (right panel) citations statistics from the Web of Science database (May 9, 2019). Red, blue and green curves are "recollision", "Elliptically polarized laser pulses" and "Coulomb in intense laser pulses" as a topic, respectively.



Figure 5: Classical schematic of the three-step model and HHG reproduced from Ref. [142]. The color code is the effective potential of the electron in interaction with the ion and the laser field. The thick red curve shows the laser field as a function of time.

In step (iii), the electron interferes and recombines with the portion of the wave function in the ground state which remained bounded. The recombination time of the recolliding electron, denoted $t_0 + \Delta t$, is such that $x(t_0 + \Delta t) \approx 0$, with Δt the excursion time. At the recombination, a photon of frequency

$$\Omega = I_p + 2U_p \left[\sin(\omega t_0 + \omega \Delta t) - \sin(\omega t_0) \right]^2, \qquad (2)$$

is emitted, where I_p is the ionization potential of the atom. The right-hand side of Eq. (2) is the energy of the electron at the recollision as a function of the ionization time t_0 , the recombination time $t_0 + \Delta t$ and the ponderomotive energy $U_p = E_0^2/4\omega^2$.

In the HHG intensity spectrum, the BTH is built of photons emitted at a frequency $\Omega < I_p$. The plateau region is built of photons radiated by recollision processes [39, 100, 146, 166, 20]. In Eq. (2), we observe that the frequency of the radiated photons depends on the times when the recolliding electrons ionize and recombine. We assume that $x_0 \approx x(t_0 + \Delta t) \approx 0$. Using Eq. (1), it implies $\sin(\omega t_0) = [\cos(\omega t_0) - \cos(\omega t_0 + \omega \Delta t)]/\omega \Delta t$. This condition fixes the excursion time of the recolliding electrons as a function of their ionization time. We substitute this condition in Eq. (2). We maximize the frequency of the radiated photons with respect to the ionization time of the recolliding electrons t_0 and their excursion time Δt , and we obtain the maximum frequency of photon radiated by recollision processes [39, 100]

$$\Omega_c \approx I_p + 3.17 \mathrm{U}_p,\tag{3}$$

for $\omega \Delta t \approx 4.09$ and $\omega t_0 \approx 0.31$. As a consequence, the trajectories which bring back the largest amount of energy to the core, around 3.17U_p, ionize early after the laser reaches its peak amplitude, and recollide around half of a laser cycle later. For a laser wavelength $\lambda = 1064$ nm and laser intensities $I = 3 \times 10^{13}$ W · cm⁻², $I = 1.3 \times 10^{13}$ W · cm⁻², $I = 9 \times 10^{12}$ W · cm⁻², $I = 7 \times 10^{12}$ W · cm⁻², $I = 5 \times 10^{12}$ W · cm⁻² (laser intensities used in Fig. 3), the harmonic cutoffs predicted by Eq. (3) are (\circ) $\Omega_c / \omega \approx 19$, (\bullet) $\Omega_c / \omega \approx 14$, (\Box) $\Omega_c / \omega \approx 13$, (\blacksquare) $\Omega_c / \omega \approx 12$ and (Δ) $\Omega_c / \omega \approx 12$, respectively. The cutoff law predicted by Eq. (3) agrees well with the experimental measurements in Fig. 3, in particular for $I > 7 \times 10^{12}$ W · cm⁻². The recollision mechanism and the physics beyond HHG are summarized and schematically represented in Fig. 6. Figure 6a shows the experimental setup, in which an intense IR laser pulse is sent on a gas of atoms. Figure 6b schematically represents the electron motion in the single-atom response and the laser field. Figure 6c represents the harmonic number corresponding to the return energy of the recolliding electrons as a function of their ionization times (upper panel) and a HHG intensity spectrum as a function of the harmonic number of the radiated photons (lower panel). An energy of return of the recolliding electrons is associated to each frequency of the radiated photons in the plateau region.

In sum, classical mechanics has been able to unravel some of the physics beyond HHG in atoms subjected to intense laser pulses. In particular, the scaling law of the harmonic cutoff predicted by the recollision model (3) agrees well with the experimental measurements [102]. The larger the harmonic cutoff and the shorter the post-processed XUV laser pulses. To increase the harmonic cutoff significantly, the amplitude of the laser must be increased or its frequency must be decreased. Nonetheless, the intensity and the frequency



Figure 6: HHG experiments and measurements for the production of attosecond pulse trains reproduced from Ref. [35]. (a) Schematic of the experiments. (b) Three-step model [39, 154] and illustration of short and long trajectories. (c) Return time as a function of the emitted harmonic number of the recolliding electron in the SFA (upper panel). Harmonic power spectrum as a function of the harmonic number (lower panel).



Figure 7: Left panel: Cartoon of an atom subjected to an intense infrared (IR) laser pulse. The red curve is the trajectory of a recolliding electron. Right panels: Cartoon of the recolliding electron which (upper panel) recombines with the ground state and produce HHG, (middle panel) rescatters with the core and induces ATI, (lower panel) exchange energy with a bound electron and induces NSDI.

of the lasers are limited by the available technological devices. Increasing the frequency cutoff is not a straithforward task, and is the focus of many studies (see Refs. [7, 44], e.g.).

The success of the recollision scenario for describing the production of HHG demonstrates the existence of classical features in these fundamentally quantum systems. Since it is one of the places where classical and quantum mechanics meet, another important role assessed by attosecond science is to test and challenge concepts, methods and interpretations arising from this mixture of classical and quantum mechanics. For instance, the "time the electron spends under the potential barrier" to ionize during step (i), refered to as "tunneling time" in Refs. [95, 94], remains under debate [30, 155, 10, 134, 95, 94, 124, 60, 152, 149]. In this thesis, we exploit the classical features of the electron dynamics after ionization in order to understand and picture, with the help of trajectories, the nonlinear phenomena observed in atoms subjected to intense and elliptically polarized laser pulses.

Nonlinear phenomena

By looking at the success for the prediction of the harmonic cutoff by the three-step model, which are obtained by ignoring the interaction between the electron and its parent ion, we could think this latter plays a minor role in the mechanism behind the production of ultrashort light sources. It is yet dominant when the electron returns to its parent ion. The electron, by experiencing strong interactions with the core, probes its internal structures and bound electron dynamics. If the electron radiates photons, these informations are found in the HHG intensity spectrum. High harmonic spectroscopy are techniques to extract these informations from the HHG spectrum. For instance, high harmonic spectroscopy is used to track the charge migration in molecules [90].

A first obvious effect of the ion-electron interaction inside atoms is the deflection of the recolliding electron

at the recollision (see middle panel of Fig. 7). The combined strong laser and ion-electron interactions can produce ionized electrons with high energies [39, 133]. This is above-threshold ionization (ATI). The deflection of the recolliding electrons by the ionic core is referred to as the *rescattering* of the electron. During the rescattering, the electrons probe the internal structure and dynamics of target ions and molecules [99]. The distributions of the ionized electrons at the detector, referred to as photoelectron momentum distributions (PMDs), encode these informations [115, 76]. This is the basis for imaging techniques, e.g., laser-induced electron diffraction [176, 115, 139] (LIED) for molecular imaging [25] and photoelectron holography [76, 22]. These techniques are often seen as "half-scattering" experiments, in analogy to the Rutherford "scattering" experiments [151], where particles are sent from infinity to probe target atoms. Here, the particles probing the targets come from the targets themselves.

Another obvious effect of the ion-electron interactions is to bound electrons. In particular, ion-electron interactions can bound electrons during the excursions of recolliding electrons. When a recolliding electron returns to the core, it exchanges energy with a bound electron and both can ionize [39, 110] (see lower panel of Fig. 7). This is non sequential double ionization (NSDI). NSDI is one of the most dramatic manifestations of electron-electron correlation in nature [17]. PMDs of doubly ionized atoms encode informations on the electron-electron correlations [141, 18].

In sum, recollisions, because they involve combinations between the strong laser and ion-electron interactions, give rise to a variety of highly nonlinear phenomena [39, 16, 41, 5, 91, 17] summarized in Fig. 7. Imaging techniques based on these highly nonlinear phenomena offers very promising outlook. However, because of their inherent technicality and the fundamental difficulty to assess the motion of the electrons in the combined laser and ion-electron interactions, are not routine procedures employed in laboratories for the real-time measurement of the motion of electrons in matter yet. In particular, movies of electron dynamics in large polyatomic biological molecules have never been observed. Attosecond science is still rather at an early stage.

Laser ellipticity and the Coulomb potential

The strength of the most fundamental ion-electron interaction is quantified by the Coulomb potential. While the role of the Coulomb potential is obvious in the rescattering processes (in ATI) and for trapping electrons close to their parent ion (in NSDI), this is not the case for its manifestations in the dynamics of the ionized electrons. The ellipticity of the laser field, which acts as a simple control knob in experiments, highlights different phenomena originating from electron dynamics [9, 63, 156, 96, 75, 47, 68, 36, 43, 114]. Figure 4 shows the rate of citations and publications with "Elliptically polarized laser pulses" and "Coulomb in intense laser pulses" as a topic. Since 1990, the interest on these topics has drastically increased. In particular, since it was noticed that changing the ellipticity of the driving laser highlights the impact of the Coulomb potential on the motion of the ionized electrons, referred to as *Coulomb effects* [28, 11, 62, 38, 93]. Put slightly differently, by changing the laser ellipticity, it was noticed that the Coulomb potential is significant, even after ionization.

Figure 8 shows PMDs and photoelectron angular distributions (PADs) of He atoms for different laser ellipticities. PMDs are distributions of the momentum of the ionized electrons at the detector, and PADs are distributions of the angle of the momentum of the ionized electrons with respect to the major polarization axis. The laser field is of the form $\mathbf{E}(t) = E_0 f(t) [\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \xi \sin(\omega t)] / \sqrt{\xi^2 + 1}$, where \mathbf{e}_x and \mathbf{e}_y are the major and minor polarization axes, respectively. The laser ellipticity is ξ and the laser envelope is f. The duration of the laser is rather long, such that the laser envelope is $f \approx 1$ for several laser cycles. In Fig. 8, we observe that as laser ellipticity is varied, the shape of the PMDs undergoes drastic changes.

In Fig. 8, for $\xi = 0$ (LP fields), the PMD is one lobe centered around the origin of the momentum space. In the PAD, we observe that this cloud is slightly asymmetric with respect to the minor polarization axis. At $\xi = 0.15$, we observe that the PMDs splits into two lobes symmetric with respect to the origin of the momentum space. For increasing ellipticity, the two lobes go further away from each other. In order to understand the changes in the PMDs, the natural theoretical framework is to apply the same procedure as used to describe the recollision scenario, namely: (i) the electron tunnel-ionizes, (ii) the motion of the electron is treated classically without the Coulomb potential. The electron ionizes outside the potential barrier induced by the laser field, and as a consequence, its initial momentum is $\mathbf{p}_0 = \mathbf{0}$. From the fundamental principle of dynamics, $\dot{\mathbf{p}} = -\mathbf{E}(t)$, the momentum of the electron is given by $\mathbf{p} = \mathbf{A}(t) - \mathbf{A}(t_0)$, where t_0 is the ionization time. The vector potential is $\mathbf{A}(t)$ such that $\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$. Asymptotically, at the detector, the vector potential of the laser field is turned off, and therefore $\mathbf{p}(t) = -\mathbf{A}(t_0)$ when time goes to infinity. The most probable ionization time of the electron is such that the amplitude of the laser field is the largest,



Figure 8: Experimental measurements of He for $I = 8 \times 10^{14}$ W \cdot cm⁻² and $\lambda = 780$ nm. Upper panels: Photoelectron momentum distribution (PMDs) (colored figures) and photoelectron angular momentum (PADs) (white figures) reproduced from Ref. [140]. For $\xi \leq 0.2$, the PMDs are mainly one lobe centered at the origin. For $\xi \geq 0.2$, the PMDs are mainly two lobes symmetric with respect to the origin. The offset angle is indicated by $\Delta\theta$ for ellipticity $\xi = 0.4$. The offset angle is the angle between the minor polarization axis \mathbf{e}_y and the maximum of the upper lobe in the PMDs. Lower panel: Maximum of the upper lobe of the PMDs as a function of the laser ellipticity ξ reproduced from Ref. [93]. The green and horizontal black curves are the prediction of the SFA given by Eq. (4) along the minor (\mathbf{e}_y) and the major (\mathbf{e}_x) polarization axes, respectively.

i.e., $\omega t_0 = k\pi$ with $k \in \mathbb{Z}$. In absence of Coulomb potential, the most probable momenta of the electron at the detector are

$$\mathbf{P} = \pm \frac{\xi E_0}{\omega \sqrt{\xi^2 + 1}} \mathbf{e}_y. \tag{4}$$

At $\xi = 0$ (LP fields), the most probable momentum of the electron at the detector is zero, in agreement with the experimental measurements of Fig. 8. For increasing ellipticities, the most probable momentum of the electron at the detector goes further away from the origin along the minor polarization axis, in qualitative agreement with experimental measurements in the upper panels of Fig. 8. For increasing ellipticity, the drift energy of the electron increases, and pushes it away from the core. The electron ionizes without recolliding.

In order to assess these qualitative changes, in Ref. [93], the location of the peaks of the PMDs is followed as a function of the laser ellipticity (lower panel of Fig. 8), denoted $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$. In the lower panel of Fig. 8, the blue and red circles are the momentum coordinates of the maximum of the upper lobe in the PMDs, P_x and P_y , respectively. The solid green curve is the positive component of \mathbf{P} given by Eq. (4) along the minor polarization axis \mathbf{e}_y . The solid black curve is the component of \mathbf{P} given by Eq. (4) along the major polarization axis \mathbf{e}_x . For $\xi \leq 0.1$, we observe that $\mathbf{P} \approx \mathbf{0}$, and as a consequence, the PMD is mainly one lobe centered at the origin. In contrast, the solid green curve increases linearly in this range of ellipticities. The persistence of the single lobe in the PMDs, revealed by the lower panel of Fig. 8, is referred to as *Coulomb focusing* [28, 38]. For $\xi \geq 0.1$, we observe that when P_y is positive, P_x is negative. The red circles agree well with the green curve. However, there is a discrepancy between the horizontal black curve and the blue circles. Indeed, P_x is negative and non-zero. In the PMDs, we observe an asymmetry with respect to the minor polarization axis. This asymmetry, revealed by the lower panel of Fig. 8, is referred to as *Coulomb asymmetry* [11, 62]. In the experimental measurements reported in Ref. [104], the final momentum of the electron exhibits the same features as in experiments [140, 93] (see Fig. 3.13). The hypothesis made in Ref. [93] is that there is a *bifurcation* when varying the ellipticity of the laser field.

Therefore, changing ellipticity reveals effects of the Coulomb potential and changes the prioritized ionization channel taken by the electrons. In this way, for instance, the electrons can probe different characteristics of the target atoms. However, there have always been fundamental obstacles for understanding and assessing the impact of the Coulomb potential on the trajectories of the ionized electrons. In particular, the interplay between the interactions of the ionized electrons with the laser and their parent ion gives rise to multiple temporal and spatial scales, yielding their dynamics highly nonlinear.

Thesis work

In this thesis, we consider single active electron atoms (except in Sec. 4.3 where an extension of the study for double active electron atoms is performed) driven by intense and elliptically polarized laser pulses. The dynamics of the electron is studied in the quantum and classical frameworks. In the classical framework, tools from nonlinear dynamics and Hamiltonian formalism are used to investigate the motion of the electron.

Thesis objectives

The overall objective of this thesis is to understand the changes in the dynamics of electrons in atoms driven by intense laser pulses for changing ellipticities, and their impact in the highly nonlinear phenomena observed in experiments. One goal is to describe the motion of electrons in the classical framework, to exploit tools from nonlinear dynamics, while taking into account the quantal nature of the electrons in atoms. In the meantime, another goal is to assess the role of the Coulomb potential in the dynamics of the ionized electrons and in the excursion of the recolliding electrons. This consists in identifying manifestations of the Coulomb potential in experimental or numerical measurements and to determine the underlying mechanisms in terms of electron trajectories. To achieve that, one targeting is to disentangle the contribution of the laser and the Coulomb fields, in order to identify and describe the role of the Coulomb potential in nonlinear phenomena observed in attosecond science.

Methods

In this thesis manuscript, we detail the numerical and analytical investigations which have been carried out. In this manuscript, we show the:

- Derivation of a hierarchy of reduced models for describing the dynamics of the electron in an intense laser field in terms of the dynamics of its guiding center.
- Computation of high-dimensional invariant structures in phase space (invariant manifolds of a family of one-dimensional invariant tori).
- Representation of high-dimensional invariant structures in two dimensions.
- Computation of classical trajectory Monte Carlo simulations (CTMCs).
- Results of high resolution statistics using graphics processing unit (GPU) calculations.

Summary of the manuscript

Chapter 1. Hamiltonian model for atoms subjected to intense and elliptically polarized laser pulses In this chapter, we derive the Hamiltonian models for single-active electron atoms driven by intense and elliptically polarized laser pulses in the quantum and classical frameworks. By comparing the Wigner quasi-probability distribution [167] of the electron and its classical distribution in phase space, we show that the quantum dynamics of the electron exhibits clear classical features after ionization. We describe a two-step model in which the electron (i) tunnel-ionizes through the potential barrier induced by the laser field, and (ii) moves classically outside the ionic core region. This two-step model allows us to exploit tools from nonlinear dynamics we use throughout the manuscript for the electrons in atoms. We introduce tools from nonlinear dynamics we use throughout the manuscript for the description of the electron dynamics in step (ii). We introduce the questions and themes which are studied in the next chapters.

Chapter 2. Reduced dynamics for atoms subjected to intense and elliptically polarized laser pulses: The guiding center model In this chapter, we derive reduced models for atoms subjected to intense and elliptically polarized laser pulses, namely: The strong field approximation (SFA), the Coulomb-perturbed strong field approximation (CCSFA) and the guiding-center (GC) models. The SFA neglects the Coulomb potential, and the CCSFA treats it as a perturbation of the laser interaction. Concerning the GC models, we derive a hierarchy of models for the dynamics of the GC. The GC motion corresponds to the averaged motion of the electron in the combined laser and Coulomb fields. The hierarchy of models is derived using the canonical Lie transforms and takes into account the Coulomb potential. The GC models are particularly efficient to approximate the electron trajectories on long time scales. As a result of averaging, the GC Hamiltonians are conserved in time, and as a consequence, one can define an energy for the electrons with the energy of their associated GC. We study the accuracy of each model as a function of the position of the electron in phase space and the integration time.

Chapter 3. The impact of the Coulomb potential in ATI: Coulomb-driven recollisions and Rydberg state creations In this chapter, we investigate the role of the Coulomb potential in step (ii) of the ionization process using four models, namely: The reference Hamiltonian, the SFA, the CCSFA and the second order GC model. The SFA is used to target where are the contributions of the Coulomb potential, such as it is done with Eq. (4) and Fig. 8. The CCSFA and the second order GC model are used to decouple the contributions of the laser and the Coulomb potential. We show that the CCSFA and the GC models are complementary, in the sense that the CCSFA is effective on short time scale dynamics while the GC model is effective to describe the Rydberg state creation and the Coulomb-driven recollisions. Finally, we use the GC model to unravel the mechanism behind the bifurcation observed in the PMDs (see lower panel of Fig. 8). The bifurcation arises from the complementarity between the quantum tunnel ionization of the electron and its subsequent classical nonlinear dynamics.

Chapter 4. Recollision mechanism at high ellipticities and its significance in HHG and NSDI: Role of the laser envelope In this chapter, we identify a highly probable recollision channel of electrons with large return energy by taking into account the effects of the pulse envelope f(t). The competition between the Coulomb force and the laser field creates a channel of ionization early after the laser field is turned on. Just as the electron is outside the core region, the amplitude of the vector potential is small, and therefore the sideways drift of the electron can be compensated by its momentum. We show that this recollision channel can be used to produce HHG with atoms driven by high elliptically polarized laser fields [near circularly polarized
(CP) fields], and is also responsible for the enhanced double ionization from specific target species subjected to CP fields observed in experiments [61, 111]. The existence condition of this recollision channel agrees well with the conditions for which the enhanced double ionization for CP fields is observed experimentally [55, 65, 61]. In addition, we notice that, under reasonable conditions, these recollisions can take place in the Attoclock setup, where recollisions are always assumed to be nonexistent [53, 152].

Chapter 5. Structure of the phase space of the recolliding electron in step (ii) In this chapter, we investigate the phase space of the electron in step (ii) using nonperturbative methods. The Coulomb potential and the laser interactions are fully taken into account. The numerical computations are detailed in Appendix D. We study the global behavior of the trajectories through the analysis of invariant structures for d = 1, 2, 3, where d is the dimension of the configuration space. In particular, we show that a set of invariant manifolds of a family of invariant tori associated with a periodic orbit which originates from the competition between the strong Coulomb and laser interactions, drive the electron dynamics in phase space. We show that RPOs, a specific family of periodic orbits introduced in Refs. [81, 80], are located in the vicinity of this high-dimensional invariant structure. We study the symmetries of the equations of motion of the electron for varying ellipticities, and how they impact the invariant objects. We show that key invariant structures which drive the recollisions exist regardless of the ellipticity.

Chapter 1

Hamiltonian model for atoms subjected to intense and elliptically polarized laser pulses

The goal of this manuscript is to analyze and interpret the dynamics of electrons for atoms subjected to intense and elliptically polarized laser pulses. In this chapter, we identify the characteristic temporal and spatial scales of the electron in intense and ultrashort laser pulses. The study of these temporal and spatial scales allows us to formulate hypotheses on the system, and to make approximations such as the single-active electron (SAE) approximation, the static ion approximation and the dipole approximation. As a consequence, we derive the quantum and classical models we use throughout the manuscript. In experimental measurements, some features are solely due to quantum effects. For instance, the patterns of interference observed in the PMDs [76], the plateau in the HHG spectrum [166, 20], or the peaks in the ATI spectrum due to multiphoton absorption [6, 57]. However, the dynamics of the electron also exhibits classical features [16]. Classical mechanics has been very successful in attosecond science. For instance, for interpreting and predicting the cutoff of the HHG spectrum [39] and its time-frequency profiles.

Questions

- What are the base models for electron dynamics for atoms driven by intense and elliptically polarized laser pulses ?
- Under which conditions does the quantum dynamics of the electron exhibits classical features ?

Plan In Sec. 1.1, we introduce the quantum model for the description of the dynamics of the atom subjected to intense and ultrashort laser pulses. In Sec. 1.2, we study the similarities between the classical and quantum dynamics of the electrons, and we determine the conditions under which the quantum model exhibits classical features. We show that, by initiating appropriately the trajectories of the electrons, they reproduce classical patterns in the quantum wave function. We exploit the classical features of the quantum wave function by describing the dynamics of the electron in two steps where: (i) the ionization of the electron is treated quantum mechanically, (ii) the dynamics of the electron after ionization is treated classically. In step (i), the ionization is described by the tunnel-ionization process [83, 8, 137, 138, 136], and provides the distribution of the initial conditions of the electrons in phase space. Step (ii) is used to understand and analyze the motion of the electrons and the mechanisms behind the phenomena measured in experiments in the light of nonlinear dynamics. We define the T-trajectory, which corresponds to the most probable trajectory according to step (i). In Sec. 1.3, we describe the qualitative behavior of the T-trajectory during step (ii). Through the analysis of the motion of the T-trajectory, we introduce the tools from nonlinear dynamics used throughout the manuscript.

1.1 Hamiltonian model

In this section we define the Hamiltonian model of a single-active electron atom driven by intense and ultrashort laser pulses. We study the spatial and temporal scales of the electron in the laser fields used in the manuscript. First, we study the field-free atom case, and we introduce the potentials describing the ion-electron interaction used throughout the manuscript. Second, we consider the atom driven by the intense laser field, and introduce the shape of the laser pulses we use throughout the manuscript. The system is described in the *laboratory frame* for a configuration space of dimension d = 1, 2, 3.



1.1.1 Orders of magnitude of an electron driven by intense laser fields

Figure 1.1: Map of the wavelength-intensity parameter space in strong-field ionization. The purple dotted region is where the dipole approximation is valid (dipole oasis). The black line is the short-wavelength dipole limit [107] where $\lambda < 1$ a.u. The magenta line is the long-wavelength dipole limit where $E_0/\omega^2 > \lambda/100$. Above the red dotted line is where the radiation pressure is non negligible [107], $U_p^2/2c^2 > 0.5$ a.u.. Above the blue dashed line is where the relativistic effects are non negligible, $2U_p/c^2 > 1$ a.u. The vertical green line indicates $\lambda = 780$ nm. The interval indicates where $\lambda = 780$ nm and $I \in [10^{12}, 10^{16}]$ W \cdot cm⁻².

We consider an atom subjected to a very intense infrared laser field. The intensity of the laser I is related to its amplitude E_0 with E_0 [a.u.] = $5.338 \times 10^{-9} (I \ [W \cdot cm^{-2}])^{1/2}$ (see Appendix B for details on atomic units and conversions). Here, we focus on an intensity range

$$I \sim 10^{12} - 10^{16} \,\mathrm{W} \cdot \mathrm{cm}^{-2},$$

i.e., $E_0 \sim 5 \times 10^{-3} - 0.5$ a.u. The wavelength of the laser λ is related to its frequency ω by ω [a.u.] = $4.555 \times 10^{-8}/\lambda$ [m]. We consider infrared laser pulses generated from a Ti:Sapphire [121] of wavelength $\lambda = 780$ nm. This corresponds to $\lambda \approx 1.5 \times 10^4$ a.u. and frequency $\omega = 0.0584$ a.u. When atoms are subjected to intense laser fields, electrons can be teared off the atom by tunneling through the potential barrier induced by the laser. Atoms are composed of inner- and outer-shell electrons. Electrons from the inner shell are more closely bound to the nucleus than electrons from the outer shell. We consider a single electron in the outer shell. We neglect the ionization of the electron [89, 31]. We consider that the atom is composed of a single-active electron (the outer electron) and an *ionic core*. The ionic core is composed of the nucleus and the electrons of the inner shell, whose dynamics is frozen. The effective charge of the ionic core seen by the outer electron is screened by the inner-shell electrons.

The duration of the laser field is of the order of tens to hundreds of femtoseconds (where 1 fs = 10^{-15} s) which corresponds to about ten laser cycles. After ionization, using a dimensional analysis, the *characteristic* velocity v_{chara} of the electron in the laser field is

$$v_{\rm chara} \sim E_0/\omega.$$
 (1.1)

During one femtosecond, according to the characteristic velocity of the electron, the electron could travel a distance of order 4 - 400 a.u. In contrast, the characteristic velocity of the ionic core in the field is at least

a thousand times smaller than the velocity of the electron. This is mainly due to the mass of the nucleus which is at least a thousand times larger than the mass of the electron. In a femtosecond, the dynamics of the ionic core is negligible compared to the characteristic size of the atom. We assume the ionic core to be static, which leads to the *static ion approximation*.

The *characteristic length* of the electron in the laser field r_{chara} is

$$r_{\rm chara} \sim E_0/\omega^2,$$
 (1.2)

which corresponds to $r_{\text{chara}} \sim 1.5 - 150$ a.u. for the laser parameters used in this manuscript. The size of the atom, of order 1 a.u., is smaller than the characteristic length of the electron in the laser. The excursions of the electrons during recollisions are relatively far from the core. In addition, the characteristic length of the electron in the laser fields is at least a hundred times smaller than its wavelength, i.e., $E_0/\omega^2 \ll \lambda$. The spatial variations of the laser field are neglected, which leads to the *dipole approximation*. In the dipole approximation, the amplitude of the magnetic force $|\mathbf{v} \times \mathbf{B}| \sim v_{\text{chara}} E_0/c$, where \mathbf{v} is the velocity of the electron, is much smaller than the amplitude of the electric force $|\mathbf{E}| \sim E_0$. The magnetic force is negligible, and the Lorentz force is $\mathbf{F} = -(\mathbf{E} + \mathbf{v} \times \mathbf{B}) \approx -\mathbf{E}$.

The *characteristic energy* of the electron in the laser field corresponds to the ponderomotive energy

$$U_p = \frac{E_0^2}{4\omega^2},\tag{1.3}$$

The rest energy of the electron in atomic units is c^2 . The relativistic effects are negligible as long as $U_p \ll c^2$. For the laser wavelength considered in this manuscript, the critical intensity beyond which relativistic effects are no longer negligible correspond to $I \sim 10^{18} \text{ W} \cdot \text{cm}^{-2}$. Therefore, the system with the laser intensity used in this manuscript is in the *non-relativistic regime*. The different regimes of approximation are illustrated in Fig. 1.1 as a function of the intensity and the wavelength of the laser.

1.1.2 Field-free single-active electron atom

Since the early twentieth century, a series of models for describing the atom were proposed [160]. For instance, the solid sphere model of J. Dalton (1803), or later, the plum pudding model of J. J. Thomson [164] (1904), the nuclear model of E. Rutherford [151] (1911) and the planetary model of N. Bohr [26] (1913) (see Appendix B for a recall on this semi-classical approach). In 1926, quantum theory became wave mechanics, initiated by E. Schrödinger. Nowadays, it is the most accurate approach to describe atoms in the non-relativistic regime.

The ionic core is assumed to be static (upon the static ion approximation), and the wave function describing the state of the electron in the atom at time t is denoted $\Psi(\mathbf{r}, t)$. The probability of presence of the electron at position \mathbf{r} at time t is given by $|\Psi(\mathbf{r}, t)|^2$. The evolution of the wave function in time is governed by the time-dependent Schrödinger equation (TDSE)

$$\hat{H}\Psi(\mathbf{r},t) = i\frac{\partial\Psi(\mathbf{r},t)}{\partial t},\tag{1.4}$$

where H is the Hamiltonian operator. In field-free atoms, the Hamiltonian operator is composed of kinetic energy terms and interaction potential terms governing the ion-electron interactions. The Hamiltonian operator of a single-active electron atom in the non-relativistic regime and in the static ion approximation reads

$$\hat{H}_{\text{free}} = \frac{\hat{\mathbf{p}}^2}{2} + V(\hat{\mathbf{r}}), \qquad (1.5)$$

where $\hat{\mathbf{p}}\Psi(\mathbf{r},t) = -i\partial\Psi(\mathbf{r},t)/\partial\mathbf{r}$ is the momentum operator and $\hat{\mathbf{r}}\Psi(\mathbf{r},t) = \mathbf{r}\Psi(\mathbf{r},t)$ is the position operator in the position representation. By substituting Hamiltonian (1.5) into the left-hand side of Eq. (1.4), we obtain the Schrödinger wave equation. The atom is initiated in the ground state of energy $-I_p$, where $I_p > 0$ is the ionization potential of the atom. The energy of the ground state of the atom is an eigenvalue of the Hamiltonian operator \hat{H}_{free} , and therefore

$$\hat{H}_{\text{free}}\Psi(\mathbf{r},t) = -I_p\Psi(\mathbf{r},t). \tag{1.6}$$

In the field-free atom, Hamiltonian (1.5) does not depend on time. With Eq. (1.4) and Eq. (1.6), we can write $-I_p\Psi(\mathbf{r},t) = i\partial\Psi(\mathbf{r},t)/\partial t$. We assume that the variables can be separated such that $\Psi(\mathbf{r},t) = \psi(\mathbf{r})\chi(t)$. We obtain $\chi(t) = \chi(0) \exp(iI_p t)$. Therefore, only the phase of the wave function of the electron changes in time in the field-free atom. The probability of presence of the electron is constant in time, $|\Psi(\mathbf{r},t)|^2 = |\psi(\mathbf{r})|^2$.

The space-dependence of the wave function of the electron is governed by the *time-independent Schrödinger* equation (TISE). The TISE is obtained by substituting Hamiltonian (1.5) into the left-hand side of Eq. (1.6). The TISE reads

$$\left[-\frac{\Delta}{2} + V(\mathbf{r})\right]\psi(\mathbf{r}) = -I_p\psi(\mathbf{r}),\tag{1.7}$$

where Δ is the Laplacian. The Hydrogen atom, for which the interaction potential $V(\mathbf{r}) = -|\mathbf{r}|^{-1}$ is solution of the three-dimensional Laplace equation, is one of the rare systems for which there exists a closed form solution for the wave function of the Schrödinger wave equation [116]. In our quantum calculations, the initial state of the electron is computed using the split-operator method with imaginary time propagation [13, 12].



Figure 1.2: (a) The blue and shaded green surfaces are the soft Coulomb potential [see Eq. (1.8)] for a = 1and the hard Coulomb potential [see Eq. (1.9)], respectively, for d = 2. (b) Soft Coulomb potential for a = 1, 2, 3 and the hard Coulomb potential (a = 0) as a function of the distance between the ion and the electron. (c) Ground state energy of the electron $-I_p$ of Eq. (1.6) obtained using the split-operator method with imaginary time propagation [13, 12]. In all panels, Z = 1.

In this manuscript, we use the *soft Coulomb potential* [78, 17] to describe the ion-electron interaction, which is commonly used in ultrafast processes involving laser-atom interactions [72, 73, 110, 81]. The soft Coulomb potential is defined as

$$V(\mathbf{r}) = -\frac{Z}{\sqrt{|\mathbf{r}|^2 + a^2}},\tag{1.8}$$

where a is the softening parameter (see Fig. 1.2a). In Fig. 1.2b, the soft-Coulomb potential is depicted for different values of the softening parameter. The softening parameter controls the depth of the potential well. This parameter models the strength of the correlation between the ion and the electron. The effective charge Z models the charge of the ion seen by the active electron, screened by the inner-shell electrons. In our analytic predictions, we use Z to generalize our results on ionization of electrons from cations, which are studied in Ref. [52], for instance. In our numerical calculations, we use Z = 1. The soft Coulomb potential is not singular at the origin, which facilitates the description of the dynamics of the electron in one dimension (for d = 1). In Fig. 1.2c, we observe that, by changing a, the ionization potential of the atom $-I_p$ changes. The softening parameter a can be used to model different target atoms. Close to the core, the soft-Coulomb potential behaves like the harmonic oscillator $V(\mathbf{r}) \approx -Z/a + Z |\mathbf{r}|^2/2a^3$. Far from the core, the soft-Coulomb potential behaves like the *hard Coulomb potential*

$$V(\mathbf{r}) = -\frac{Z}{|\mathbf{r}|}.\tag{1.9}$$

The soft and hard Coulomb potential are the ion-electron potentials used in this manuscript. They are rotationally invariant, as it is the case for interaction potential in atoms. In some places of the manuscript, we use the notation $\mathcal{V}(r) = V(\mathbf{r})$ with $r = |\mathbf{r}|$ to generalize the results to rotationally invariant potentials.

1.1.3 Intense laser field perturbation

The Hamiltonian of the field-free atom used for describing the electron dynamics in ultrashort processes is given by Eq. (1.5). We use an intense laser field elliptically polarized to perturb the atom. In the *dipole approximation*, the expression of the electric field does not depend on the space variables. The characteristic length scale of the electron is not large enough to perceive the spatial variations of the laser field. Unless stated otherwise, the electric field is of the form

$$\mathbf{E}(t) = \frac{E_0 f(t)}{\sqrt{\xi^2 + 1}} \left[\mathbf{e}_x \cos(\omega t + \varphi_{\text{CEP}}) + \mathbf{e}_y \xi \sin(\omega t + \varphi_{\text{CEP}}) \right].$$
(1.10)

The amplitude of the laser is E_0 , its frequency is $\omega = 0.0584$ a.u. and its carrier-envelope phase (CEP) is φ_{CEP} . The period of the laser field is denoted $T = 2\pi/\omega$. The axes \mathbf{e}_x and \mathbf{e}_y are referred to as the major polarization axis and the minor polarization axis, respectively. The electric field is related to the vector potential by $\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$. The magnetic field, which is the curl of the vector potential, is zero under the dipole approximation. The laser polarization is $\xi \in [0, 1]$. For $\xi = 0$, the laser is linearly polarized (LP). For $\xi > 0$, the laser polarization is *anticlockwise*. For $\xi = 1$, the laser is circularly polarized (CP). We call low (resp. high) elliptically polarized laser fields, lasers for which the polarization is close to linear (resp. circular). The scaling $(\xi^2 + 1)^{-1/2}$ is there to preserve the amplitude of the Poynting vector for varying ellipticity ξ (see Sec. B.2.1). The envelope of the laser is f(t). In experiments, the duration of the laser pulse is usually obtained from the full width at half maximum (FWHM). The typical shape of the laser envelope f(t) is Gaussian due to the laser beam profile in the propagation direction. Figure 1.3 shows three shapes of laser envelope: The Gaussian envelope [175] $f(t) = \exp[-(t-4T)^2/2T^2]$, the trapezoidal envelope [110, 81] and the cosine envelope [165, 29] $f(t) = \cos^4[\pi(t-4T)/8T]$. For the trapezoidal envelope is $f(t) = t/T_u$ for $t \in [0, T_u], f(t) = 1$ for $t \in [T_u, T_u + T_p], f(t) = (T_u + T_p + T_d - t)/T_d$ for $t \in [T_u + T_p, T_u + T_p + T_d]$, and f(t) = 0 otherwise. Here, T_u , T_p and T_d are the duration of the ramp-up, plateau and ramp-down of the laser pulse, respectively. During the plateau, the laser field is T-periodic. This is particularly well suited for tools from nonlinear dynamics, as discussed in Sec. 1.3.1.



Figure 1.3: Laser field as a function of time per laser cycle for $\xi = 0.5$ and $\varphi_{\text{CEP}} = 0$. The red and blue curves are the laser field component along the major (\mathbf{e}_x) and minor (\mathbf{e}_y) polarization axes, respectively. The thick black curves are the laser envelope f(t) and -f(t). (a) Gaussian envelope: $f(t) = \exp[-(t-4T)^2/2T^2]$ (used in Ref. [175] for instance). (b) Trapezoidal envelope of 2-4-2: $T_u = 2$, $T_p = 4$ and $T_d = 2$ (used in Refs. [110, 81] for instance). (c) Cosine envelope: $f(t) = \cos^4[\pi(t-4T)/8T]$ (used in Refs. [165, 29] for instance).

The total Hamiltonian operator of the system, in the framework where the laser field is treated classically, is of the form

$$\hat{H}(t) = \hat{H}_{\text{free}} + \hat{H}_{\text{int}}(t).$$
 (1.11)

The interaction between the electron and the laser field is governed by $\hat{H}_{int}(t)$. The interaction between the electron and the laser has an explicit time dependence. In principle there is no eigenvalue of $\hat{H}(t)$ associated with the state of the electron at time t when the laser is turned on. The Hamiltonian operator between the electron and the laser in the velocity gauge reads [135]

$$\hat{H}(t) = \frac{1}{2} \left[\hat{\mathbf{p}} + \mathbf{A}(t) \right]^2 + V(\hat{\mathbf{r}}),$$

where $\hat{H}_{int}(t) = \hat{\mathbf{p}} \cdot \mathbf{A}(t) + |\mathbf{A}(t)|^2/2$ and $\mathbf{A}(t)$ is the vector potential. In the velocity gauge, the interaction Hamiltonian is written in terms of the momentum operator. In order to determine the expression of the

CHAPTER 1. ATOMS SUBJECTED TO INTENSE LASER FIELDS

interaction potential in terms of the position operator, we consider the unitary transformation $\bar{\Psi}(\mathbf{r},t) = \hat{U}(t)\Psi(\mathbf{r},t)$, with

$$\hat{U}(t) = \exp\left[i\hat{\mathbf{r}}\cdot\mathbf{A}(t)\right]. \tag{1.12}$$

Substituting this transformation in Eq. (1.4), and identifying with the TDSE $\hat{H}\bar{\Psi} = i\partial\bar{\Psi}/\partial t$, we obtain the expression of the new Hamiltonian $\hat{H} = \hat{U}\hat{H}\hat{U}^{\dagger} - i\hat{U}\partial\hat{U}^{\dagger}/\partial t$. The unitary transformation of Eq. (1.12) corresponds to a translation in momentum space. The first term reads $\hat{U}\hat{H}\hat{U}^{\dagger} = |\hat{\mathbf{p}}|^2/2 + V(\hat{\mathbf{r}})$. The second term reads $-i\hat{U}\partial\hat{U}^{\dagger}/\partial t = \hat{\mathbf{r}}\cdot\mathbf{E}(t)$, where we have used $\mathbf{E}(t) = -\partial\mathbf{A}(t)/\partial t$. Therefore, the Hamiltonian in the *length gauge* reads [135]

$$\hat{H}(t) = \frac{\hat{\mathbf{p}}^2}{2} + V(\hat{\mathbf{r}}) + \hat{\mathbf{r}} \cdot \mathbf{E}(t), \qquad (1.13)$$

where bars have been removed. In the length gauge, $\hat{H}_{int}(t) = \hat{\mathbf{r}} \cdot \mathbf{E}(t)$. The Hamiltonian in the *length gauge* highlights the effective potential of the electron. The effective potential seen by the electron oscillates with the laser field at a frequency ω .

In the classical treatment of the dynamics, the electron is seen as a particle. The position of the electron is \mathbf{r} and its canonical conjugate momentum is \mathbf{p} . The Hamiltonian governing the motion of the electron in phase space is the classical analog of the Hamiltonian operator given by Eq. (1.13). The classical Hamiltonian of the single-active electron in the dipole approximation reads

$$H(\mathbf{r}, \mathbf{p}, t) = \frac{|\mathbf{p}|^2}{2} + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t).$$
(1.14)

The motion of the electron in phase space is determined by solving Hamilton's equations $\dot{\mathbf{r}} = \partial H/\partial \mathbf{p}$ and $\dot{\mathbf{p}} = -\partial H/\partial \mathbf{r}$. Hamiltonian (1.14) is studied throughout the manuscript.

1.2 Description of the electronic dynamics in two steps

In this section, we study the quantum dynamics of the electron in atoms subjected to an intense laser field. We show that, after ionization, the quantum dynamics of the electron exhibits classical features. We use the Wigner quasi-probability distribution [167] to highlight the quantum-classical correspondence in phase space. The study of the quantum-classical correspondence allows us to introduce the two-step model which is used in Chap. 3 to study PMDs.

1.2.1 Comparison between the quantum and the classical description of the dynamics

Figure 1.4 shows the probability of presence of the electron $|\Psi(x,t)|^2$ as a function of the position and time per laser cycle. The wave function $\Psi(x,t)$ is solution of the TDSE given by Eq. (1.4) for Hamiltonian (1.13) for d = 1. The potential is the soft Coulomb potential given by Eq. (1.8). The model atom is H, for which $I_p = 0.5$ a.u. and $a = \sqrt{2}$. We use a trapezoidal envelope 2–4–2 ($T_u = 2T$, $T_p = 4T$ and $T_d = 2T$).

In Fig. 1.4, we observe that the maximum of the probability of presence of the electron is around the origin x = 0 for all time. It corresponds to the ground state of the electron which remains populated during the simulation. At time t = 8T, when the laser field is turned off, only around 95% of the wave function is outside the core (i.e., |x| > 10 a.u.). At t = T, when the amplitude of the laser field reaches a local maximum of $E_0/2$, we observe that a portion of the wave packet is released from the atom. At t = 1.5T, when the amplitude of the laser field reaches a local maximum of $3E_0/4$, we observe that a larger portion of the wave packet is released from the atom than at time t = T. This ionization channel is observed every time the amplitude of the laser field is large. After the ionization, further away from the core, we observe white stripes which correspond to local maxima of the probability of presence of the electron. Far from the core, at |x| > 10 a.u., we observe that the local maxima of the probability of presence of the electron.

In Fig. 1.4, we show the positions of classical trajectories of Hamiltonian (1.14) for d = 1 as a function of time per laser cycle. The trajectories are initiated outside the potential barrier of the atom, at $x_0 = -I_p \mathbf{E}(t_0) \cdot \mathbf{e}_x / |\mathbf{E}(t_0)|^2$ at time $t = t_0$ with zero initial momentum (see Sec. 1.2.2.1). The magenta trajectory ionizes without returning to the parent ion, this is a *direct ionization*. We observe that the magenta trajectory exhibits the same features as the white stripes of the probability of presence of the ionized quantum wave packets. In particular, right after ionization, the magenta trajectory overlaps with a white stripe for more



Figure 1.4: Probability of presence of the electron in logarithmic scale solution of the TDSE (1.4) with Hamiltonian (1.13) for d = 1 as a function of the position x and the time per laser cycles. The interaction potential is the soft Coulomb potential (1.8) with Z = 1 and $a = \sqrt{2}$ and Z = 1 ($I_p = 0.5$ a.u., corresponding to H atom). The laser intensity is $I = 10^{14}$ W \cdot cm⁻², the wavelength is $\lambda = 780$ nm and the laser envelope is trapezoidal 2–4–2 ($T_u = 2T$, $T_p = 4T$ and $T_d = 2T$). The red, pink and purple trajectories are initiated according to the ADK theory (see Eq. (1.23)). The red trajectory is a *subcycle recollision*, the pink trajectory is a *Coulomb-driven recollision* and the magenta trajectory is a *direct ionization* (see Sec. 3.1.2.3 for details).

than one laser cycle. The red and pink trajectories also overlap with a white stripe for more than a laser cycle after ionization. After ionization, the red trajectory goes far from the core, and then returns to the core, at x = 0, in less than one laser cycle. This is a *subcycle recollision*. After the first recollision, the red trajectory recollides multiple times, and then ionizes. The pink trajectory goes far from the core, at about 50 a.u. Then, the electron comes back to the core after multiple laser cycles. This is a multiple laser cycle recollision, referred to as a *Coulomb-driven recollision*. The nature of the trajectories is discussed in details in Chap. 3. Hence, far from the core, after ionization, the quantum wave function manifests similarities with the purely classical trajectories of Hamiltonian (1.14).

In order to highlight the similarities of the classical and quantum dynamics after ionization, we consider the scattering experiment [166, 20]. In the scattering experiments, the wave packet is initiated at the peak amplitude of the laser field, at the quiver radius E_0/ω^2 . The initial wave function is $\Psi(x,0) = (5\pi)^{-1/4} \exp[-(x - E_0/\omega^2)^2/10]$. The laser envelope is f = 1. In order to visualize the evolution of the wave function in phase space, we use the Wigner quasi-probability distribution [167]

$$\mathcal{W}(x, p_x, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Psi^*\left(x - \frac{y}{2}, t\right) \Psi\left(x + \frac{y}{2}, t\right) \exp\left(-ip_x y\right) \,\mathrm{d}y,\tag{1.15}$$

where $\Psi(x,t)$ is solution of the TDSE given by Eq. (1.4) for Hamiltonian (1.13). Figure 1.5 shows snapshots of the Wigner quasi-probability distribution and the classical trajectories of Hamiltonian (1.14). Initially, the classical trajectories of Hamiltonian (1.14) are distributed according to the initial condition of the Wigner quasi-probability distribution $\mathcal{W}(x, p_x, 0)$. By substituting the initial conditions of the wave function in Eq. (1.15), we obtain $\mathcal{W}(x, p_x, 0) = (2/\pi) \exp(-5p_x^2) \exp[-(x - E_0/\omega^2)^2/5]$. Therefore, the most probable trajectory is initiated at the quiver radius $x = E_0/\omega^2$ with zero initial momentum.

In Fig. 1.5, we observe that the evolution of the classical distribution and the Wigner distribution in phase space is very similar, at least qualitatively. For instance, at t = 1.3T, the classical distribution and the Wigner distribution are both centered at $x \approx 0$ and $p_x \approx -1$. At t = 1.8T, both distributions are centered at $x \approx 0$ and $p_x \approx 1$. The spread of the distributions of the classical trajectories and the Wigner



Figure 1.5: Snapshots of the evolution of the Wigner quasi-probability distributions (1.15) (white panels) and the classical electron trajectories (grey panels) in the scattering experiment. The interaction potential is the soft Coulomb potential (1.8) with Z = 1 and $a = \sqrt{2}$ and Z = 1 ($I_p = 0.5$ a.u., corresponding to H atom). The laser intensity is $I = 10^{14}$ W \cdot cm⁻², the wavelength is $\lambda = 780$ nm and the laser envelope is f = 1. Initially, the wave function is $\Psi(x,0) = (5\pi)^{-1/4} \exp[-(x - E_0/\omega^2)^2/10]$, and the classical trajectories are distributed according to the initial Wigner quasi-probability distribution $\mathcal{W}(x, p_x, 0) = (2/\pi) \exp(-5p_x^2) \exp[-(x - E_0/\omega^2)^2/5]$. The dynamics of the classical trajectories is governed by Hamiltonian (1.14) for d = 1. The maximum of the absolute value of the Wigner quasi-probability distribution is set to unity for each time. All quantities are in a.u.

quasi-probability distributions is also very similar. The classical dynamics of the electron is the skeleton of its quantum dynamics. The distributions of the classical trajectories are structured along lines. These lines are *invariant manifolds* which drive the electron dynamics. How invariant manifolds drive the dynamics of the electron is studied in details in Chap. 5. In Fig. 1.5, we observe that the main features absent in the classical distribution of the electron are the interference patterns in the Wigner quasi-probability distribution. These features are due to the interferences between the recolliding wave packet and parts of the wave packet which remains trapped close to the core [166, 21]. These interferences play a central role in HHG, which is discussed in Sec. 4.2. The interference patterns can be retrieved from the classical action in a semi-classical approach [103].

In this manuscript, we focus on the purely classical features that the quantum dynamics of the electron exhibits after ionization. We take advantage of the similarities between the classical and quantum dynamics of the electron to understand, with the help of classical trajectories, the mechanisms underlying the nonlinear phenomena observed in experiments. In this manuscript, we use the quantum theory of ionization to obtain the initial classical distribution in phase space. This procedure is referred to as the *classical trajectory Monte Carlo* (CTMC) simulations, and has two steps:

- In step (i), the electron tunnel-ionizes through the potential barrier induced by the laser. The quantum theory of tunnel-ionization [83, 137, 138, 136, 8] is used to determine the initial distribution of the electron in phase space.
- In step (ii), the nonlinear dynamics of the electron is treated with a purely classical model, in the Hamiltonian formalism, in terms of trajectories in phase space.



Figure 1.6: Left panel: Two-step model of the electron. The blue surface is the effective potential $V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t_0)$. The red arrow is the tunnel-ionization of the electron corresponding to step (i). The blue arrow is the electron trajectory corresponding to step (ii). Right panel: Effective potential of Hamiltonian (1.13) in the adiabatic approximation (1.17) with $\mathbf{E}(t) = E_0 \mathbf{e}_{\parallel}(t_0)$, for $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, a = 0 (hard Coulomb potential), Z = 1, and $I_p = -0.5$ a.u. The blue curve is the effective potential $-1/|r_{\parallel}| + E_0 r_{\parallel}$. The blue shaded region is the classical forbidden region. The green dashed curve is the effective potential in the SFA $r_{\parallel}E_0$. The red shaded region is the initial wavefunction of the electron. The vertical dotted line are the boundaries between the regions I, II and III.

1.2.2 Step (i): Ionization of the electron from the ground state

When the laser field oscillates, it creates a potential barrier through which the electron can tunnel-ionize [83, 8, 137, 138, 136], as shown in the left panel of Fig. 1.6. The characteristic velocity of the electron in the ground state is $\sqrt{2I_p}$. From a semi-classical point of view, the characteristic ionization time of the electron is the time it spends under the potential barrier of length I_p/E_0 . The ionization frequency is therefore $\omega_i = E_0 \sqrt{2/I_p}$. Typically, for an atom with $I_p \sim 1$ eV and for the laser parameters considered in this manuscript, the ionization frequency is around $\omega_i \sim 4 \times 10^{-2} - 4$ a.u. $\approx 10^{-18} - 10^{-16}$ rad \cdot s⁻¹. The Keldysh parameter [83]

$$\gamma = \frac{2\omega}{\omega_i} = \frac{\sqrt{2I_p\omega}}{E_0},\tag{1.16}$$

is used to estimate the dominant ionization process. We notice that if we consider the characteristic velocity of the electron under the potential barrier to be $v_{\text{chara}} \sim E_0/\omega$ instead of $\sqrt{2I_p}$, the ionization frequency becomes $\omega_i = E_0^2/\omega I_p$ and the ratio $2\omega/\omega_i = 2I_p\omega^2/E_0^2$, which corresponds to γ^2 . If $\gamma \ll 1$, the barrier oscillates slowly compared to the ionization frequency, and the dominant ionization process is the *adiabatic* tunnel ionization [83, 8]. If $\gamma \gg 1$, the barrier oscillates quickly compared to the ionization frequency, and the dominant ionization process is *multiphoton absorption* [6, 57]. For $\gamma \sim 1$, the process is in between tunnel ionization and multiphoton absorption. During this so-called nonadiabatic tunnel-ionization [137, 138, 136], the wavepacket absorbs photons during the tunneling [86, 124, 125]. For the laser parameters considered in this manuscript and $I_p \sim 1$ eV, the Keldysh parameter is around $\gamma \sim 3 \times 10^{-2} - 3$. Therefore, the regime of ionization for the parameters investigated in this manuscript are the adiabatic and nonadiabatic tunnel ionization. Many tunneling ionization rates have been derived with various techniques, see for instance Refs. [172, 27, 104, 86]. In this section, we show the results for the two main ionization theories, namely the Ammosov-Delone-Krainov [83, 8] (ADK) and the Perelomov-Popov-Terent'ev [137, 138, 136] (PPT) ionization theories. We summarize the derivation of the ADK theory and we summarize the results of the PPT theory. Then, we compare the results obtained in the adiabatic and nonadiabatic tunneling theory, and we identify the initial conditions of the T-trajectory which corresponds to the most probable trajectory according to the tunneling ionization rate.

1.2.2.1 Ammosov-Delone-Krainov (ADK) ionization rate

The summary of the derivation of the ADK theory of ionization follows Refs. [83, 8, 117, 123]. We consider the adiabatic regime $\gamma \ll 1$, in which the frequency of oscillations of the barrier is very small compared to the frequency of ionization. We denote t_0 the time at which the electron is outside the potential barrier. We use the adiabatic approximation in which the laser field is static

$$\mathbf{E}(t) \approx \mathbf{E}(t_0) = |\mathbf{E}(t_0)| \mathbf{e}_{\parallel}(t_0), \tag{1.17}$$

where $\mathbf{e}_{\parallel}(t_0) = \mathbf{E}(t_0)/|\mathbf{E}(t_0)|$ is the direction of the electric field. The polarization plane is $(\mathbf{e}_x, \mathbf{e}_y)$ and therefore $\mathbf{e}_{\parallel}(t_0) = [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_x + [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_y$ and $\mathbf{e}_{\perp}(t_0) = -[\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_y]\mathbf{e}_x + [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_y$. The Hamiltonian operator governing the evolution of the wave function is given by Eq. (1.13). The ionization occurs relatively far from the core, therefore we consider the hard Coulomb potential to describe the ion-electron interaction $V(\mathbf{r}) = -Z/|\mathbf{r}|$. Figure 1.6 shows the effective potential of Hamiltonian (1.13) in the adiabatic approximation along $\mathbf{e}_{\parallel}(t_0)$. In the right panel of Fig. 1.6, three regions are depicted. The saddle point $\mathbf{r}^* = -\sqrt{Z/|\mathbf{E}(t_0)|}\mathbf{e}_{\parallel}(t_0)$ is where the Coulomb interaction and the laser interaction have the same amplitude, $|V(\mathbf{r}^*)| = |\mathbf{r}^* \cdot \mathbf{E}(t_0)|$. Each region corresponds to a regime of approximation. The wave function in region I and II are denoted $\Psi_{\rm I}$ and $\Psi_{\rm II}$, respectively.

Region I: Atom in the ground state In region I, for which $|\mathbf{r}| < |\mathbf{r}^{\star}|$, the Coulomb potential is dominant. We assume that the Hamiltonian operator governing $\Psi_{I}(\mathbf{r})$ corresponds to the Hamiltonian operator of the field-free atom (1.5). The wave function in region I is described by Eq. (1.7). For rotationally invariant potential, the wavefunction is of the form $\Psi_{I}(\mathbf{r}) = R_{nl}(|\mathbf{r}|)Y_{l}^{m}(\mathbf{r}/|\mathbf{r}|)$, where $Y_{l}^{m}(\mathbf{r}/|\mathbf{r}|)$ are spherical harmonics [116], $R_{nl}(|\mathbf{r}|)$ is the radial wave function, n is the principal quantum number, l is the angular quantum number and m is the magnetic quantum number. In Refs. [83, 8, 123], the asymptotic solution of the radial wave function (in $|\mathbf{r}|$) is used in the theory of ionization. For hard Coulomb potential (1.9), the asymptotic radial wavefunction [116] is of the form

$$\Psi_{\mathrm{I}}(\mathbf{r}) \approx C_{nl} |\mathbf{r}|^{Z/2I_p - 1} \exp(-\sqrt{2I_p} |\mathbf{r}|) Y_l^m(\mathbf{r}/|\mathbf{r}|), \qquad (1.18)$$

where C_{nl} is a constant which depends on the wavefunction of the atom near the origin (see Ref. [8, 123] for an approximation of the expression of C_{nl} , and Ref. [116] for the expression of the spherical harmonics). The probability of presence of the electron decreases exponentially with its distance from the ionic core.

Region II: Tunnel ionization and WKB approximation In region II, for which $|\mathbf{r}^*| < -r_{\parallel} < |\mathbf{r}_{ADK}|$, the laser interaction is dominant. We assume that the Coulomb potential is neglected. In the adiabatic approximation, the Hamiltonian operator (1.13) is autonomous (no explicit time dependence), and the eigenvalue is the energy of the ground state $-I_p$. The wave function in region II is given by

$$\left[\frac{\hat{\mathbf{p}}^2}{2} + \hat{\mathbf{r}} \cdot \mathbf{E}(t_0)\right] \Psi_{\mathrm{II}}(\mathbf{r}) = -I_p \Psi_{\mathrm{II}}(\mathbf{r}).$$
(1.19)

We consider the wavefunction in terms of the position operator along the laser field direction $\hat{r}_{\parallel} = \hat{\mathbf{r}} \cdot \mathbf{e}_{\parallel}(t_0)$, transverse to the laser field direction $\hat{r}_{\perp} = \hat{\mathbf{r}} \cdot \mathbf{e}_{\perp}(t_0)$ and perpendicular to the polarization plane $\hat{z} = \hat{\mathbf{r}} \cdot \mathbf{e}_z$, with $\mathbf{e}_{\parallel}(t_0) \times \mathbf{e}_{\perp}(t_0) = \mathbf{e}_z$. In the position representation, the momentum operators are denoted $\hat{p}_{\parallel} = -i\partial/\partial r_{\parallel}$, $\hat{p}_{\perp} = -i\partial/\partial r_{\perp}$ and $\hat{p}_z = -i\partial/\partial z$. We perform a partial Fourier-transform [123] in the transverse and perpendicular direction to the laser field. We denote $\Upsilon_{\mathrm{II}}(r_{\parallel}, p_{\perp}, p_z) = \mathcal{F}[\Psi_{\mathrm{II}}(r_{\parallel}, r_{\perp}, z)](p_{\perp}, p_z)$. The TISE (1.19) becomes

$$\left[\frac{\partial^2}{\partial r_{\parallel}^2} + k(r_{\parallel})^2\right] \Upsilon_{\rm II}(r_{\parallel}, p_{\perp}, p_z) = 0, \qquad (1.20)$$

where $k(r_{\parallel}) = [-2(I'_p + r_{\parallel}|\mathbf{E}(t_0)|)]^{1/2}$ and $2I'_p = 2I_p + p_{\perp}^2 + p_z^2$ is the effective energy of the electron. In order to determine wave function in region II from Eq. (1.20), we use the Wentzel–Kramers–Brillouin (WKB) approximation [117]. The WKB approximation is valid if $|\partial k/\partial r_{\parallel}| \ll |k|^2$. We assume that $\Upsilon_{\rm II}(r_{\parallel}, p_{\perp}, p_z) = \exp[iv_j(r_{\parallel}, p_{\perp}, p_z)]$. We substitute this form of the wave function in Eq. (1.20) and rewriting the equation in a fixed point equation, we obtain

$$v_j(r_{\parallel}, p_{\perp}, p_z) = \pm \int^{r_{\parallel}} \left[k(s)^2 + i \frac{\partial^2 v_j(s, p_{\perp}, p_z)}{\partial s^2} \right]^{1/2} \mathrm{d}s.$$
(1.21)

The solution of $v_j(r_{\parallel}, p_{\perp}, p_z)$ is determined by recurrence using the Picard-Lindelöf theorem. At the zeroth order, we assume that $|\partial^2 v_0 / \partial r_{\parallel}^2| \ll |\partial v_0 / \partial r_{\parallel}|^2$, or equivalently that $|\partial^2 v_0 / \partial r_{\parallel}^2| \ll |k(r_{\parallel})|^2$. Therefore, $v_0(r_{\parallel}, p_{\perp}, p_z) = \pm \int^{r_{\parallel}} k(s) \, ds$. At the first order, we substitute the solution of $v_0(r_{\parallel}, p_{\perp}, p_z)$ in the right-hand side of Eq. (1.21). Using a Taylor expansion with the assumption $|\partial^2 v_0 / \partial r_{\parallel}^2| \ll |k(r_{\parallel})|^2$, one obtains $v_1(r_{\parallel}, p_{\perp}, p_z) \approx \pm \int^{r_{\parallel}} k(s) \, ds + i \log \sqrt{k(r_{\parallel})} + C$, with *C* a constant. At the first order, $\Upsilon_{\rm II} \approx \exp[iv_1(r_{\parallel}, p_{\perp}, p_z)]$. In region II, $k(r_{\parallel})^2 < 0$ and therefore $k(r_{\parallel}) = i\Im k(r_{\parallel})$ with $\Im k(r_{\parallel}) = [2(I'_p + r_{\parallel} |\mathbf{E}(t_0)|)]^{1/2}$. The wave function for $r_{\parallel} \in [-|\mathbf{r}_{\rm ADK}|, -|\mathbf{r}^{\star}|]$ is given by

$$\Upsilon_{\rm II}(r_{\parallel}, p_{\perp}, p_z) \approx \Upsilon_{\rm II}(-|\mathbf{r}^{\star}|, p_{\perp}, p_z) \sqrt{\frac{k(-|\mathbf{r}^{\star}|)}{k(r_{\parallel})}} \\
\exp\left[\frac{(2I'_p + 2r_{\parallel}|\mathbf{E}(t_0)|)^{3/2}}{3|\mathbf{E}(t_0)|} - \frac{(2I'_p - 2|\mathbf{r}^{\star}||\mathbf{E}(t_0)|)^{3/2}}{3|\mathbf{E}(t_0)|}\right].$$
(1.22)

Region III: Distribution of the initial conditions of the electron in phase space In region III, for which $r_{\parallel} < -|\mathbf{r}_{ADK}|$, we consider the dynamics to be classical. The tunnel exit of the electron is outside the barrier in the direction of the electric field. In the adiabatic approximation, the energy of the electron after ionization is $2I'_p = 2I_p + p_{\perp}^2 + p_z^2$. In the adiabatic approximation, the initial conditions of the electron are

$$\mathbf{r}_{\text{ADK}} = -\mathbf{e}_{\parallel}(t_0) \frac{I'_p}{2|\mathbf{E}(t_0)|} \left(1 + \sqrt{1 - \frac{4Z|\mathbf{E}(t_0)|}{I'_p^2}} \right) \approx -\mathbf{e}_{\parallel}(t_0) \frac{I_p}{|\mathbf{E}(t_0)|}, \quad (1.23a)$$

$$\mathbf{p}_{\text{ADK}} = \mathbf{e}_{\perp}(t_0)p_{\perp} + p_{z,0}\mathbf{e}_z. \tag{1.23b}$$

The initial position of the electron corresponds to $|\mathbf{p}_{ADK}|^2/2 + V(\mathbf{r}_{ADK}) + \mathbf{r}_{ADK} \cdot \mathbf{E}(t_0) = -I_p$. In Eq. (1.23a), we have used that the electron ionizes relatively far from the core $4Z|\mathbf{E}(t_0)|/I'_p^2 \ll 1$ and with a small initial kinetic energy compared to the ionization potential $I_p \gg |\mathbf{p}_{ADK}|^2/2$. Each trajectory is weighted by the ionization rate associated with the initial conditions. The ADK ionization rate is given by $W_{ADK}(t_0, \mathbf{p}_{ADK}) = |\Upsilon_{II}(-|\mathbf{r}_{ADK}|, p_{\perp}, p_z)|^2$. In Eq. (1.22), we use $|\mathbf{r}^*| = 0$ and the expansion $(2I_p + |\mathbf{p}_{ADK}|^2)^{3/2} \approx (2I_p)^{3/2} + |\mathbf{p}_{ADK}|^2\sqrt{2I_p}$. The ADK ionization rate, for a given ionization time t_0 and initial momentum \mathbf{p}_{ADK} is therefore

$$W_{\rm ADK}(t_0, \mathbf{p}_{\rm ADK}) \propto \exp\left[-\frac{2(2I_p)^{3/2}}{3|\mathbf{E}(t_0)|}\right] \times \exp\left[-\frac{\sqrt{2I_p}}{2|\mathbf{E}(t_0)|} \left(p_{\perp}^2 + p_{z,0}^2\right)\right].$$
 (1.24)

In Eq. (1.24), the ionization rate is written in terms of proportionality for two reasons: In the CTMCs calculations, only the relative amplitude of the ionization rate is useful (see Sec. D.1.1), and in experiments, the constant is obtained by interpolating the ionization rate at a given intensity. Equation (1.24) is therefore a convenient form for the CTMCs computations and for post-processing purposes. In Ref. [123], the expression of the constant is reported and is obtained by matching the asymptotic expression of the wave function in region I (1.18) with the wave function in region II.

1.2.2.2 Perelomov-Popov-Terent'ev (PPT) ionization rate

In the nonadiabatic regime $\gamma \lesssim 1$, the frequency of the barrier is of the same order as the frequency of the ionization rate. The oscillations of the barrier must be taken into account and the adiabatic approximation (1.17) no longer holds. In 1966, a series of papers [137, 138, 136] has been published, which introduced some techniques to account for the variations of the laser field using the Green functions formalism and the complex trajectories (saddle point method) for a short-range potential $V(\mathbf{r}) \propto \delta(\mathbf{r})$, with δ the Dirac distribution function. Here, we summarize the results of the PPT theory: The initial conditions of the electron are parametrized by the ionization time t_0 and its momentum \mathbf{p}_{PPT} at t_0 ,

$$\mathbf{r}_{\text{PPT}} = \mathbf{e}_{\parallel}(t_0) \frac{|\mathbf{E}(t_0)|}{\omega^2} (1 - \cosh \tau_0),$$
 (1.25a)

$$\mathbf{p}_{\text{PPT}} = \mathbf{e}_{\parallel}(t_0)p_{\parallel} + \mathbf{e}_{\perp}(t_0)p_{\perp} + \mathbf{e}_z p_{z,0}, \qquad (1.25b)$$

with $\mathbf{e}_{\parallel}(t_0) = \mathbf{E}(t_0)/|\mathbf{E}(t_0)|$ and $\mathbf{e}_{\perp}(t_0) = -[\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_y]\mathbf{e}_x + [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_y$. In other words, p_{\parallel} is the initial momentum of the electron along the laser field direction at ionization, p_{\perp} is the initial momentum transverse to the laser field direction, and $p_{z,0}$ is the initial momentum perpendicular to the polarization plane $(\mathbf{e}_x, \mathbf{e}_y)$. The time $\tau_0 = \tau_0(\gamma_0)$ is solution of the transcendental equation

$$\sinh^2 \tau_0 - \xi^2 \left(\cosh \tau_0 - \frac{\sinh \tau_0}{\tau_0} \right)^2 = \gamma_0(t_0)^2, \tag{1.26}$$

where $\gamma_0(t_0) = \omega \sqrt{2I_p}/|\mathbf{E}(t_0)|$. For $\tau_0(\gamma_0) \ll 1$, we obtain $\tau_0(\gamma_0) \approx \gamma_0$. The PPT ionization rate [138] for the CTMC calculations reads

$$W_{\rm PPT}(t_0, \mathbf{p}_{\rm PPT}) \propto \exp\left[-\frac{2I_p}{\omega}g(\gamma_0)\right] \times \exp\left\{-\frac{1}{\omega}\left[c_{\parallel}(\gamma_0)p_{\parallel}^2 + c_{\perp}(\gamma_0)\left(p_{\perp} - p_{\perp,0}^{\rm max}\right)^2 + c_z(\gamma_0)p_{z,0}^2\right]\right\}, \quad (1.27)$$

where the functions g is

$$g(\gamma_0) = \left(1 + \frac{1+\xi^2}{2\gamma_0^2}\right)\tau_0(\gamma_0) - (1-\xi^2)\frac{\sinh 2\tau_0(\gamma_0)}{4\gamma_0^2} - \xi^2\frac{\sinh^2\tau_0(\gamma_0)}{\gamma_0^2\tau_0(\gamma_0)}.$$
(1.28)

The coefficients c_{\parallel} , c_{\perp} and c_z , which are inversely proportional to the square of the standard deviation of the distribution along the longitudinal and transverse momentum, are given by

$$c_{\parallel}(\gamma_0) = \tau_0(\gamma_0) - \sigma(\gamma_0) \tanh \tau_0(\gamma_0), \qquad (1.29a)$$

$$c_{\perp}(\gamma_0) = \tau_0(\gamma_0) + \sigma(\gamma_0) \xi^2 \frac{[\tau_0(\gamma_0) - \tanh \tau_0(\gamma_0)]^2}{\tau_0(\gamma_0)^2 \tanh \tau_0(\gamma_0)},$$
(1.29b)

$$c_z(\gamma_0) = \tau_0(\gamma_0), \qquad (1.29c)$$

with the notation

$$\sigma(\gamma_0) = \left(1 - \xi^2 + \xi^2 \frac{\tanh \tau_0(\gamma_0)}{\tau_0(\gamma_0)}\right)^{-1}.$$

The coefficients satisfy $c_{\parallel} < c_{\perp} \leq c_z$, implying that the distributions are more spread out along the longitudinal direction than along the transverse or perpendicular directions. The most probable initial transverse momentum $p_{\perp,0}^{\max}$ is

$$p_{\perp,0}^{\max} = \frac{\xi E_0}{\omega \sqrt{\xi^2 + 1}} \left(1 - \frac{\sinh \tau_0(\gamma_0)}{\tau_0(\gamma_0)} \right).$$
(1.30)

1.2.2.3 T-trajectory and comparison of adiabatic and nonadiabatic theories

The ionization rate in the ADK adiabatic theory and the PPT nonadiabatic theory are given in Eq. (1.24) and Eq. (1.27), respectively. In the adiabatic (ADK) theory of ionization, the electric field is assumed to be static during the ionization of the electron. The associated Keldysh parameter (1.16) is very small, $\gamma \ll 1$. In other terms, the potential barrier under which the electron ionizes is very thin, and the ionization is almost instantaneous. In the PPT ionization rate, the "time spent by the electron under the potential barrier" [152] is given by τ_0 , solution of Eq. (1.26). In the adiabatic limit for which $\gamma_0 \to 0$, we obtain $\tau_0 \approx \gamma_0$ by Taylor expanding the left-hand side of Eq. (1.26) to the second order in τ_0 . Therefore, in the semi-classical representation, tunnel-ionization is "instantaneous" in the adiabatic limit. The first order Taylor expansion in γ_0 of Eq. (1.28) is $g(\gamma_0) \approx (2/3)\gamma_0$, and therefore $\exp(-2I_pg(\gamma_0)/\omega) \approx \exp[-2(2I_p)^{2/3}/3|\mathbf{E}(t_0)|]$ in the adiabatic limit. The first order Taylor expansion in γ_0 of the coefficients given in Eqs. (1.29) are $c_{\parallel} \approx 0$ and $c_{\perp} \approx c_z \approx \gamma_0$. The first order Taylor expansion in γ_0 of the most probable transverse momentum given in Eq. (1.30) is $p_{\perp,0}^{\max} \approx 0$. Therefore, $\exp\{-[c_{\parallel}p_{\parallel}^2 + c_{\perp}(p_{\perp} - p_{\perp,0}^{\max})^2 + c_z p_z^2]/\omega\} \approx \exp[-\sqrt{2I_p}(p_{\perp}^2 + p_{z,0}^2)]$, and $W_{\rm PPT} \approx W_{\rm ADK}$ in the adiabatic limit. In addition, the first order expansion initial position of the electron in the PPT theory given in Eq. (1.25) is $\mathbf{r}_{\rm PPT} \approx \mathbf{r}_{\rm ADK}$ in the adiabatic limit. Therefore, the nonadiabatic (PPT) theory provides the same results as the adiabatic (ADK) theory in the adiabatic limit.

In both theories, the maximum of the ionization rate is reached when the electron ionizes at the peak amplitude of the laser field. In other terms, the most probable time at which the electron is outside the potential barrier is such that the laser field is at the peak amplitude. For a laser field as given in Eq. (1.10) and f = 1, the peak amplitude of the laser $E_0/\sqrt{\xi^2 + 1}$ is reached at time $\omega t_0 + \varphi_{\text{CEP}} = k\pi$, with $k \in \mathbb{Z}$. In the adiabatic theory, the most probable initial position and momentum of the electron are

$$\mathbf{r}_{\text{ADK}}^{\text{T}} = -\mathbf{e}_{\parallel}(t_0) \frac{I_p \sqrt{\xi^2 + 1}}{E_0},$$
 (1.31a)

$$\mathbf{p}_{\mathrm{ADK}}^{\mathrm{T}} = \mathbf{0}, \qquad (1.31\mathrm{b})$$

where we have used Eqs. (1.23) and (1.24). The most probable trajectory according to the ionization rate is referred to as the T-trajectory. In the ADK theory, the initial position and momentum of the T-trajectory are given by Eqs. (1.31) with t_0 which maximizes the amplitude of the laser field. The T-trajectory ionizes in the opposite direction of the laser field direction. In the nonadiabatic theory, the most probable initial position and momentum of the electron are

$$\mathbf{r}_{\rm PPT}^{\rm T} = \mathbf{e}_{\parallel}(t_0) \frac{E_0}{\omega^2 \sqrt{\xi^2 + 1}} \left(1 - \cosh \tau\right), \qquad (1.32a)$$

$$\mathbf{p}_{\text{PPT}}^{\text{T}} = \mathbf{e}_{\perp}(t_0) \frac{\xi E_0}{\omega \sqrt{\xi^2 + 1}} \left(1 - \frac{\sinh \tau}{\tau} \right), \qquad (1.32\text{b})$$

where τ is the solution of Eq. (1.26) for $\gamma_0 = \gamma \sqrt{\xi^2 + 1}$. We have used Eqs. (1.25) and (1.27). In the PPT theory, the initial position and momentum of the T-trajectory are given by Eqs. (1.32) with t_0 maximizing the amplitude of the laser field. The T-trajectory in the PPT theory ionizes closer to the core than in the ADK theory. Hence, the ionization rate associated with the T-trajectory is the maximum ionization rate, and the T-trajectory captures the dominant behavior of the ionized electrons.

1.2.3 Step (ii): Classical motion of the ionized electron

After ionization, the dynamics of the electron is described purely classically. The Hamiltonian governing the motion of the electron in phase space is given by Eq. (1.14). It corresponds to the classical analog of the Hamiltonian operator given by Eq. (1.13). Figure 1.6 depicts step (ii).

Close to the core, the ion-electron interaction in Hamiltonian (1.14) is dominant compared to the laser interaction. If there is no laser field, $\mathbf{E}(t) = \mathbf{0}$, Hamiltonian (1.14) is autonomous. There are *d* degrees of freedom in the system (*d* is the dimension of the configuration space). The potential is rotationally invariant (i.e., the force is central), and as a consequence the angular momentum of the electron is conserved, which corresponds to d - 1 constants of the motion in involution. The system is integrable in the Liouville sense and cannot exhibit chaos. As a consequence, there is no sensitivity with respect to the initial conditions.

Far from the core, the laser interaction in Hamiltonian (1.14) is dominant compared to the ion-electron interaction. If there is no ion-electron interaction, $V(\mathbf{r}) = 0$, Hamiltonian (1.14) is explicitly time-dependent and the equations of motion of the electron are linear. In particular, the momentum of the electron is

$$\mathbf{p}(t) = \mathbf{p}_0 - \mathbf{A}(t_0) + \mathbf{A}(t), \tag{1.33}$$

where $\mathbf{p}_0 = \mathbf{p}(t_0)$ is the initial momentum after ionization, t_0 is its ionization time and the vector potential is such that $\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$. This case is referred to as the strong field approximation (SFA). In the SFA, the drift momentum of the electron $\mathbf{p}(t) - \mathbf{A}(t) = \mathbf{p}_0 - \mathbf{A}(t_0)$ is conserved in time. Therefore, there are dconstants of the motion in involution in the SFA. The Hamiltonian system in the SFA is also integrable.

Figure 1.7a, Fig. 1.7c and Fig. 1.7e show the position of the ADK T-trajectory as a function of time per laser cycle t/T for different values of the ionization potential for d = 1. The dotted line is the SFA trajectory



Figure 1.7: The dimension of the configuration space is d = 1, the laser intensity is $I = 10^{14}$ W · cm⁻², the softening parameter is a = 1, the effective charge is Z = 1, the laser field is given by Eq. (1.10) with $\varphi_{\text{CEP}} = 0$ and f = 1. Upper panels (a), (c), (e): Position of the ADK T-trajectory as a function of time t per laser cycle T. The horizontal line indicates the position of the ionic core at x = 0. The dotted line is the SFA trajectory. Middle panels (b), (d), (f): T-trajectory in the ADK theory in phase space (x, p_x) . The ionization potential is (a-b) $I_p = 0.5$ a.u., (c-d) $I_p = 0.2$ a.u., (e-f) $I_p = 0.1$ a.u. (g) Poincaré section of the electron dynamics given in Eq. (5.2). The squares are the initial conditions of the ADK T-trajectory. The triangles are the T-trajectory in phase space under the Poincaré map (5.2).

and the solid line is the trajectory of Hamiltonian (1.14) for a laser field of the form (1.10) and f = 1. The electron ionizes at $t_0 = 0$, at the peak amplitude of the laser field. In the SFA, the trajectory of the electron is $x(t) = -E_0/\omega^2 [1 + \gamma^2/2 - \cos(\omega t)]$, with γ given by Eq. (1.16). In the SFA, the T-trajectory oscillates in time without recolliding, i.e., without crossing x = 0. In contrast, the T-trajectory of Hamiltonian (1.14) exhibits different qualitative behaviors. In Fig. 1.7a, the T-trajectory of Hamiltonian (1.14) recollides with the core multiple times. In Fig. 1.7c, the T-trajectory of Hamiltonian (1.14) is trapped close to the core for half of a laser cycle before ionizing and recolliding. In Fig. 1.7e, the T-trajectory of Hamiltonian (1.14) is bounded and does not ionize. Therefore, the T-trajectory with and without taking into account the Coulomb potential are qualitatively very different. Even when the laser interaction is dominant compared to the ion-electron interaction after ionization [see Fig. 1.7a for instance], the ion-electron interaction acts significantly on the dynamics of the electron after ionization. In Chap. 3, we study the impact of the Coulomb potential on the motion of the electron using the comparison between the trajectories in the SFA and of Hamiltonian (1.14).

The difficulty in the analyses of the dynamics of the electron arises when the combined Coulomb and strong laser field must be both taken into account, i.e., $\mathbf{E}(t) \neq \mathbf{0}$ and $V(\mathbf{r}) \neq 0$. In this case, the angular momentum and the drift momentum of the electron are no longer conserved, and Hamiltonian (1.14) depends explicitly on time. The number of conserved quantities in the system is always smaller than the number of degrees of freedom. The system is not integrable in the Liouville sense, making the combined Coulomb and laser fields a fully nonlinear and chaotic system. In this manuscript, we study the classical motion of the electron after ionization given by the reference Hamiltonian (1.14) by taking into account the Coulomb and laser fields. In Chap. 2, we derive reduced models to describe the electron dynamics of Hamiltonian (1.14). In Chap. 5, we investigate qualitatively the phase space of the electron using nonperturbative methods.

1.3 Qualitative analysis of the T-trajectory in phase space

The T-trajectory of the electron is the most weighted trajectory according to the ionization rate of the quantum theory. The T-trajectory captures the dominant behavior of the ionized electrons. Understanding its behavior allows one to understand the global behavior of the ionized electrons. In this section, we describe qualitatively the dynamics of the T-trajectory in phase space, and we introduce the tools we use throughout the manuscript.

1.3.1 Dynamics of the electron for d = 1: Poincaré section

We first focus on LP fields ($\xi = 0$). For LP fields, the dynamics of the electron can be reduced to one dimensions (for d = 1) for $y = z = p_y = p_z = 0$. For the laser field of the form (1.10) with f = 1 and for the soft Coulomb potential (1.8), Hamiltonian (1.14) for d = 1 reads

$$H(x, p_x, t) = \frac{p_x^2}{2} - \frac{1}{\sqrt{x^2 + 1}} + xE_0 \cos(\omega t).$$
(1.34)

In absence of laser envelope, the CEP is chosen as $\varphi_{\text{CEP}} = 0$ without loss of generality. Figures 1.7b, 1.7d and 1.7f show the T-trajectory of Hamiltonian (1.34) in phase space, in the plane (x, p_x) , for different ionization potentials. The ionization time of the T-trajectory is chosen at $t_0 = 0$ (peak amplitude of the laser field). The initial condition of the electron is depicted with a square, at $x_{\text{ADK}}^T = -I_p/E_0$ and $p_{x,\text{ADK}}^T = 0$ [see Eqs. (1.23)].

In Fig. 1.7b, the T-trajectory goes far from the core, its momentum changes sign, and then comes back to the core and recollides with an energy approaching $2U_p$. At the recollision, there is a sudden peak of momentum around x = 0 due to the ion-electron interaction. Then, the T-trajectory recollides multiple times and ionizes. We observe that the shape of the T-trajectory during the multiple recollisions looks like the shape of the black curve. The black curve is a *periodic orbit* of Hamiltonian (1.34) of period T. One piece of the periodic orbit is far from the core, and another piece is close to the core. It is referred to as a recolliding periodic orbit [81, 80, 109, 3] (RPO). After ionization, the electron is driven by the *stable manifold* of the RPO, mimics the shape of the RPO, and then ionizes through its *unstable manifold*, as it is shown in Fig. 1.8a. Invariant objects in phase space, such as for instance the periodic orbits, play an instrumental role in driving the electron dynamics. Analyzing the shape of the invariant objects in phase space allows us to understand and predict the motion of the electron. In Chap. 5, we study the invariant objects structuring the phase space and driving the recollisions of the electron for d = 1, 2, 3.

One way to analyze and represent invariant objects in phase space is to use Poincaré sections. For d = 1, there are 1 + 1/2 degrees of freedom: One degree of freedom for the position and momentum of the electron (x, p_x) , and a half degree of freedom for time t. Therefore, the phase space is three-dimensional: One dimension for the position, one dimension for momentum and one dimension for time. Hamiltonian (1.34) is T-periodic, H(x, p, t + T) = H(x, p, t) for all x, p and t. As a consequence, the Hamiltonian flow $\varphi_0^t(x, p_x)$, which maps the point (x, p_x) at time t = 0 to the point $\varphi_0^t(x, p_x)$ at time t following Hamilton's equations, is also T-periodic. We consider the Poincaré map

$$\boldsymbol{\mathcal{P}}: (x, p_x) \mapsto \boldsymbol{\varphi}_0^T(x, p_x), \tag{1.35}$$

also referred to as the stroboscopic map. The Poincaré map (1.35) maps the phase-space variables (x, p_x) to a two-dimensional subspace through a stroboscopic plot of the dynamics. Therefore, the dynamics in phase space can be represented in two dimensions. The Poincaré section depicted in Fig. 1.7g is the stroboscopic plot obtained by applying multiple times the Poincaré map (1.35) on a set of initial conditions at time t = 0. The Poincaré section highlights the structure of the phase space. Under the Poincaré map (1.35), periodic orbits of period T, such as for instance the one depicted in Figs. 1.7b, 1.7d and 1.7f, become fixed points. For instance, around the origin, there is a fixed point corresponding to a periodic orbit of period T which is around the bottom of the ion-electron potential. Around the fixed point, we observe one-dimensional curves, which are *invariant tori* of the Poincaré map (1.35). The representation and the computation of invariant tori is performed in Sec. D.2.4.

In Figs. 1.7f, the T-trajectory populates the region close to the core, where we observe invariant curves. The motion of the T-trajectory is *quasi-periodic*, i.e., the electron almost comes back to its initial position in a given interval of time, as shown by the position of the triangles and the squares. As a consequence, the T-trajectory in light blue is bounded and cannot ionize. Notice that this is inconsistent with the quantum

theory of ionization, since the initial conditions correspond to ionized trajectories. This discrepancy can be overcome by including the Coulomb potential in the derivation of the quantum tunneling theory. For $I_p = 0.1$ a.u., the electron ionizes over the barrier for $I = 10^{14}$ W · cm⁻², and not by tunnel-ionization as it is assumed in Sec. 1.2.2. Still, we use the case $I_p = 0.1$ a.u. to highlight the role of the population of states of the T-trajectory by the quantum theory of ionization. The population of classical states by the quantum ionization plays an instrumental role, for instance, in the mechanism behind the bifurcation in the PMDs (see Fig. 8). This is studied in Chap. 3.

Further away from the origin, surrounding the last invariant torus, we observe a region of phase space in which there are no invariant curves, but a large density of points. In Fig. 1.7g, the pink trajectory populates this region. We observe that the T-trajectory is trapped close to the ionic core for a half of a laser cycle after ionization. Therefore, the region surrounding the last invariant curve is referred to as a *chaotic region*. Notice that the chaotic region is responsible for the resonant-excitation with subsequent ionization [113, 112]. The finite time Lyapunov exponent $\log |\lambda|/t$, where λ is the largest eigenvalue of the tangent flow (see Appendix C.1), measures the sensitivity with respect to the initial conditions by quantifying the degrees of exponential growth of the distance between two trajectories initiated close to each other. The finite time Lyapunov exponent of the pink trajectory, half of a laser cycle after ionization, is about 0.08. Therefore, in the chaotic region, two trajectories initiated close to each other leave quickly far apart each other. Usually, perturbative methods fail for predicting the dynamics of the electron in the chaotic region. Further away from the chaotic region, the density of points is smaller. We expect the dynamics of the T-trajectory populating this region to be accurately described by perturbative methods. In Chap. 2, we derive reduced models for the electron dynamics for arbitrary ellipticity and d = 1, 2, 3. The reduced models are used in Chap. 3 to highlight the impact of the Coulomb potential in the PMDs.

Therefore, despite the invariant objects driving the electron in phase space are identical in panels (b,d,f) of Fig. 1.7, the nature of the T-trajectory depends strongly on the state it populates after ionization. In Chap. 3, the states populated by the T-trajectory and its impact in photoelectron momentum distributions (PMDs) measured in experiments [93, 104] is studied for arbitrary ellipticities and d = 1, 2, 3.

1.3.2 Dynamics of the electron in higher dimensions

For arbitrary ellipticities, due to the form of the laser field given by Eq. (1.10) and Hamiltonian (1.14), the configuration space must be at least two dimensional (d > 1). For a *d*-dimensional configuration space, there are d + 1/2 degrees of freedom, and therefore the dimension of the phase space is 2d + 1. The dimension of the Poincaré section is therefore 2d, and cannot be represented in a straightforward way in two dimensions for d > 1. The dimension of the invariant objects is also larger, and therefore their analysis and their representation is computationally challenging. This makes the analysis of the phase space more delicate.

In Chap. 2, we derive a hierarchy of reduced models for describing the motion of the electrons in the framework of their guiding center (GC). The Hamiltonian of the guiding center is autonomous, and therefore it allows us to define an energy for the electron in the combined Coulomb and laser field. The energy of the guiding center is an adiabatic invariant of the dynamics of the electron. This energy is of particular interest for identifying the nature of the T-trajectory, as shown in Chap. 3. In addition, for d = 2, the number of degrees of freedom in the guiding center models is two, and their energy is conserved. Therefore, we can plot *Poincaré sections* of the Hamiltonian of the guiding center. It becomes possible to describe the dynamics of the electron with Poincaré sections as it has been done in Sec. 1.3.1 for the T-trajectory in Fig. 1.7.

In Chap. 4, we investigate the recollision mechanism in atoms driven by circularly polarized (CP) and highly elliptically polarized laser fields. The existence of recollisions in CP fields is still under debate [39, 61, 111, 58]. In particular, the three-step model, cartooned in Fig. 5, predicts that "For circularly polarized light, [...] the electron trajectory never returns to the vicinity of the ion.". However, experimental measurements [55, 61], supported by theoretical measurements [111, 81], show the existence of recollisions driven by CP fields for certain target species. For CP fields and f = 1, when mapping the phase-space coordinates of the electron in the laboratory frame to the rotating frame, in which the laser field is static, the laser interaction does no longer depend on time. As a consequence, the Hamiltonian is autonomous, and the dimension of the phase space is decreased to 2d. The rotating frame reveals a variety of invariant objects in phase space, such as for instance fixed points and periodic orbits. In particular, it reveals the existence of RPOs capable of driving the electron back to the core [81], depicted in red in Fig. 1.8c. These periodic orbits are located in high energy regions, and exist regardless the target species. Just as the electron is outside the core region, the initial phase-space coordinates of the T-trajectory are close to the energy of the blue dot depicted in Fig. 1.8c. However, in the rotating frame and for f = 1, the energy of the electron is constant in time,



Figure 1.8: (a)–(b) Calculation for LP fields and d = 1 reproduced from Ref. [80]. The wavelength of the laser is $\lambda = 780$ nm, the ion-electron interaction is a soft Coulomb potential (1.8). (b) Recolliding periodic orbits for (left panel) $I = 10^{14}$ W · cm⁻² and (right panel) $I = 10^{15}$ W · cm⁻². The red and black curves are the RPOs of Hamiltonian (1.34) and in the SFA, respectively. (a) The color code is the number of recollisions with initial condition $x_0 = 0$ in the plane of the momentum and the laser phase for $I = 10^{14}$ W · cm⁻². The black curves are the stable manifolds \mathcal{W}^s and the grey curves are the unstable manifolds \mathcal{W}^u of the fixed point associated with the periodic orbit under the Poincaré section x = 0 and dx/dt < 0. (c) RPOs for d = 2 and CP fields in the rotating frame reproduced from Ref. [81]. The grey surface is the zero-velocity surface [71] given by Eq. (4.10). The blue dot is the saddle point of the zero-velocity surface. The red curves are the RPOs.

and the T-trajectory cannot reach the high energy states. Hence, the rotating frame, besides decreasing the phase-space dimension, clearly illustrates the difficulty of obtaining recollisions in CP fields. In Chap. 4, we investigate the way the electron populates high energy states in atoms driven by CP fields and the conditions for which it recollides.

1.4 Conclusions

In sum, we have studied the dynamics of the electron in the quantum framework. We have highlighted the classical features of the wave function using, among others, the Wigner quasi-probability distribution. In particular, we have shown that after ionization, the dynamics of the electron is mainly classical. We have introduced a two-step model in which the ionization of the electron is treated quantum mechanically, and its subsequent dynamics is treated classically. First, treating the dynamics of the electrons after ionization classically, instead of quantum mechanically, allows us to describe the underlying mechanisms in terms of electron trajectories. Trajectories, which are local paths in space, allows us to visualize and interpret more clearly the motion and the behaviors of electrons in time. Second, the two-step model allows us to distinguish phenomena due to both the population of initial conditions by quantum ionization and the subsequent classical motion, and phenomena solely due to the subsequent classical motion after ionization. Indeed, in the classical description of the dynamics, the initial conditions of the electron is of prime importance, and phenomena, such as for instance the bifurcation in the PMDs (studied in Chap. 3) or the absence of recollisions in CP fields for specific target species (studied in Chap. 4), are due to both the population of initial conditions by quantum ionization and the subsequent classical motion of the electron. In contrast, phenomena, such as for instance the Rydberg states creations and Coulomb-driven recollisions (also studied in Chap. 3), are mainly due to the subsequent classical motion of the electron after ionization.

CHAPTER 1. ATOMS SUBJECTED TO INTENSE LASER FIELDS

In the classical framework, the dynamics of the electron is governed by Hamiltonian (1.14). In this manuscript, two main methods are used for studying Hamiltonian (1.14). In Chap. 2, we use *perturbative methods* to determine reduced models for describing the motion of the electron in phase space. In Chap. 5, we use *non perturbative methods* to analyze the dynamics of the electron in phase space. In particular, we identify invariant objects which structure phase space and drive recollisions.

Chapter 2

Reduced dynamics for atoms subjected to intense and elliptically polarized laser pulses: The guiding center model

The laser-atom interaction gives rise to complex phenomena, involving multiple temporal and spatial scales. The phenomena arising from short time-scale and long time-scale processes manifest themselves in different aspects of the measurements, for different values of the parameters, and also at distinct times along the same trajectory. The multiple temporal and spatial scales arise from the competition between the strong laser and Coulomb forces. The co-existence of short vs long time-scale processes in Hamiltonian (1.14) is illustrated in the upper panel of Fig. 2.1, which shows the excursion time per laser cycle $\Delta t/T$ of the electron as a function of the PPT initial conditions given by Eqs. (1.25) with $p_{\parallel} = p_{z,0} = 0$. The excursion time Δt is the time when the electron returns to the core, i.e., the smallest positive time such that $|\mathbf{r}(t + \Delta t)| = R$, with R = 5 a.u. For Hamiltonian (1.14), we observe the four main features of the electron is trapped in a Rydberg state (grey regions), the electron returns to the core on a subcycle time scale (light purple region for which $\Delta t/T \leq 1$) and the electron returns to the core after multiple laser cycles (colored layers in the grey regions for which $\Delta t/T > 1$).

For instance, the red trajectory in Fig. 1.4 (case d = 1) is a typical trajectory which returns to the core on a subcycle time scale (for which $\Delta t/T \leq 1$). In this case, the electron leaves relatively far from the core, and comes back mainly due to the oscillations of the laser field. This is referred to as a *subcycle recollision*. During the excursion of the electron outside the core, the laser field is dominant, and the Coulomb potential acts perturbatively compared to the laser interaction on short time scales. In Sec. 2.1, we derive the strong field approximation (SFA) and the Coulomb-corrected strong field approximation (CCSFA) based on the treatment of the Coulomb potential as a perturbation of the laser interaction. The middle panels of Fig. 2.1 show the excursion time of the electron $\Delta t/T$ in the SFA and CCSFA. We observe that the region with subcycle recollisions (for which $\Delta t/T \leq 1$) is well described by the SFA and the CCSFA. In these regions, hard recollisions [84] (the electron returns on the core) and soft recollisions [82] (the electron returns in the vicinity of the core) coexist, depending on the initial conditions.

The pink trajectory in Fig. 1.4 (case d = 1) and the trajectories in Fig. 2.2a, Fig. 2.2c and Fig. 2.2f are typical trajectories which return to the core after multiple laser cycles (for which $\Delta t/T > 1$). In this case, the electron displays fast oscillations due to its interaction with the laser, and its drift momentum changes slowly in time due to its interaction with the ion. Then, the electron returns to the core. This is referred to as a *Coulomb-driven recollision*. However, we observe on the middle panels of Fig. 2.1 that the long time-scale processes, such as the Rydberg state creation and the return of the electron after multiple laser cycles, which are the processes the most probable according to the PPT ionization rate as seen in Fig. 2.1, are not captured by the SFA and the CCSFA.

Questions

- How can we analyze the motion of the electron in the combined Coulomb and laser fields on long time scales ?
- Can we distinguish short- and long time-scale processes ?



Figure 2.1: Excursion time of the electron Δt per laser cycle T as a function of the initial conditions $(t_0, p_\perp, p_{\parallel} = p_{z,0} = 0)$ for $I = 8 \times 10^{13}$ W \cdot cm⁻², Z = 1, a = 1 and $\xi = 0.4$ of the reference Hamiltonian (1.14) with the soft Coulomb potential, the SFA [39], the CCSFA [62] and the GC model $G_2 = (H_2, \Phi_2)$. The time Δt is the smallest positive time such that $|\mathbf{r}(t + \Delta t)| = 5$ a.u. The grey regions are where the electron final energy is negative after the end of the pulse (at t = 10T). The white regions are where the electron undergoes a direct ionization, i.e., an ionization without returning to the core. The black dashed lines are contours of constant ionization rate $W_{\rm PPT}(t_0, p_\perp \mathbf{e}_\perp(t_0))$ for $W_{\rm PPT}/\max(W_{\rm PPT}) = 10^{-1}$, 10^{-5} and 10^{-15} , from bottom to top. The momentum p_\perp is scaled by E_0/ω .

Plan Reduced dynamics for atoms subjected to intense and elliptically polarized laser pulses depends on the type of trajectories and phenomena to assess. Figure 2.1 reflects the complementarity of the CCSFA and the GC models on short vs long time-scale processes. In this chapter, we derive reduced models to describe the motion of the electron after ionization. In Sec. (2.1), we recall the two models used in the community for the description of the fast time-scale dynamics of the electron, namely the SFA and the CCSFA. In Sec. 2.2, we describe the procedure we use for deriving the hierarchy of reduced models for the GC dynamics in strong laser fields based on averaging over the fast motion of the electron using Lie transforms. In Sec. 2.3, we first compare these reduced models with the dynamics associated with Hamiltonian (1.14). The comparison between these models highlights the models which are best suited for a qualitative and quantitative agreement with the parent dynamics. In particular, we show the relevance of two models in the hierarchy. The model G₂ (second order in the hierarchy of models) provides the leading behavior of the trajectories and is the most robust close to the core. The model G₅ (fifth order of the hierarchy of models) improves the quantitative agreement with trajectories relatively far from the core. Then we analyze the dynamics of the GC models in phase space, highlighting regular and chaotic regions and their relation with the trajectories of Hamiltonian (1.14).

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2.1 The Coulomb potential seen as a perturbation of the laser interaction: The SFA and the CCSFA

The reference Hamiltonian is defined in Eq. (1.14). Figure 2.1 shows that the dynamics of the electron is particularly well described by the SFA and the CCSFA for trajectories which recollide early after ionization. This suggests that such approximations are well suited for describing the motion of the electron on short time scales. We consider the Coulomb potential as a perturbation of the laser interaction. Under this assumption, we write Hamiltonian (1.14) as

$$H(\mathbf{r}, \mathbf{p}, t) = \frac{|\mathbf{p}|^2}{2} + \mathbf{r} \cdot \mathbf{E}(t) + \epsilon V(\mathbf{r}),$$

where we have introduced an ordering parameter ϵ for bookkeeping purposes. The equations of motion of the electron are

$$\dot{\mathbf{r}} = \mathbf{p},$$

 $\dot{\mathbf{p}} = -\mathbf{E}(t) - \epsilon \frac{\partial V(\mathbf{r})}{\partial \mathbf{r}}$

We consider the correction due to the Coulomb interaction on short time scales, hence $\mathbf{r} = \mathbf{r}^{\text{SFA}} + \epsilon \Delta \mathbf{r} + O(\epsilon^2)$ and $\mathbf{p} = \mathbf{p}^{\text{SFA}} + \epsilon \Delta \mathbf{p} + O(\epsilon^2)$. We substitute these equations in the equations of motion (2.1) and we identify each term by order of ϵ .

2.1.1 Strong field approximation (SFA)

The lowest order in ϵ provides the SFA electron phase-space trajectory

$$\mathbf{r}^{\text{SFA}}(t) = \mathbf{r}_0 + [\mathbf{p}_0 - \mathbf{A}(t_0)] (t - t_0) + [\mathbf{\Sigma}(t) - \mathbf{\Sigma}(t_0)] / \omega^2, \qquad (2.2a)$$

$$\mathbf{p}^{\text{SFA}}(t) = \mathbf{p}_0 - \mathbf{A}(t_0) + \mathbf{A}(t), \qquad (2.2b)$$

where $\omega^2 \mathbf{A}(t) = \partial \boldsymbol{\Sigma} / \partial t$. The initial conditions are $\mathbf{r}(t_0) = \mathbf{r}_0$ and $\mathbf{p}(t_0) = \mathbf{p}_0$.

In the SFA, the motion of the electron is composed of a drift momentum $\mathbf{p}^{\text{SFA}}(t) - \mathbf{A}(t) = \mathbf{p}_0 - \mathbf{A}(t_0)$ which is conserved in time. When the laser field is turned off, the vector potential is zero and the momentum of the electron is the initial drift momentum of the electron, $\mathbf{p} = \mathbf{p}_0 - \mathbf{A}(t_0)$. We consider the laser field given by Eq. (1.10) with f(0) = 1 and $\varphi_{\text{CEP}} = 0$. According to the ADK theory [see Sec. 1.2.2.1], the electron ionizes most probably at the peak amplitude of the laser field $(t_0 = 0)$ and with zero initial momentum $\mathbf{p}_0 = \mathbf{0}$. As a consequence, for LP fields $(\xi = 0)$, the most probable final energy of the electron is $|\mathbf{p}|^2/2 = 0$. For CP fields $(\xi = 1)$, the most probable final energy of the electron is $|\mathbf{p}|^2/2 = 2U_p/(\xi^2 + 1)$. Therefore, the distributions of the energy of the ionized electrons subjected to LP ($\xi = 0$) and CP ($\xi = 1$) fields is 0 and $U_p/(\xi^2 + 1)$, respectively, in agreement with experimental measurements [40]. Revealing the mechanism behind the differences between the distributions of the energy of the ionized electrons subjected to LP and CP fields was the first success of the SFA.

Later, in Ref. [39], the classical motion of the electron in the SFA succeeded in describing the mechanics of the recollision. In the SFA, in addition to the drift momentum of the electron $\mathbf{p}^{\text{SFA}}(t) - \mathbf{A}(t) = \mathbf{p}_0 - \mathbf{A}(t_0)$ which is conserved in time, the motion of the electron is also composed of fast oscillations $\mathbf{\Sigma}(t)/\omega^2$ of amplitude E_0/ω^2 . The electron leaves far from the core, then returns to the core when the laser field changes direction and recollides. The maximum return kinetic energy of the recolliding electron, $3.17U_p$, explained the HHG cutoff observed in experiments [102, 39]. In addition, the trajectories and the return time of the electron in the SFA were successful to predict the time-frequency profile of the HHG spectrum [147]. The trajectories which contribute the most to HHG recollide in a subcycle after ionization. Therefore, physics of HHG is built on short-time scale processes which are well described by the SFA.

In Chap. 3, the SFA is used to highlight the contributions of the Coulomb potential in ATI experiments. In Chap. 4, the SFA is used to describe the conditions for which the electron returns and gains energy in high elliptically polarized laser fields.

2.1.2 Coulomb-corrected strong field approximation (CCSFA)

The first order in ϵ provides the correction due to the Coulomb interaction on the SFA trajectory, which reads

$$\Delta \mathbf{r}(t) = \int_{t_0}^t \Delta \mathbf{p}(s) \, \mathrm{d}s, \qquad (2.3a)$$

$$\Delta \mathbf{p}(t) = -\int_{t_0}^t \frac{\partial V\left(\mathbf{r}^{\text{SFA}}(s)\right)}{\partial \mathbf{r}} \, \mathrm{d}s.$$
(2.3b)

The CCSFA is also used in a semiclassical framework in Refs. [136, 143, 145, 144] and in a classical framework in Refs. [62, 82, 93, 80, 84, 85].

The CCSFA in the purely classical approach was successful to describe the asymmetry with respect the minor polarization axis in the distribution of the momentum of the ionized electrons [62] (see Figs. 8). A good agreement is observed between the predictions of this model and the experimental measurements for relatively large ellipticity. In Ref. [93], we observe a disagreement between the predictions of the CCSFA and the experimental measurements. As mentioned above, the CCSFA is valid to determine the correction of the Coulomb interaction for short times (e.g., $t - t_0 \sim T$) regardless of the ellipticity. Looking at Eqs. (2.2a), if the initial drift momentum of the electron $\mathbf{p}_0 - \mathbf{A}(t_0)$ is sufficiently large, the Coulomb correction (2.3b) is significant only for a short time after ionization. According to the PPT theory, the initial drift momentum is of order $|\mathbf{p}_0 - \mathbf{A}(t_0)| \sim \xi E_0/\omega$, hence, we expect the CCSFA to be valid in ATI only for large ellipticity.

2.2 Derivation of the hierarchy of models for the guiding center (GC)

Figure 2.2 displays six typical trajectories of Hamiltonian (1.14) involving long time-scale dynamics. We notice that these trajectories display fast oscillations around a mean trajectory, which we call the GC trajectory. What is the dynamics of these GC trajectories? We use this clear separation of scales to derive models for the GC dynamics. Specifically, we average Hamiltonian (1.14) over the fast motion using canonical transforms in order to simplify the electron dynamics and clearly distinguish the different ionization channels for the electron. As a consequence, we derive a hierarchy of averaged models which fully take into account the Coulomb potential. In these models, the electron in the combined strong laser and Coulomb fields follows a GC trajectory. Actually, there are several possible GCs, depending on the order up to which the averaging is performed.

2.2.1 Canonical Lie transforms

In this section, we recall some basic features on canonical transformations in the framework of *canonical Lie* transforms [33, 34]. We consider a Hamiltonian system with phase-space variables \mathbf{z} , a Hamiltonian $H(\mathbf{z})$ and a Poisson bracket $\{\cdot, \cdot\}$ (either a canonical or non-canonical bracket. See Sec. C.2 for details). Canonical

Lie transforms are near-identity change of coordinates $\mathbf{z} \mapsto \bar{\mathbf{z}}(\mathbf{z})$, generated by a scalar function $S(\mathbf{z})$, called the generating function, and given by

$$\bar{\mathbf{z}} = \exp(-\mathcal{L}_S) \, \mathbf{z} = \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} \mathcal{L}_S^k \mathbf{z},$$

$$= \mathbf{z} - \{S, \mathbf{z}\} + \frac{1}{2} \{S, \{S, \mathbf{z}\}\} + \dots,$$
(2.4)

where \mathcal{L}_S is the Liouville operator defined by $\mathcal{L}_S F = \{S, F\}$. Canonical Lie transforms have several properties:



Figure 2.2: Typical electron trajectories for $I = 10^{14}$ W \cdot cm⁻², d = 2, Z = 1, a = 1 and $\omega = 0.05$ a.u. in the polarization plane (x, y). The ellipticities are: (a) $\xi = 0$, (b–e) $\xi = 0.5$, (f) $\xi = 1$. The dark blue curve is the electron trajectory of Hamiltonian (1.14) with the soft Coulomb potential. The cyan and red curves are the trajectories of the models G₂ and G₅, respectively, with initial conditions far from the ionic core. For each model, the solid and dashed curves are the GC and the approximate trajectories, respectively. The lightly shaded circle is the position of the ionic core at the origin, and the black circle surrounding the origin is $|\mathbf{r}| = 15$ a.u. All quantities are in atomic units.

- $F(\exp(\mathcal{L}_S)\mathbf{z}) = \exp(\mathcal{L}_S)F(\mathbf{z})$, which comes from the Leibniz rule,
- $\{\exp(\mathcal{L}_S)F, \exp(\mathcal{L}_S)G\} = \exp(\mathcal{L}_S)\{F, G\}$, which comes from the Jacobi identity and the antisymmetry of the Poisson bracket,

for any scalar functions $F(\mathbf{z})$ and $G(\mathbf{z})$. As a consequence of the first Property and of the scalar invariance $\overline{F}(\overline{\mathbf{z}}) = F(\mathbf{z})$, these changes of variables modify any observable $F(\mathbf{z})$, for instance the Hamiltonian $H(\mathbf{z})$, into

$$\overline{F}(\mathbf{z}) = \exp\left(\mathcal{L}_S\right) F(\mathbf{z}).$$
 (2.5)

In particular, the generating function is an observable, and $\bar{S}(\bar{\mathbf{z}}) = S(\mathbf{z})$. Using Eq. (2.5), one obtains $\bar{S}(\mathbf{z}) = \exp(\mathcal{L}_S)S(\mathbf{z})$. Since $\exp(\mathcal{L}_S)S(\mathbf{z}) = S(\mathbf{z})$, one has $\bar{S}(\mathbf{z}) = S(\mathbf{z})$, i.e., the generating function does not change form with respect to the canonical Lie transforms. The second Property ensures that these changes of coordinates do not affect the expression of the Poisson bracket, i.e., they are canonical transformations. One significant advantage of these transformations is that they are explicit functions and they can be easily inverted: $\mathbf{z} = \exp(\mathcal{L}_S)\bar{\mathbf{z}}$. This way, we can recover all the information on the particle dynamics from the transformed (averaged) system.

These canonical Lie transforms are particularly well suited for perturbation theory. If the Hamiltonian is of the form $H = H_0 + \epsilon W$, where H_0 is the Hamiltonian of the unperturbed system, W is the perturbation and ϵ is an ordering (small) parameter, a canonical Lie transform generated by a generating function $S(\bar{z})$ (of order ϵ), applied to H is able to remove the unwanted part of the perturbation, called \widetilde{W} , and move its influence to higher orders in ϵ . More explicitly, at the lowest order, the expression of the new Hamiltonian expressed in the new variables \bar{z} is

$$\bar{H} = \exp(\mathcal{L}_S) H$$

$$= H_0 + \epsilon W + \{S, H_0\} + \epsilon \{S, W\} + \frac{1}{2} \{S, \{S, H_0\}\} + \dots$$
(2.6)

Choosing appropriately the generating function S such that $\{S, H_0\} = -\epsilon \widetilde{W}$, unwanted terms in the perturbation W can be pushed from order ϵ to order ϵ^2 , meaning that the order ϵ in the Hamiltonian becomes $W - \widetilde{W}$. For instance, one can suppress fast oscillating terms contained in W. Then, the associated canonical change of coordinates is determined using Eq. (2.4). The advantage of canonical Lie transforms vs other types of canonical transforms is that they are easily manipulable algebraically, which is well suited, for instance, for the numerical implementation of iterative procedures.

2.2.2 Averaging the electron dynamics

We notice that these transformations are defined for autonomous systems. Hamiltonian (1.14) has an explicit time dependence through the electric field. Therefore, we first increase phase space to include time t, and consider its canonically conjugate variable k. The extended Hamiltonian (1.14) becomes

$$H(\mathbf{r}, \mathbf{p}, t, k) = k + \frac{|\mathbf{p}|^2}{2} + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t).$$
(2.7)

The extended Poisson bracket is

$$\{F,G\} = \frac{\partial F}{\partial \mathbf{r}} \cdot \frac{\partial G}{\partial \mathbf{p}} - \frac{\partial F}{\partial \mathbf{p}} \cdot \frac{\partial G}{\partial \mathbf{r}} + \frac{\partial F}{\partial t} \frac{\partial G}{\partial k} - \frac{\partial F}{\partial k} \frac{\partial G}{\partial t}, \qquad (2.8)$$

where the operators $\partial/\partial \mathbf{r} = (\partial/\partial x, \partial/\partial y, \partial/\partial z)$ and $\partial/\partial \mathbf{p} = (\partial/\partial p_x, \partial/\partial p_y, \partial/\partial p_z)$. The hypothesis we make for the derivation of our hierarchy of reduced models is that the characteristic time of the ionized electron trajectory is large compared to a laser cycle $T = 2\pi/\omega$, i.e., $\omega \mapsto \omega/\epsilon$ where ϵ is an ordering parameter which is explicitly introduced for bookkeeping purposes. Performing the canonical change of coordinates $\bar{t} = t/\epsilon$ and $\bar{k} = \epsilon k$, and re-scaling the energy, Hamiltonian (2.7) becomes

$$H^{(0)}(\mathbf{r}, \mathbf{p}, t, k; \epsilon) = k + \epsilon \left[\frac{|\mathbf{p}|^2}{2} + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t) \right],$$
(2.9)

where we have removed the bars in the new variables. We apply canonical Lie transforms as described above in order to perform the averaging of Hamiltonian (2.9) over the fast time scale, by pushing time-dependent terms in the Hamiltonian to higher order terms in ϵ .

2.2.2.1 Gauge velocity transformation

As an example, we consider the transformation from the length gauge to the velocity gauge [135] in Hamiltonian (2.9). It is given by the following change of coordinates

$$\begin{aligned} \mathbf{\dot{r}} &= \mathbf{r}, \\ \bar{\mathbf{p}} &= \mathbf{p} - \epsilon \mathbf{A}(t), \end{aligned}$$

where $\mathbf{A}(t)$ is the vector potential defined by $\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$. This transformation is a canonical change of coordinates which can be formulated as a canonical Lie transform generated by

$$S^{(1)} = \epsilon \,\mathbf{r} \cdot \mathbf{A}(t). \tag{2.10}$$

The Hamiltonian in the velocity-gauge coordinates becomes

$$H^{(1)} = \exp(\mathcal{L}_{S^{(1)}})H^{(0)}$$

$$= \bar{k} + \epsilon \left[\frac{1}{2}\left(\bar{\mathbf{p}} + \epsilon \mathbf{A}(t)\right)^2 + V(\bar{\mathbf{r}})\right] = \bar{k} + \epsilon \left[\frac{|\bar{\mathbf{p}}|^2}{2} + V(\bar{\mathbf{r}})\right] + \epsilon^2 \bar{\mathbf{p}} \cdot \mathbf{A}(t) + \epsilon^3 \frac{\mathbf{A}^2(t)}{2}.$$
(2.11)

We observe that the time-dependence in Hamiltonian (2.9), present in $\mathbf{E}(t)$, is of order ϵ , while in Hamiltonian (2.11) this time-dependence is moved to order ϵ^2 .

2.2.2.2 Iterative procedure

We iterate the above-procedure to higher order in ϵ . We assume that after the *N*-th step of the procedure, all the time-dependent terms in the present averaged Hamiltonian are removed up to order ϵ^N , that is, the time-dependence of the averaged Hamiltonian is of order ϵ^{N+1} . We assume that the total generating function up to order ϵ^N is known. The total generating function at this step is

$$S^{(N)}(\mathbf{r}, \mathbf{p}, t; \epsilon) = \sum_{n=1}^{N} \epsilon^n S_n(\mathbf{r}, \mathbf{p}, t), \qquad (2.12)$$

and the corresponding averaged Hamiltonian is denoted

$$H^{(N)} = \exp(\mathcal{L}_{S^{(N)}})H^{(0)}$$

= $k + \sum_{n=1}^{N} \epsilon^n h_n(\mathbf{r}, \mathbf{p}) + \epsilon^{N+1} R_{N+1}(\mathbf{r}, \mathbf{p}, t; \epsilon),$

where $h_n(\mathbf{r}, \mathbf{p})$ are the coefficients in the series expansion of the Hamiltonian that no longer depends on time, while $R_{N+1}(\mathbf{r}, \mathbf{p}, t; \epsilon)$ is the remainder of the Hamiltonian which still depends on time. The objective of the iterative method is to find the modified generating function $S^{(N+1)}$ [which amounts to finding the extra function S_{N+1} in Eq. (2.12)] to remove the time-dependence in the term R_{N+1} at the lowest order.

The averaged Hamiltonian $H^{(N+1)}$ whose time-dependence is of order ϵ^{N+2} is

$$\begin{aligned} H^{(N+1)} &= \exp(\mathcal{L}_{S^{(N+1)}})H^{(0)}, \\ &= \exp(\epsilon^{N+1}\mathcal{L}_{S_{N+1}})\exp(\mathcal{L}_{S^{(N)}})H^{(0)} + O(\epsilon^{N+2}), \\ &= \exp(\epsilon^{N+1}\mathcal{L}_{S_{N+1}})H^{(N)} + O(\epsilon^{N+2}), \\ &= H^{(N)} + \epsilon^{N+1}\left(R_{N+1} + \frac{\partial S_{N+1}}{\partial t}\right) + O(\epsilon^{N+2}). \end{aligned}$$

The time-fluctuating terms in R_{N+1} are denoted $\overbrace{R_{N+1}}^{N+1}$, and are defined by

$$\widetilde{R_{N+1}} = R_{N+1} - \frac{1}{T} \int_0^T \mathrm{d}t \; R_{N+1}.$$
(2.13)

In order to eliminate the time-fluctuating terms at order ϵ^{N+1} , the component S_{N+1} of the generating function $S^{(N+1)}$ is chosen as

$$S_{N+1} = -\int \mathrm{d}t \ \widetilde{R_{N+1}(\mathbf{r}, \mathbf{p}, t; 0)},\tag{2.14}$$

where the primitive is chosen such that the mean value of S_{N+1} with respect to $t \in [0, T]$ is zero. At each step the functions R_n have to be computed up to order M where M is the last order for which the averaged Hamiltonian will be computed analytically. We perform these computations using a symbolic computation software.

2.2.2.3 Averaged Hamiltonians

We apply the above-described procedure to push the time-dependence in Hamiltonian (2.9) to order ϵ^8 using $H^{(7)} = \exp(\mathcal{L}_{S^{(7)}})H^{(0)}$. Below we provide the explicit expression for $S^{(6)}$. The higher-order components are too lengthy to report and their expressions are not particularly enlightening.

$$S^{(6)} = \epsilon \,\mathbf{r} \cdot \mathbf{A}(t) - \frac{\epsilon^2}{\omega^2} \,\mathbf{p} \cdot \mathbf{E}(t) + \frac{\epsilon^3}{\omega^2} \mathbf{A}(t) \cdot \left(\frac{\mathbf{E}(t)}{4} + \frac{\partial V}{\partial \mathbf{r}}\right) - \frac{\epsilon^4}{\omega^4} \mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{E}(t) \cdot \frac{\partial V}{\partial \mathbf{r}}\right) + \frac{\epsilon^5}{\omega^4} \left[\left(\frac{\mathbf{E}(t)}{4} + \frac{\partial V}{\partial \mathbf{r}}\right) \cdot \frac{\partial}{\partial \mathbf{r}} - \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}}\right)^2 \right] \left(\mathbf{A}(t) \cdot \frac{\partial V}{\partial \mathbf{r}}\right) - \frac{5\epsilon^6}{8\omega^6} \mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}}\right)^2 V - \frac{\epsilon^6}{\omega^6} \left[\left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}}\right) \left(\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}}\right) - \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}}\right)^3 + 2\left(\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}}\right) \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}}\right) \right] \left(\mathbf{E}(t) \cdot \frac{\partial V}{\partial \mathbf{r}}\right). \quad (2.15)$$

Here, we have used the fact that the electric field is monochromatic and satisfies $\omega^2 \mathbf{E}(t) = -\partial^2 \mathbf{E}/\partial t^2$. The averaged Hamiltonian $H^{(7)}$ is

$$H^{(7)} = \bar{k} + \epsilon \left[\frac{|\bar{\mathbf{p}}|^2}{2} + V(\bar{\mathbf{r}}) \right] + \epsilon^3 U_p + \epsilon^5 \frac{U_p}{\omega^2(\xi^2 + 1)} \left(\frac{\partial^2 V}{\partial x^2} + \xi^2 \frac{\partial^2 V}{\partial y^2} \right) + \epsilon^7 \frac{U_p}{\omega^4(\xi^2 + 1)} \left[\left| \frac{\partial}{\partial \mathbf{r}} \left(\frac{\partial V}{\partial x} \right) \right|^2 + \xi^2 \left| \frac{\partial}{\partial \mathbf{r}} \left(\frac{\partial V}{\partial y} \right) \right|^2 \right] + O(\epsilon^8),$$
(2.16)

where all the derivatives are evaluated at $\bar{\mathbf{r}}$. By truncating the Hamiltonian at a given order, we notice that the reduced (time-independent) Hamiltonians up to order ϵ^7 are of the form

$$H(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \frac{|\bar{\mathbf{p}}|^2}{2} + V_{\text{eff}}(\bar{\mathbf{r}}),$$

with an effective potential V_{eff} . In particular, this highlights a particular property in the reduction process that the reduction procedure does not generate $\bar{\mathbf{p}}$ -dependent terms in the Hamiltonian other than the kinetic energy, up to order ϵ^7 . At order ϵ^8 , the term which is generated in $H^{(8)}$ is linear in the momenta; therefore, it can easily be eliminated by a translation in $\bar{\mathbf{p}}$ (which is a canonical transformation). At order ϵ^9 , the terms which are generated are quadratic in the momenta $\bar{\mathbf{p}}$, and we do not consider these terms in what follows. Therefore, our analysis is valid up to order ϵ^8 . In such a way, it is particularly convenient to define effective potentials. Depending on the order of truncation, we obtain three reduced Hamiltonians

$$H_2(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \frac{|\bar{\mathbf{p}}|^2}{2} + V(\bar{\mathbf{r}}), \qquad (2.17a)$$

$$H_5(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \frac{|\bar{\mathbf{p}}|^2}{2} + V(\bar{\mathbf{r}}) + \frac{U_p}{\omega^2(\xi^2 + 1)} \left(\frac{\partial^2 V}{\partial x^2} + \xi^2 \frac{\partial^2 V}{\partial y^2}\right), \qquad (2.17b)$$

$$H_{7}(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \frac{|\bar{\mathbf{p}}|^{2}}{2} + V(\bar{\mathbf{r}}) + \frac{U_{p}}{\omega^{2}(\xi^{2}+1)} \left(\frac{\partial^{2}V}{\partial x^{2}} + \xi^{2}\frac{\partial^{2}V}{\partial y^{2}}\right) \\ + \frac{U_{p}}{\omega^{4}(\xi^{2}+1)} \left[\left|\frac{\partial}{\partial \mathbf{r}}\left(\frac{\partial V}{\partial x}\right)\right|^{2} + \xi^{2}\left|\frac{\partial}{\partial \mathbf{r}}\left(\frac{\partial V}{\partial y}\right)\right|^{2}\right], \qquad (2.17c)$$

where we have removed the small parameter ϵ which was originally introduced for bookkeeping purposes. Each of these Hamiltonians describes the dynamics of the GC at a different level of approximation. As a result of averaging, the Hamiltonians (2.17) are conserved, in contrast to Hamiltonian (1.14). We notice that the effective potentials depend on the main parameters of the electric field, its amplitude and its ellipticity, with the exception of H_2 .

The corresponding change of coordinates which maps $H^{(0)}(\mathbf{r}, \mathbf{p}, t, k; \epsilon)$ to $H^{(7)}$ is given by Eq. (2.4) and

	Φ_2	Φ_3	Φ_4	Φ_5	Φ_6	Φ_7
H_2	G_2	G_3	G_4	0	0	0
H_5	0	0	0	G_5	G_6	0
H_7	0	0	0	0	0	G_7

Table 2.1: The GC models $G_n = (H_m, \Phi_n)$, where $n \ge m$. The Hamiltonians H_m are given by Eqs. (2.17), and the mapping from the electron to the GC coordinates are given by Eqs. (2.18).

its series expansion up to order ϵ^6 is given by

$$\begin{split} \bar{\mathbf{r}} &= \mathbf{r} - \frac{\epsilon^2}{\omega^2} \mathbf{E}(t) - \frac{\epsilon^4}{\omega^4} \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V - \frac{2\epsilon^5}{\omega^4} \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\mathbf{A}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V \\ &- \frac{\epsilon^6}{8\omega^6} \frac{\partial}{\partial \mathbf{r}} \left(\overline{\mathbf{E}}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right)^2 V - \frac{\epsilon^6}{\omega^6} \frac{\partial}{\partial \mathbf{r}} \left[\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}} - 3 \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right)^2 \right] \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V \\ &- \frac{2\epsilon^6}{\omega^6} \left(\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) \frac{\partial V}{\partial \mathbf{r}} + O(\epsilon^7), \end{split}$$
(2.18a)
$$\bar{\mathbf{p}} &= \mathbf{p} - \epsilon \mathbf{A}(t) - \frac{\epsilon^3}{\omega^2} \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{A}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V + \frac{\epsilon^4}{\omega^4} \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right)^2 \frac{\partial V}{\partial \mathbf{r}} + O(\epsilon^7) \\ &- \frac{\epsilon^5}{\omega^4} \frac{\partial}{\partial \mathbf{r}} \left[\left(\frac{\mathbf{E}(t)}{4} + \frac{\partial V}{\partial \mathbf{r}} \right) \cdot \frac{\partial}{\partial \mathbf{r}} - \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right)^2 \right] \left(\mathbf{A}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V + \frac{\epsilon^6}{8\omega^6} \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\overline{\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}}} \right)^2 \frac{\partial V}{\partial \mathbf{r}} \\ &+ \frac{\epsilon^6}{\omega^6} \frac{\partial}{\partial \mathbf{r}} \left[\left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}} \right) - \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right)^3 + 2 \left(\frac{\partial V}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \right] \left(\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V \\ &+ O(\epsilon^7). \end{aligned}$$
(2.18b)

If one needs to know the averaged Hamiltonian and/or the system of coordinates at the order ϵ^N , one needs to truncate the $O(\epsilon^{N+1})$ terms of Eqs. (2.16) and/or (2.18), respectively. The expressions (2.18) are used below to reconstruct the trajectories of the electrons from the trajectories of the GCs.

Result 1: Hierarchy of guiding-center (GC) models

The hierarchy of models for the GC is composed of a time-independent Hamiltonian H_m [see Eqs. (2.17)] and a transformation from the electron coordinates to the GC coordinates:

$$\Phi_n: (\mathbf{r}, \mathbf{p}) \mapsto (\bar{\mathbf{r}}, \bar{\mathbf{p}}), \tag{2.19}$$

whose truncated expressions are given by the truncations of Eqs. (2.18). The orders m and n refer to the order of the Hamiltonian and the transformation, respectively, after truncation of the perturbative expansion. We refer to the model

$$\mathbf{G}_n = (H_m, \Phi_n)$$

as the *n*-th order GC model, where $n \ge m$, and we show the set of (m, n) in Table 2.1.

2.2.2.4 Links with the Kramers-Henneberger potential

At order ϵ^5 , our model is linked to the Kramers-Henneberger (KH) treatment of the motion of a charged particle in an external time-periodic electric field [42, 161, 120]. In a nutshell, the classical KH theory amounts

CHAPTER 2. GUIDING CENTER MODEL

to performing a canonical Lie transform generated by

$$S^{(2)} = \epsilon \, \mathbf{r} \cdot \mathbf{A}(t) - \frac{\epsilon^2}{\omega^2} \, \mathbf{p} \cdot \mathbf{E}(t),$$

on Hamiltonian (2.9), where the first term is used for moving into the velocity gauge. The corresponding change of coordinates is given by $\bar{\mathbf{p}} = \mathbf{p} - \epsilon \mathbf{A}(t)$ and $\bar{\mathbf{r}} = \mathbf{r} - \epsilon^2 \mathbf{E}(t)$. The resulting Hamiltonian becomes exactly

$$H_{\rm KH} = \bar{k} + \epsilon \left[\frac{|\bar{\mathbf{p}}|^2}{2} + V \left(\bar{\mathbf{r}} + \frac{\epsilon^2}{\omega^2} \mathbf{E}(t) \right) \right] + \epsilon^3 \frac{\mathbf{E}^2(t)}{2\omega^2}.$$

The term of order ϵ^3 in the Hamiltonian $H_{\rm KH}$ can be easily removed by performing an additional transformation. In KH theory, the remaining time-dependence in the potential is removed by an integral over time of the effective potential. For instance, if we expand the KH effective potential up to order ϵ^7 and we average over $t \in [0, T]$, it becomes

$$V_{\rm KH} = V + \epsilon^4 \frac{U_p}{\omega^2(\xi^2 + 1)} \left(\frac{\partial^2 V}{\partial x^2} + \xi^2 \frac{\partial^2 V}{\partial y^2}\right) + O(\epsilon^8).$$

In this framework, the effective potential always only depends on the position variables (and not on the momenta), and there is no contribution at order ϵ^6 , contrary to our derivations. The origin of this discrepancy is that performing an averaging using an integral over the fast timescales is only correct to the lowest order (here ϵ^4), but it fails at higher orders. In particular, it does not include additional drift velocities at order ϵ^7 (as opposed to H_7 which includes an additional term of order ϵ^7), which drive the charged particles at longer time scales. In addition, the reconstructed trajectories do not include corrections on orders higher than ϵ^2 . One needs to perform canonical changes of coordinates to properly average the fast motions. Expressions beyond order ϵ^4 and results obtained using these higher orders are therefore incorrect.

We hereby take the opportunity to reiterate the advantage of using canonical Lie transforms in the reduction procedure: Since these transformations are invertible and their inverse can be algebraically computed, information on the original system [as described by Hamiltonian (1.14)] can be fully recovered using the dynamics of the reduced Hamiltonians. The model G_5 contains more information than what is provided by KH theory. In particular, the KH theory does not provide Φ_5 , and as a consequence, we are not able to reconstruct consistently the trajectory from $H_{\rm KH}$.

2.2.2.5 Influence of a slowly varying laser envelope

We consider a slowly varying envelope $f(\epsilon t)$ compared to the frequency of the laser, such that $|\partial^k f(\epsilon t)/\partial t^k| \ll \omega^k$. The *k*th derivative of the envelope is of order ϵ^k . We perform a similar derivation as in Sec. 2.2.2.2. The difference is that in Eqs. (2.13) and (2.14), we use an integration by parts to determine the expression of the generating function at order *n*. In this section, the electric field and the vector potential for f = 1 are denoted $\mathbf{E}_1(t) = E_0/\sqrt{\xi^2 + 1}[\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \xi \sin(\omega t)]$ and $\mathbf{A}_1(t) = E_0/\omega\sqrt{\xi^2 + 1}[-\mathbf{e}_x \sin(\omega t) + \mathbf{e}_y \xi \cos(\omega t)]$, respectively. With these notations, the electric field and the vector potential read $\mathbf{E}(t) = f(\epsilon t)\mathbf{E}_1(t)$ and $\mathbf{A}(t) = f(\epsilon t)\mathbf{A}_1(t)$, respectively. The generating function reads

$$\begin{split} S^{(6)} &= \epsilon f(\epsilon t) \mathbf{r} \cdot \mathbf{A}_{1}(t) - \frac{\epsilon^{2}}{\omega^{2}} \left(f(\epsilon t) \mathbf{p} + \omega f'(\epsilon t) \mathbf{r} \right) \cdot \mathbf{E}_{1}(t) \\ &+ \frac{\epsilon^{3}}{\omega^{2}} \mathbf{A}_{1}(t) \cdot \left(\frac{\mathbf{E}_{1}(t)}{4} + \frac{\partial V}{\partial \mathbf{r}} - \omega^{2} f''(\epsilon t) \mathbf{r} - 2f'(\epsilon t) \mathbf{p} \right) \\ &- \frac{\epsilon^{4}}{\omega^{4}} \left[\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{E}_{1}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V - 3\omega^{2} f''(\epsilon t) \mathbf{p} \cdot \mathbf{E}_{1}(t) - \omega^{3} f'''(\epsilon t) \mathbf{r} \cdot \mathbf{E}_{1}(t) + 3f'(\epsilon t) \mathbf{E}_{1}(t) \cdot \frac{\partial V}{\partial \mathbf{r}} \\ &+ \frac{3\omega}{8} f(\epsilon t) f'(\epsilon t) \left(|\mathbf{E}_{1}(t)|^{2} - \omega^{2} |\mathbf{A}_{1}(t)|^{2} \right) \right] \\ &+ \frac{\epsilon^{5}}{\omega^{4}} \left\{ \left[\left(f(\epsilon t)^{2} - 1 \right) \frac{\mathbf{E}_{1}(t)}{4} + \left(f(\epsilon t) - 1 \right) \frac{\partial V}{\partial \mathbf{r}} \right] \cdot \frac{\partial}{\partial \mathbf{r}} - \left(f(\epsilon t) - 1 \right) \left(\mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \right)^{2} \right\} \left(\mathbf{A}_{1}(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V \\ &+ \frac{\epsilon^{5}}{\omega^{4}} \left[4f'''(\epsilon t) \mathbf{p} \cdot \mathbf{A}_{1}(t) + \omega^{4} f''''(\epsilon t) \mathbf{r} \cdot \mathbf{A}_{1}(t) - 4\omega f'(\epsilon t) \mathbf{p} \cdot \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{A}_{1}(t) \cdot \frac{\partial V}{\partial \mathbf{r}} \right) \\ &- \frac{3\omega^{2}}{8} \left(f'(\epsilon t)^{2} + 3f(\epsilon t) f''(\epsilon t) \right) \mathbf{E}_{1}(t) \cdot \mathbf{A}_{1}(t) \right]. \end{split}$$

The Hamiltonian reads

$$H^{(5)} = \bar{k} + \epsilon \left[\frac{|\bar{\mathbf{p}}|^2}{2} + V(\bar{\mathbf{r}}) \right] + \epsilon^3 f(\epsilon t)^2 \mathbf{U}_p + \epsilon^5 \frac{f(\epsilon t) \mathbf{U}_p}{\omega^2 (\xi^2 + 1)} \left[f(\epsilon t) \left(\frac{\partial^2 V}{\partial x^2} + \xi^2 \frac{\partial^2 V}{\partial y^2} \right) - 3(1 + \xi^2) \omega^2 f''(\epsilon t) \right].$$

Up to the fifth order, the Hamiltonian does not depend on the derivatives of the laser envelope. Hence, at the fifth order, the drift velocities of the electron depend on the variations of the laser envelope. At the first orders, the change of coordinates is given by

$$\bar{\mathbf{r}} = \mathbf{r} - \frac{\epsilon^2}{\omega^2} f(\epsilon t) \mathbf{E}_1(t) - 2 \frac{\epsilon^3}{\omega} f'(\epsilon t) \mathbf{A}_1(t) + O(\epsilon^4),$$

$$\bar{\mathbf{p}} = \mathbf{p} - \epsilon f(\epsilon t) \mathbf{A}_1(t) + \frac{\epsilon^2}{\omega} f'(\epsilon t) \mathbf{E}_1(t) - \frac{\epsilon^3}{\omega^2} \left[f(\epsilon t) \frac{\partial}{\partial \mathbf{r}} \left(\mathbf{A}_1(t) \cdot \frac{\partial}{\partial \mathbf{r}} \right) V - \omega f''(\epsilon t) \mathbf{A}_1(t) \right]$$

$$+ O(\epsilon^4).$$

$$(2.20a)$$

When the laser field is turned off, the envelope of the laser becomes $f(\epsilon t) = 0$. In Eqs. (2.20), we observe that when the laser field is turned off, $\bar{\mathbf{r}} = \mathbf{r}$ and $\bar{\mathbf{p}} = \mathbf{p}$. As a consequence, when the laser field is turned off, the guiding-center trajectory and the electron trajectory are the same.

2.3 Analysis of the guiding-center (GC) models

In this section, we analyze the different GC models, composed of a time-independent Hamiltonian H_m and a canonical transformation Φ_n . In what follows, we restrict the analysis to the soft-Coulomb potential [78, 17] [see Eq. (1.8)] with softening parameter a = 1.

2.3.1 Comparison between Hamiltonian (1.14) and the reduced models

Figure 2.2 shows typical electron trajectories (dark blue curve) of Hamiltonian (1.14), the GC trajectories of G_2 (solid cyan curve) and G_5 (solid red curve) and the associated reconstructed trajectories (dashed curves), for d = 2, $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$, $\omega = 0.05$ a.u. Different ellipticities are considered: $\xi = 0, \xi = 0.5$ and $\xi = 1$. The GC trajectories $(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$ are computed by solving the forward and backward equations of motion of the corresponding GC Hamiltonians (2.17) with initial conditions $\Phi_n(\mathbf{r}(t_0), \mathbf{p}(t_0))$, where t_0 is chosen such that $|\mathbf{r}(t_0)| > 50$ a.u. (~ $2E_0/\omega^2$). We observe that the cyan and red solid curves guide the oscillating dark blue curves. Therefore, the electron oscillates around a GC. For instance, in Figs. 2.2c-d, we see that the trajectory ionizes if the GC motion is unbounded, and it returns to the ionic core if the GC returns to the core. Moreover, we observe a qualitative agreement between the electron trajectory and the reconstructed trajectory using the models G_2 and G_5 . In addition, we observe an overlap almost everywhere between the dark blue curve and the red dashed curve, a signature of a very good quantitative agreement between the electron trajectory and the reconstructed trajectory of the model G_5 . For the G_2 , the overlap is mainly observed far from the ionic core, i.e., for shorter integration times (less than 10T). Below, we provide more thorough analyses to see if and when this agreement between the reduced models and the true trajectories holds. We consider the case d = 1 for clarity. In what follows, the parameters are $\xi = 0$, $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$. and $\omega = 0.05$.

As a consequence of d = 1, the electron and GC phase-space coordinates are reduced to $(\mathbf{r}, \mathbf{p}) = (x\mathbf{e}_x, p\mathbf{e}_x)$ and $(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = (\bar{x}\mathbf{e}_x, \bar{p}\mathbf{e}_x)$, respectively. Looking at longer trajectories as it is done in Fig. 2.3, we observe multiple returns of the electron to the ionic core. The upper panel of Fig. 2.3 shows a typical trajectory (dark blue curve) of Hamiltonian (1.14), and the GC trajectory for G₅ (red curve) for every interval of time when the electron is far from the ionic core. The GC trajectory is solution of the forward and backward equations of motion of Hamiltonian (2.17b), with initial conditions $\Phi_5(x(t_0), p(t_0))$, such that $|x(t_0)| > 50$ a.u. In the lower panel of Fig. 2.3, the dark blue curve is the GC energy $H_5(\Phi_5(x(t), p(t)))$, i.e., at each time, the transformation Φ_5 is performed on the electron phase-space coordinates, and its associated energy H_5 is computed. The red curves are the GC energy of Hamiltonian (2.17b) for initial conditions $\Phi_5(x(t_0), p(t_0))$, i.e., the energy of the GC of G₅, which is conserved. In Fig. 2.3, we observe that the GC reproduces well the mean trajectory of the electron for several time intervals when the electron is far from the ionic core, in a similar way as it was observed in Fig. 2.2. As a consequence, the GC energy of the electron $H_5(\Phi_5(x(t), p(t)))$ is approximately conserved in a piece-wise manner in time. However, we notice that the energy strongly varies during close encounters between the electron and its ionic core. In addition, once the electron has undergone a close encounter, the GC energy of the electron jumps to another energy level.



Figure 2.3: Typical electron trajectory of Hamiltonian (1.14) for $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$, d = 1, $\xi = 0$, Z = 1 and $\omega = 0.05$, and the GC trajectory of the model G₅, for multiple initial conditions $\Phi(x(t_0), p(t_0))$ such that $|x(t_0)| > 50$ a.u., with forward and backward integration of the equations of motion of H_5 . The grey areas are where the GC position is $|\bar{x}| < 35$ a.u. Upper panel: Dark blue and red curves are the electron and the GC trajectory, respectively. In the grey regions, the red curves would typically look like the red curve in the top panel of Fig. 2.6. Lower panel: The dark blue curve is the GC energy $H_5(\Phi_5(x(t), p(t)))$. The red curves are the GC energy of G₅, given by $H_5(\Phi_5(x(t_0), p(t_0)))$, respectively. The horizontal black line is $\mathcal{E} = 0$. Here x and \mathcal{E} are in atomic units.

These observations on the reconstructed trajectories and on the GC energy lead us to consider two different methods for comparing in a more systematic way Hamiltonian (1.14) with the *n*-th order GC model $G_n = (H_m, \Phi_n)$. They consist of:

- (i) Computing trajectories of Hamiltonian $H_m(\bar{\mathbf{r}}, \bar{\mathbf{p}})$ for $t \in [t_0, t_f]$ with initial conditions $\Phi_n(\mathbf{r}(t_0), \mathbf{p}(t_0))$, and then performing the inverse change of coordinates $\Phi_n^{-1}(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$ to obtain the reconstructed trajectories.
- (ii) Computing GC energies $H_m(\Phi_n(\mathbf{r}(t), \mathbf{p}(t)))$ with $(\mathbf{r}(t), \mathbf{p}(t))$ the trajectory of Hamiltonian (1.14) for $t \in [t_0, t_f]$.

In what follows, we use these two methods to test the validity and the benefits of the reduced models. Using method (i), the reconstructed trajectories of the model must be close to the true electron trajectory for the models to be relevant. This method is employed in Fig. 2.4. Using method (ii), by definition, the GC energy of the electron $H_m(\Phi_n(\mathbf{r}(t), \mathbf{p}(t)))$ must be conserved up to some order for the reduced models to be relevant. This method is employed in Figs. 2.5 and 2.6. In addition, we use these tests to compare the reduced models and to provide some guidelines on which models should be used for practical purposes.

2.3.1.1 Reconstructed trajectories

Figure 2.4 shows the most probable distance error $\delta x_n(t)$ between the reconstructed trajectories of the model G_n and the electron trajectory, such that

$$\delta x_n(t) = |\Pi_x(\Phi_n^{-1}(\bar{x}(t), \bar{p}(t))) - x(t)|, \qquad (2.21)$$

where x(t) is the trajectory of Hamiltonian (1.14), $\bar{x}(t)$ is the GC trajectory of Hamiltonian H_m with initial condition $\Phi_n(x(0), p(0))$, and Π_x is the projection from phase space onto the position component, i.e., $\Pi_x(x,p) = x$. The most probable distance error is determined using the maximum of the kernel density estimation [157] of the distance error as a function of t/T. Specifically, it is determined in two steps for a fixed t/T: First, we compute the kernel density estimation of our data, then, we locate its maximum. The equations of motion for \bar{x} and \bar{p} for the models $G_2 = (H_2, \Phi_2)$, $G_3 = (H_2, \Phi_3)$ and $G_4 = (H_2, \Phi_4)$ are the



Figure 2.4: Most probable distance error δx_n [see Eq. (2.21)] as a function of time t per laser cycle T (in log-log scale) for $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$, d = 1, $\xi = 0$, Z = 1 and $\omega = 0.05$ a.u. The electrons are initialized such that the initial velocity of the GC of G₂ is zero, and its initial position is normally distributed, with a mean value 1000 a.u. and a standard deviation 5 a.u. The initial laser phase is uniformly distributed $\varphi_{\text{CEP}} \in [0, 2\pi]$. The dashed lines are the linear fit curves. Inset: Mean value of the position of the electron $\langle x \rangle$ as a function of t/T. Here δx_n is in atomic units.

same, given that the Hamiltonians are the same. The differences between these models come from the change of coordinates, as taken into account in the determination of the initial conditions of the GC trajectory and in the reconstruction of the trajectory from the GC phase-space coordinates. This is also the case when comparing the models $G_5 = (H_5, \Phi_5)$ and $G_6 = (H_5, \Phi_6)$.

The distance error between the electron trajectories and the reconstructed trajectories is increasing for increasing time. Moreover, the distance error is increasing faster for $n = \{2, 3, 4\}$ than for $n = \{5, 6, 7\}$. As a consequence, at $t \approx 100T$, the distance errors δx_n for $n = \{2, 3, 4\}$ are two orders of magnitude greater than the ones for $n = \{5, 6, 7\}$. More quantitatively, the most probable distance error δx_n scales as

$$\delta x_n \propto |t|^{\alpha_n}$$

We observe that $\alpha_n \approx 2.1$ for $n = \{2, 3, 4\}$, and $\alpha_n \approx 1.1$ for $n = \{5, 6, 7\}$.

Counter-intuitively, we observe no significant quantitative improvements, neither between the models G_2 , G_3 and G_4 , nor between the models G_5 , G_6 and G_7 . Hence, far from the ionic core, the corrective terms in the change of coordinates Φ_3 and Φ_4 , are negligible (at least for the chosen parameters), and the models G_2 , G_3 and G_4 provide similar results. In the same way, the corrective terms in Hamiltonian H_7 , compared with H_5 , are negligible, as well as the corrective terms in the change of coordinates Φ_6 and Φ_7 .

2.3.1.2 Guiding-center (GC) energy

We complement the analysis of the trajectories by looking at a specific property of the reduced models, namely the conservation of energy. In Fig. 2.5, an ensemble of trajectories is initiated such that the initial velocity of the GC G₅ is zero, and the initial position of the GC G₅ is normally distributed with a mean value 100 a.u. and a standard deviation 5 a.u. The initial laser phase is uniformly distributed $\varphi_{\text{CEP}} \in [0, 2\pi]$. The distribution in the lower panel of Fig. 2.5 represents the distribution of the GC energy $H_5(\Phi_5(x(t), p(t)))$ as a function of t/T. In the upper panel, the dark blue curve is a typical electron trajectory in the ensemble. The red curve is the GC energy $H_5(\Phi_5(x(t), p(t)))$ of the electron trajectory corresponding to the dark blue curve in the upper panel.

For t < 7T, we observe in the lower panel that the distribution is peaked around the initial GC energy of the electron. During this time, the electrons are far from the ionic core. At $t \sim 8T$, the electrons get close to the ionic core, and the GC energy distribution starts to spread out. When the electrons are close to the ionic core, their dynamics is highly nonlinear due to the competition between the strong laser and Coulomb fields, and the energy curve of a single trajectory in the lower panel starts varying significantly. In the meantime, the inset in the upper panel shows that the approximate trajectory of the reduced model no longer reproduces the electron trajectory. For t > 9T, the electron is far from the ionic core, and the red curve in the lower panel stops varying. It means that the model G_5 is again relevant, but for a different energy level than the initial energy. The arrow indicates the jump of the GC energy after the close encounter with the ionic core. Close encounters with the ionic core are short time processes, and therefore cannot be averaged in time. It

CHAPTER 2. GUIDING CENTER MODEL

is expected that the fast-time average we perform fails to describe the various energy exchanges happening on these short time scales.

Figure 2.6 shows the most probable relative energy error $\delta e_n(t)$ for the models G_n as a function of the distance between the electron and the ionic core |x(t)|, such that

$$\delta e_n(t) = \left| \frac{H_m(\Phi_n(x(t), p(t))) - H_m(\Phi_n(x(0), p(0)))}{H_m(\Phi_n(x(0), p(0)))} \right|,$$
(2.22)

where (x(t), p(t)) are the electron phase-space coordinates at time t. The most probable energy error is the maximum of the kernel density estimation [157] of the energy error. It is determined using the same technique as for computing the most probable distance error (see Fig. 2.4). The initial conditions are the same as in Fig. 2.4, and the integration is stopped when the electron reaches x = 1 a.u.

As expected, we observe that the energy error δe_n increases when |x| decreases, i.e., as the electron approaches the ionic core. Far away from the ionic core, we observe that the most probable energy error δe_n scales as

$$\delta e_n \propto |x|^{-\beta_n},$$

with $\beta_n \approx 3.0$ for $n = \{2, 3, 4\}$, and $\beta_n \approx 6.5$ for $n = \{5, 6, 7\}$. As in Sec. 2.3.1.1, for we observe no significant quantitative improvements, neither between the models G_2 , G_3 and G_4 , nor between the models G_5 , G_6 and G_7 .

A cross-over is observed between all the models when the electron reaches ~ 35 a.u. In particular, from Fig. 2.6, we observe that $G_2 = (H_2, \Phi_2)$ gives the smallest energy errors among the reduced models close to the ionic core. The main reason is that H_2 and Φ_2 do no contain derivatives of the potential. As such, G_2 constitutes the most robust model among the hierarchy. Far from the ionic core (> 35 a.u.), the higher order models provide a better quantitative agreement with the electron trajectories as shown in Figs. 2.4 and 2.6. The efficiency of the higher-order models appears far from the ionic core, around 35 a.u.

2.3.1.3 Discussion

The relative failure of the higher order models close to the ionic core can be understood by looking at the magnitude of the corrective terms in the Hamiltonians H_m and in the changes of coordinates Φ_n . Figure 2.7 shows $h_1 = V(\bar{x})$, $h_5 = (U_p/\omega^2)V''(\bar{x})$ and $h_7 = (U_p/\omega^4)V''(\bar{x})^2$ as a function of the distance between the GC and the ionic core. The term h_n corresponds to the time-independent term in Hamiltonian (2.16) of order ϵ^n . We observe an overlap between these terms, where $h_1 \sim h_5$ for $|\bar{x}| \approx x_c$. Far from the ionic core, we approximate the soft-Coulomb potential by a hard-Coulomb potential $V(\bar{x}) \approx -1/|\bar{x}|$ and we compute h_n explicitly. We deduce that $h_1 \sim h_5$ for $\bar{x} \approx x_c$ with

$$x_c \sim E_0/\omega^2$$
,

which is approximately equal to 21 a.u. for $I = 10^{14}$ W \cdot cm⁻² and $\omega = 0.05$ a.u. Therefore, for $|\bar{x}| \gg x_c$, the terms are ordered such that $h_1 > h_5 > h_7$. In this case, the series in the perturbative expansion of Hamiltonian (2.16) and the change of coordinates (2.18) are likely converging, and the models G_n for n > 2are relevant for the GC dynamics. For $|\bar{x}| \ll x_c$, the terms are ordered such that $h_1 < h_5 < h_7$. In this case, the series in the perturbative expansion of Hamiltonian (2.16) and the change of coordinates (2.18) becomes very far from the true trajectory, and the models G_n for n > 2 are no longer relevant for the dynamics of Hamiltonian (1.14).

For parameters which push the electron far from the ionic core, the higher order models become more relevant and precise. For instance, in the Perelomov-Popov-Terent'ev [137, 138, 136] (PPT) theory of ionization, initially the electron is at a distance $|\mathbf{r}| \sim (E_0/\omega^2)\gamma$ from the ionic core, where $\gamma = \omega \sqrt{2I_p}/E_0$ is the Keldysh parameter with I_p the ionization potential. For small or intermediate Keldysh parameter (i.e., the tunneling regime), the electron ionizes close to the ionic core compared to the quiver radius. Here, we expect the model G_2 to be more relevant because of its robustness close to the ionic core. However, for large Keldysh parameter $(\gamma \gg 1)$, the electron ionizes far from the ionic core compared to the quiver radius. In particular, in a recent experiment [170] in which photoelectron momentum distributions were measured, the parameters –laser intensity $I_0 = 6 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, laser wavelength $\lambda = 400 \text{ nm}$ and atom Ar– correspond to a Keldysh parameter of $\gamma \sim 3$. Therefore, in the multiphoton regime $(\gamma \gg 1)$ and in particular for the set of parameters used in this experiment, we expect the higher orders models G_5 or G_7 to be more relevant.



Figure 2.5: Electron trajectory for $I = 10^{14}$ W · cm⁻², d = 1, Z = 1 and $\omega = 0.05$ a.u. The grey areas are where the GC position is $|\bar{x}| < 35$ a.u. Upper panel: The dark blue curve is the electron trajectory of Hamiltonian (1.14) as a function of time t per laser cycle T. The solid and dashed red curves are the GC and the approximate trajectory for G₅, respectively, with initial condition $t_0 = 0$. The inset is a zoom of the region around the grey area. Lower panel: Logarithm of the distribution of the GC energy $H_5(\Phi_5(x(t), p(t)))$ as a function of t/T. The GCs are initialized with a normal distribution with mean value 100 a.u. and standard deviation 5a.u., with zero-velocity and a uniformly distributed initial laser phase $\varphi_{\text{CEP}} \in [0, 2\pi]$. The red curve is the GC energy $H_5(\Phi_5(x(t), p(t)))$ of the dark blue curve in the upper panel. Here x and \mathcal{E} are in atomic units.



Figure 2.6: Most probable energy error δe_n [see Eq. (2.22)] as a function of the distance between the electron and the ionic core |x| (in log-log scale), for $I = 10^{14}$ W · cm⁻², d = 1, Z = 1 and $\omega = 0.05$ a.u. The initial conditions are the same as in Fig. 2.4. The dashed lines are the linear approximation for $|x| \in [1, 35]$ a.u. The inset shows a zoom of the curves. Here |x| is in atomic units.



Figure 2.7: Representation of h_n as a function of the distance between the GC and the ionic core $|\bar{x}|$ (in log-log scale) for $I = 10^{14}$ W \cdot cm⁻², d = 1, Z = 1 and $\omega = 0.05$. The term h_n corresponds to the term of order $O(\epsilon^n)$ in the Hamiltonian (2.16), which are: $h_1 = V(\bar{x})$, $h_5 = (U_p/\omega^2)V''(\bar{x})$, and $h_7 = (U_p/\omega^4)V''(\bar{x})^2$. The dashed lines are the asymptotic behaviors of these terms for soft-Coulomb potential (1.8), which are for $|x| \gg 1$: $|h_1| \propto |\bar{x}|^{-1}$, $|h_5| \propto |\bar{x}|^{-3}$ and $|h_7| \propto |\bar{x}|^{-6}$. The vertical line is $x_c = E_0/\omega^2$. Here h_n and $|\bar{x}|$ are in atomic units.



Figure 2.8: Cartoon of scenario of step (ii) for d = 1 using the second order GC model $G_2 = (H_2, \Phi_2)$. The thin lines are constant energy surfaces $H_2(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \mathcal{E}$, and the yellow thick line is the separatrix of the GC energy $\mathcal{E} = 0$. The parameters are $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$, Z = 1 and $\xi = 0$. The thick cyan line is the GC trajectory. The thick blue lines are the reconstructed trajectory $\Phi_2^{-1}(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$.

2.3.2 Guiding-center (GC) phase-space dynamics

In this section, we compare the dynamics of the GC given by the three main Hamiltonians H_2 , H_5 and H_7 in Eqs. (2.17). In the case of Hamiltonian H_2 , the final momentum of the GC is computed as a function of its initial conditions.

2.3.2.1 For Hamiltonian H_2

The first (non-trivial) element of the hierarchy is $G_2 = (H_2, \Phi_2)$ and the phase space of the electron is depicted in Fig. 2.8. This model was identified above as the most robust one in the hierarchy for the analysis of the GC dynamics, since its GC energy error is lower than for any other models close to the ionic core. We notice that this is the only reduced Hamiltonian which does not depend on the parameters of the laser field. The dependence on the laser field is in the change of variables Φ_2 . Moreover, if the potential is rotationally invariant, as is the case for atoms, the resulting Hamiltonian is integrable since the angular momentum is conserved in addition to the Hamiltonian.

The change of variables is exactly given by

$$\bar{\mathbf{r}} = \mathbf{r} - \epsilon^2 \mathbf{E}(t) / \omega^2,$$

$$\bar{\mathbf{p}} = \mathbf{p} - \epsilon \mathbf{A}(t).$$

What is particularly convenient with this GC model is that the potential is taken into account in the Hamiltonian and the electric field in the change of variables.

Ionization occurs if and only if the energy of the GC $\mathcal{E} = H_2(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$ is positive. Otherwise the motion of the electron is bounded since the GC moves on a quasi-periodic orbit. The laser parameters have no influence on the motion of the GCs (and this holds up to the fourth-order model). They only influence how the electron swirls around the quasi-periodic orbit. We consider potentials invariant under rotations $V(\mathbf{r}) = \mathcal{V}(|\mathbf{r}|)$. The Hamiltonian reads

$$H_2(\bar{\mathbf{r}}, \bar{\mathbf{p}}) = \frac{|\bar{\mathbf{p}}|^2}{2} + \mathcal{V}(|\mathbf{r}|).$$

The guiding-center motion is planar due to the conservation of the angular momentum. We perform the polar-nodal canonical transformation [66], $(\bar{\mathbf{r}}, \bar{\mathbf{p}}) \mapsto (r, \theta, \nu, p_r, p_{\theta}, p_{\nu})$,

$$\bar{\mathbf{r}} = r\mathbf{Q}\mathbf{e}_x, \quad \bar{\mathbf{p}} = \mathbf{Q}(\mathbf{e}_x p_r + \mathbf{e}_y p_\theta/r),$$

where the total rotation matrix is $\mathbf{Q} = \mathbf{R}_z(\nu)\mathbf{R}_x(i)\mathbf{R}_z(\theta)$, where \mathbf{R}_x and \mathbf{R}_z are given in Appendix A, $\cos i = p_\nu/p_\theta$ and $\sin i = [1 - (p_\nu/p_\theta)^2]^{1/2}$. The GC Hamiltonian for the hard Coulomb potential becomes

$$H_2(r,\theta,\nu,p_r,p_{\theta},p_{\nu}) = \frac{p_r^2}{2} + \frac{p_{\theta}^2}{2r^2} + \mathcal{V}(r).$$
(2.23)

For rotationnally invariant potentials, the Hamiltonian is autonomous and p_{θ} , ν and p_{ν} are conserved. Three quantities in involution are conserved and the system has three degrees of freedom. Therefore the system is integrable in the Liousville sense. In analogy with the quantum case, the angular quantum numbers land m are associated with the conservation of p_{θ} and p_{ν} , respectively. In order to compute the asymptotic configuration of the guiding center with Hamiltonian (2.23), one needs to determine the initial conditions in the polar-nodal coordinates as a function of the initial conditions in the Cartesian coordinates. The momenta are given by $p_r = \bar{\mathbf{p}} \cdot \bar{\mathbf{r}} / |\bar{\mathbf{r}}|$, $p_{\theta} = |\mathbf{L}|$ and $p_{\nu} = \mathbf{L} \cdot \mathbf{e}_z$, with $\mathbf{L} = \bar{\mathbf{r}} \times \bar{\mathbf{p}}$. The distance from the origin is $r = |\bar{\mathbf{r}}|$. Concerning the angles, we first introduce the unitary vectors

$$\mathbf{e}_1 = \begin{cases} \mathbf{e}_x & \text{if } \mathbf{e}_z \times \mathbf{L} = \mathbf{0}, \\ \mathbf{e}_z \times \mathbf{L} / |\mathbf{e}_z \times \mathbf{L}| & \text{otherwise,} \end{cases}$$

and $\mathbf{e}_2 = \mathbf{L}/|\mathbf{L}| \times \mathbf{e}_1$. The angles θ and ν as a function of the Cartesian coordinates are given by

$$\cos\theta = \mathbf{e}_1 \cdot \bar{\mathbf{r}} / |\bar{\mathbf{r}}|, \quad \sin\theta = \mathbf{e}_2 \cdot \bar{\mathbf{r}} / |\bar{\mathbf{r}}|,$$

and

$$\cos\nu = \mathbf{e}_1 \cdot \mathbf{e}_x, \quad \sin\nu = \mathbf{e}_1 \cdot \mathbf{e}_y.$$

If $\mathbf{e}_z \times \mathbf{L} = \mathbf{0}$, sin i = 0 and as a consequence the angle of rotation is $\theta + \nu$. By fixing $\mathbf{e}_1 = \mathbf{e}_x$ in this case, we choose arbitrary $\nu = 0$ in order to be consistent with the definition of θ and ν . The energy $\mathcal{E} = H_2(r, \theta, \nu, p_r, p_\theta, p_\nu)$, the angular momentum p_θ , ν and p_ν are clearly constants of the motion.

If $\mathcal{E} > 0$, the GC trajectory is unbounded and the electron reaches the detector. We consider the case when the GC is far from the ionic core, and as a consequence, we can approximate $\mathcal{V}(r) \approx -Z/r$. The asymptotic configuration (when r goes to infinity) is given by $p_r = \sqrt{2\mathcal{E}}$. Concerning the final scattering angle θ , if $p_{\theta} = 0$, $\theta = \theta_0$ (if $p_{r,0} > 0$) and $\theta_0 + \pi$ (if $p_{r,0} < 0$). If $p_{\theta} \neq 0$, the final scattering angle is given by

$$\theta = \begin{cases} \theta_0 + \sin^{-1} u_0 + \sin^{-1} \beta & \text{if } p_{r,0} > 0, \\ \theta_0 + \pi/2 + \sin^{-1} \beta & \text{if } p_{r,0} = 0, \\ \theta_0 + \pi - \sin^{-1} u_0 + \sin^{-1} \beta & \text{if } p_{r,0} < 0, \end{cases}$$
(2.24)

with $u_0 = \beta [p_{\theta}^2/(Zr_0) - 1]$, $\beta = (2\mathcal{E}p_{\theta}^2/Z^2 + 1)^{-1/2}$ and $r_0 = |\bar{\mathbf{r}}_0|$. Finally, the final momentum of the GC in the Cartesian coordinates is given by

$$\bar{\mathbf{p}} = p_r \mathbf{Q} \mathbf{e}_x.$$

We notice that in the two-dimensional case where the dynamics is in the polarization plane $(\mathbf{e}_x, \mathbf{e}_y)$, we have $\mathbf{L} \times \mathbf{e}_z = \mathbf{0}$. Therefore, $|p_{\theta}| = |p_{\nu}|$, $\sin i = 0$ and \mathbf{R}_{ν} is the identity matrix. Therefore, in this case, the final momentum reads $\bar{\mathbf{p}} = p_r(\mathbf{e}_x \cos\theta + \mathbf{e}_y \sin\theta \cos i)$, where $\cos i = +1$ (resp. -1) for $p_{\nu} > 0$ (resp. < 0).

If $\mathcal{E} < 0$, the typical GC trajectory is on a Kepler orbit, where E is the energy of the orbit, as it is the case for the GC trajectories in Figs. 2.2a–c and Fig. 2.2f. One of the particularities of these orbits is that the radial momentum $\mathbf{\bar{p}} \cdot \mathbf{\bar{r}}/|\mathbf{\bar{r}}|$ vanishes twice in a revolution cycle: Once when the GC trajectory is at the perihelion r_{-} (minimum distance from the ionic core) and $d(\mathbf{\bar{p}} \cdot \mathbf{\bar{r}}/|\mathbf{\bar{r}}|)/dt > 0$, and once when the GC trajectory is at the aphelion r_{+} (maximum distance from the ionic core) and $d(\mathbf{\bar{p}} \cdot \mathbf{\bar{r}}/|\mathbf{\bar{r}}|)/dt < 0$ such that

$$r_{\pm} = \frac{Z}{2|\mathcal{E}|} \left(1 \pm \sqrt{1 - 2p_{\theta}^2 |\mathcal{E}|/Z^2} \right), \qquad (2.25)$$

The aphelion and the perihelion are such that $r_+ + r_- = Z/|\mathcal{E}|$, imposing that for a given energy, the larger the aphelion, the smaller the perihelion, i.e., the closer the electron gets to the ionic core.

2.3.2.2 For Hamiltonians H_5 and H_7

When going to higher-order models, the Hamiltonian H_2 gets perturbed by h_5 (and h_7). As a consequence, since the perturbation mainly affects the trajectories that pass close to the ionic core, i.e., $r_- < x_c \sim E_0/\omega^2$,


Figure 2.9: Poincaré sections $\mathbf{\bar{r}} \cdot \mathbf{\bar{p}} = 0$ and $d(\mathbf{\bar{p}} \cdot \mathbf{\bar{r}}/|\mathbf{\bar{r}}|)/dt < 0$ in the polarization plane (\bar{x}, \bar{y}) , for potential (1.8), $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$, Z = 1 and $\mathcal{E} = -0.01$ a.u. The cyan, red and cyan thick lines are the boundaries of the Poincaré sections. The grey shaded areas are $|\mathbf{\bar{r}}| < E_0/\omega^2$. The Hamiltonians are: (a) H_2 , (b–e) H_5 , and (f–i) H_7 . The ellipticities are: (b,f) $\xi = 0$, (c,e,g,i) $\xi = 0.5$, (d,h) $\xi = 1$. The frequencies are $\omega = 0.05$ except for (d,h) $\omega = 0.025$ (larger quiver radius). All axes are the same as for (a) unless stated otherwise. All quantities are in atomic units.

we shall see that the most perturbed trajectories in the higher-order models are the ones with a large aphelion $Z/|\mathcal{E}| - E_0/\omega^2 < r_+ < Z/|\mathcal{E}|.$

The fact that the energy of the GCs is conserved is a property which is preserved by construction of the reduction procedure: $\mathcal{E} = H_5(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$ or $\mathcal{E} = H_7(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$ is conserved in time. Consequently, for d = 2, the dimension of phase space is reduced from 5 to 3 + 1. Here 3 + 1 means that phase space is foliated by constant energy surfaces of dimension 3. The advantage is that one can visualize the dynamics using Poincaré sections. Figure 2.9 shows the Poincaré sections $\bar{\mathbf{r}} \cdot \bar{\mathbf{p}} = 0$ and $d(\bar{\mathbf{p}} \cdot \bar{\mathbf{r}}/|\bar{\mathbf{r}}|)/dt < 0$ for $\mathcal{E} = -0.01$ a.u. and the soft-Coulomb potential (1.8), where $\bar{\mathbf{p}} \cdot \bar{\mathbf{r}}/|\bar{\mathbf{r}}| = d|\bar{\mathbf{r}}|/dt$ is the radial momentum of the electron. This Poincaré section corresponds to the position of the GC when it turns back towards the ionic core. The region close to the ionic core, i.e., around $|\bar{\mathbf{r}}| \sim E_0/\omega^2$ (grey areas), is not relevant since the reduction procedure is only valid far away from the ionic core.

For linear polarization ($\xi = 0$), in Fig. 2.9b and Fig. 2.9f, we observe two distinct dynamical behaviors of the GC trajectories. These figures display a chaotic layer far from the ionic core around $Z/|\mathcal{E}| - E_0/\omega^2 < |\bar{\mathbf{r}}| < Z/|\mathcal{E}|$. This ring corresponds to trajectories with a small perihelion and a large aphelion, that come close to the ionic core $|\bar{\mathbf{r}}| < E_0/\omega^2$. These trajectories are the most affected by the perturbation h_5 in the Hamiltonian H_5 according to our discussion in Sec. 2.3.1.3, and typically correspond to the trajectories for which the electron comes back to the ionic core, as it is depicted in Fig. 2.2a, Fig. 2.2c and Fig. 2.2f. The width of this ring is of order E_0/ω^2 . Secondly, we observe a regular region for $Z/2|\mathcal{E}| < |\bar{\mathbf{r}}| < Z/|\mathcal{E}| - E_0/\omega^2$. This region corresponds to trajectories with a perihelion greater than E_0/ω^2 , of the same order of their aphelion. These are the trajectories least affected by the perturbations h_5 and h_7 . As a consequence, we observe orbits that are mostly preserved from the unperturbed Hamiltonian H_2 , and typically correspond to the trajectories for which the electron stays far from the ionic core, as it is depicted in Fig. 2.2b. Also, we observe two elliptic islands for $(\bar{x}, \bar{y}) \sim (\pm 1/2|\mathcal{E}|, 0)$. These islands correspond to mainly circular GC orbits, which become stable with the coupling with the electric field encapsulated in the effective potential of H_5 or H_7 . Finally, the Poincaré sections for H_5 and H_7 look similar. This is consistent with our earlier observation that h_7 does not significantly affect the electron trajectories for these parameters and far from the ionic core.

For elliptical polarization ($\xi = 0.5$), in Fig. 2.9c and Fig. 2.9g, the observations are similar to the linear case, which reinforces the generality of the discussion above. However, for circular polarization ($\xi = 1$), in Fig. 2.9d and Fig. 2.9h, we no longer observe chaotic behavior in the GC dynamics. Indeed, Hamiltonians H_5 and H_7 are rotationally invariant, and as a consequence, the GC angular momentum is conserved. Therefore, the dimension of phase space is reduced from 3 + 1 to 2 + 2 and the system is integrable. We observe that the elliptic islands we observed for $\xi = 0$ and 0.5 around $(\bar{x}, \bar{y}) \sim (\pm Z/2|\mathcal{E}|, 0)$ are no longer present in the circular polarization case, as a consequence of the rotational invariance.



Figure 2.10: Poincaré sections $\mathbf{\bar{r}} \cdot \mathbf{\bar{p}} = 0$ and $d(\mathbf{\bar{p}} \cdot \mathbf{\bar{r}}/|\mathbf{\bar{r}}|)/dt < 0$ of H_5 in the polarization plane (\bar{x}, \bar{y}) , for potential (1.8), $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$ and Z = 1. The red thick lines are the boundaries of the Poincaré sections. The grey shaded areas are $|\mathbf{\bar{r}}| < E_0/\omega^2$. The ellipiticities are (left panel) $\xi = 0$ and (right panel) $\xi = 0.5$. The frequencies are $\omega = 0.05$. All quantities are in atomic units.

In the previous sections we have seen that for the parameters $I = 10^{14}$ W · cm⁻² and $\omega = 0.05$ a.u., the models H_5 and H_7 are almost equivalent since h_5 is several order of magnitude higher than h_7 in the region where the higher order models are relevant, $|\mathbf{\bar{r}}| > E_0/\omega^2$ (see Fig. 2.7). This is verified by comparing the Poincaré sections of Figs. 2.9b–d and Figs. 2.9f–h. However, in Figs. 2.9e–i, we observe that when $\omega = 0.025$ a.u. so that the quiver radius is of the same order as the distance between the GC and the ionic core $(E_0/\omega^2 \sim 85$ a.u.), the dynamics between Hamiltonians H_5 and H_7 differs significantly. The reduced models are not relevant in these regions. Hence, the reduced models G₅ and G₇ are significantly different when the characteristic distance between the GC and the ionic core is the same as the quiver radius (at least for this range of parameters). Similar observations would have been made for $\omega = 0.05$ a.u. by lowering the GC energy \mathcal{E} as observed in Fig. 2.10.

2.4 Conclusions

In sum, we have derived a hierarchy of reduced models G_n for the GC dynamics of the electron interacting with the combined strong laser and Coulomb fields. The reduced models G_n are composed of an averaged Hamiltonian H_m governing the GC dynamics [Eqs. (2.17)] and a transformation Φ_n which maps the electron phase-space coordinates onto the GC phase-space coordinates [Eqs. (2.19)]. As a rule of thumb, these models are relevant when the electron is relatively far away from the ionic core (typically when its distance from the core exceeds one quiver radius), which happens in a piece-wise manner in time. The models do not describe the short events when the electron recollides with the ionic core. However, the models capture the dynamics of the electron on long time scales.

We have singled out two models G_2 and G_5 : The first model provides the leading behavior of the trajectories, is the most tractable one due to its simplicity and the most robust close to the core. In order to improve the quantitative agreement, a higher-order model such as G_5 has to be used.

All these models allow the distinction between direct ionizations and rescattering with the ionic core by looking at the GC energy of the trajectory. This is in particular very useful when the photoelectron momentum distributions are analyzed for imaging the target. Also, we were able to define an energy of the electron far away from the core for this time-dependent system. The rescattering events can be seen as jumps in energy as a result of the transfer of energy from the ionic core to the electron.

Chapter 3

The impact of the Coulomb potential in ATI: Coulomb-driven recollisions and Rydberg state creations

After electrons tunnel ionize, the interplay between their interactions with the laser and their parent ion yields their dynamics highly nonlinear. These nonlinearities give rise to rich and diverse paths the electrons can take towards the detectors. In Ref. [127], it is shown experimentally that electrons, after tunnel ionization, can also be trapped in Rydberg states. The Rydberg states of atoms are excited states in which the energy of the bound electrons in the field-free atom is close to zero, and as a consequence, they can be several atomic units far from the core. The left panel of Fig. 3.1 shows the yield of Rydberg states created from He subjected to an intense laser field as a function of the laser intensity. While the yield of ionization slowly decreases for increasing ellipticity, we observe that the yield of Rydberg states drastically decreases for increasing ellipticities.

When electrons reach the detector with or without recolliding, their final momentum can be measured, and photoelectron momentum distributions (PMDs) can be drawn. Typical PMDs for He are shown in Fig. 8. In Ref. [93], the location of the maximum of the upper lobe in the PMDs measured in experiments (upper panel of Fig. 8), denoted $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$, is tracked for varying ellipticities. In the right panel of Fig. 3.1, the blue and red circles are the momentum coordinates of the maximum of upper lobe in the PMDs P_x and P_y , respectively. The horizontal black and green curves are the predicitions of P_x and P_y in absence of Coulomb potential, i.e., in the SFA, given by Eq. (4). For $\xi \leq 0.1$, we observe that $\mathbf{P} \approx \mathbf{0}$, and as a consequence, the PMD is mainly one lobe centered at the origin. In contrast, the solid green curve increases



Figure 3.1: Left panel: Experimental measurements for $I = 10^{15} \text{ W} \cdot \text{cm}^{-2}$ reproduced from Ref. [127]. The black squares (\blacksquare) and circles (\circ) are the yield of He⁺ and He^{*} as a function of the ellipticity, respectively. Both measurements have been set equal at $\xi = 0$. Right panel: Experimental measurements of the PMD of He for $I = 8 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$ and $\xi = 0.4$ reproduced from Ref. [140] (zoom of Fig. 8 of this manuscript).

linearly in this range of ellipticities. The persistence of the single lobe in the PMDs, revealed by the right panel of Fig. 8, is referred to as *Coulomb focusing* [28, 38]. For $\xi \gtrsim 0.1$, we observe that when P_y is positive, P_x is negative and non-zero. The red circles agree well with the green curve. However, there is a discrepancy between the horizontal black curve and the blue circles. In the PMDs, we observe an asymmetry with respect to the minor polarization axis. This asymmetry, revealed by the right panel of Fig. 8, is referred to as *Coulomb asymmetry* [11, 62]. In the experimental measurements reported in Ref. [104], the final momentum of the electron exhibits the same features as in experiments [140, 93] (see Fig. 3.13). The hypothesis made in Ref. [93] is that there is a *bifurcation* when varying the ellipticity of the laser field. We observe that the ellipticity for which there is a bifurcation in the right panel of Fig. 3.1, the yield of Rydberg states decreases drastically in the left panel of Fig. 3.1.

Questions

- What are the dynamical mechanisms inducing Coulomb asymmetry and Coulomb focusing observed in the PMDs ?
- What are the mechanisms behind the drastic changes triggered by varying ellipticity measured in experiments, such as for instance, the drastic decrease of the yield of Rydberg states for increasing ellipticities and the bifurcation in the PMDs as observed in Fig. 3.1 ?

Plan In this chapter, we use the two-step model described in Sec. 1.2 to investigate the interplay between the tunnel ionization and the Coulomb interaction in above-threshold ionization (ATI). We find that the Coulomb field of the ion makes its presence known even in highly intense laser fields, in contrast to the assumptions of the SFA. The dynamics of the electron after ionization: is analyzed with the reference Hamiltonian (1.14) and three reduced models for an arbitrary laser polarization: the SFA, the CCSFA, and the second order guiding center model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1). These models illustrate clearly the Coulomb effects, in particular *Coulomb focusing* and *Coulomb asymmetry*. We show that the CCSFA and the GC are complementary, in the sense that the CCSFA describes well short time-scale phenomena (shorter than a laser cycle) for which the Coulomb interaction is significant on short time scales, such as in *subcycle recollisions*, while the GC is well suited for describing long time-scale phenomena (longer than a laser cycle) for which the Coulomb interaction is significant on long time scales, such as in *Coulomb-driven recollisions* and *Rydberg state creation*.

The chapter is organized as follows: In Sec. 3.1, we analyze the PMDs and the initial conditions of the trajectories leading to the PMDs with the four models. In particular, we identify the set of initial conditions leading to Rydberg state creation and Coulomb-driven recollisions. We show that these processes are only well described by the GC model. In Sec. 3.2, we use the GC model to describe the mechanisms behind Rydberg state creation and Coulomb-driven recollisions. In particular, the GC model allows us to define a domain of initial conditions, which we refer to as the *rescattering domain*, leading to Rydberg state creations and Coulomb-driven recollisions. Finally in Sec. 3.3, we investigate the shape of the rescattering domain using the GC model. We show how the shape and location of the rescattering domain manifest themselves in experiments, in particular, in the bifurcation of the PMDs (see Fig. 8).

Unless stated otherwise, we use the PPT initial conditions $(\mathbf{r}_0, \mathbf{p}_0, t_0) = (\mathbf{r}_{\text{PPT}}, \mathbf{p}_{\text{PPT}}, t_0)$ given by Eq. (1.25). We consider the electric field given by Eq. (1.10), with $\varphi_{\text{CEP}} = 0$, and a trapezoidal envelope $T_u = 0$, $T_p = 8T$ and $T_d = 2$. At time $T_f = T_u + T_p + T_d$, the laser field is turned off. Here, we mainly focus on the analyses of the electron dynamics during the plateau, and we consider $\mathbf{A}(t) \approx -f(t)E_0[\mathbf{e}_x \sin(\omega t) - \xi \mathbf{e}_y \cos(\omega t)]/\omega\sqrt{\xi^2 + 1}$ and $\mathbf{\Sigma}(t) \approx \mathbf{E}(t) [\mathbf{E}(t) = -\partial \mathbf{A}(t)/\partial t$, $\omega^2 \mathbf{A}(t) = \partial \mathbf{\Sigma}(t)/\partial t$, see Eqs. (2.2) and Eqs. (2.3)].

Publications

- [51] J. Dubois, S. A. Berman, C. Chandre, T. Uzer, Phys. Rev. A 99, 053405 (2019).
- [49] J. Dubois, S. A. Berman, C. Chandre, T. Uzer, Phys. Rev. Lett. 121, 113202 (2018).

3.1 Photoelectron momentum distributions (PMDs)

In this section, we analyze the influence of short vs. long time-scale microscopic phenomena on macroscopic measurements like the photoelectron momentum distributions in light of the reduced models described in Chap. 2, in particular the CCSFA and the GC models, and their complementarity.



Figure 3.2: PMD for $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$ and $\mathbf{A}(t) = (E_0/\omega) \cos^4(\omega t/2T)[\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \sin(\omega t)]$ computed with the TDSE for d = 2. The softening parameter is a = 0.7, the ionization potential is $I_p \approx 0.5$ a.u. (close to H), and Z = 1. The offset angle Θ corresponds to the angle between the minor polarization axis (\mathbf{e}_x in this case) and the location of the momentum at the maximum of the PMD. In the SFA, the offset angle is $\Theta_{\text{SFA}} = 0$. Momenta are in a.u.

3.1.1 Short time-scale dynamics

In this section, we consider the electron dynamics for which the Coulomb potential acts significantly on a short time scale after ionization. In particular, we study the scattering of the electron during a direct ionization in a CP field ($\xi = 1$) using the four models. In Fig. 3.2, we show the PMD of H solution of the TDSE. We observe the PMD is shifted with respect to the minor polarization axis \mathbf{e}_x of an angle Θ , referred to as the *scattering angle* or the *offset angle*. In this section, we study the scattering angle Θ as a function of the parameters, a signature of the Coulomb effect.

3.1.1.1 Scattering after ionization

First, we consider a circularly polarized (CP) field ($\xi = 1$), used for attoclock measurements [165, 106]. For ellipticity close to circular, the initial drift momentum is large and the electron moves away from the ionic core quickly. Therefore, the corrections due to the Coulomb potential on the electron trajectories occur on a short time scale, and we expect the CCSFA to be accurate. In attoclock measurements, the observable is the offset angle Θ . We assume that it corresponds to the scattering angle of the T-trajectory (see Sec. 1.2.2.3)

$$\Theta = \tan^{-1}(P_y/P_x),$$

where $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y + P_z \mathbf{e}_z$ is the final momentum of the T-trajectory. In order to see the Coulomb asymmetry in a PMD from a CP field, a short laser pulse has to be used [165, 106] (see Fig. 3.2 for the PMD in CP fields for short laser pulses). Otherwise, the PMD would resemble to a ring around the origin. Figure 3.3 shows the T-trajectory final momentum as a function of the intensity I for $\xi = 1$. For P_y (upper panel), we notice that the dashed black curves (CCSFA), the solid black curves (GC model) and the crosses [reference Hamiltonian (1.14)] overlap for $I \in [10^{12}, 10^{16}] \text{ W} \cdot \text{cm}^{-2}$, and hence a good agreement between these three



Figure 3.3: T-trajectory final momentum $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$ as function of the laser intensity I for $\xi = 1$ and Z = 1. The crosses are the T-trajectory final momentum of the reference Hamiltonian (1.14), where P_x and P_y are in blue and red, respectively. The dotted, dashed and solid black curves are the T-trajectory final momentum of the SFA, the CCSFA and the GC model, respectively. The dashed and solid green lines are the approximated and the asymptotic T-trajectory final momentum using Eqs. (3.1) and (3.2), respectively. Momenta are scaled by E_0/ω .

models is observed. In addition, we notice that the value of P_y predicted by these three models is lower compared to the SFA model. This is a microscopic (at the level of the trajectory) signature of the Coulomb focusing. The green dashed curves are approximations of the CCSFA given by Eq. (3.1). By considering that the initial drift momentum of the T-trajectory is large, the integrand in Eq. (2.3b) is large for a very short time after ionization, so we make the approximation $\Sigma(t) \approx \mathbf{E}(t_0) + \omega^2(t-t_0)\mathbf{A}(t_0) - \omega^2(t-t_0)^2\mathbf{E}(t_0)/2$. As a consequence, the SFA trajectory (2.2a) is quadratic in time. Taking $V(\mathbf{r}) \approx -Z/|\mathbf{r}|$ and the initial conditions of the T-trajectory in the PPT ionization theory (1.32) to be such that $(\mathbf{p}_{PPT}^T \cdot \mathbf{e}_{\perp})(t-t_0) \ll$ $|\mathbf{r}_{PPT}^T| + (t-t_0)^2|\mathbf{E}(t_0)|/2$ (which becomes valid at high intensity) the correction of the asymptotic drift momentum is given by

$$\Delta \mathbf{p} \approx \frac{\pi Z \, \mathbf{e}_{\parallel}(t_0)}{(2|\mathbf{r}_{\rm PPT}^T|)^{3/2} \sqrt{|\mathbf{E}(t_0)|}} - \frac{Z(\mathbf{p}_{\rm PPT}^T \cdot \mathbf{e}_{\perp}) \, \mathbf{e}_{\perp}(t_0)}{2|\mathbf{r}_{\rm PPT}^T|^2 |\mathbf{E}(t_0)|}.$$
(3.1)

The unitary vectors are $\mathbf{e}_{\parallel}(t_0) = \mathbf{E}(t_0)/|\mathbf{E}(t_0)|$ and $\mathbf{e}_{\perp}(t_0) = -[\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_y]\mathbf{e}_x + [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_y$. In Ref. [62], a similar result is derived for $\mathbf{p}_{\text{ADK}}^T \cdot \mathbf{e}_{\perp}(t_0) = 0$. We observe that the approximation of the CCSFA [Eq. (3.1)] becomes good only at high intensity $I \gtrsim 10^{15} \text{ W} \cdot \text{cm}^{-2}$, where the drift momentum $|\mathbf{p}_0 - \mathbf{A}(t_0)| \sim E_0/\omega$ is very large and where the electron spends a very short time close to the ionic core. At a very high intensity $I \sim 10^{16} \text{ W} \cdot \text{cm}^{-2}$, all models converge to the same value predicted by the SFA $P_y^{\text{SFA}} = (E_0/\omega)/\sqrt{2}$.

For P_x (lower panel), we observe that the dashed black curves (CCSFA) and the crosses [Hamiltonian (1.14)] overlap for $I \in [10^{12}, 10^{16}] \text{ W} \cdot \text{cm}^{-2}$. The solid black curve (GC model) agrees well with the crosses [reference Hamiltonian (1.14)] only for intensities such that $I \leq 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$. This intensity range corresponds to a Keldysh parameter $\gamma \geq 1.6$ for which the electron initial position is $|\mathbf{r}_{\text{PPT}}| \geq E_0/\omega^2$, i.e., for which the GC model is quantitatively accurate. When the electron ionizes close to the ionic core, there is a large contribution of the Coulomb potential. Mapping the electron coordinates to its GC coordinates [Eqs. (2.18)], and evaluating the Coulomb interaction on its GC only [G₂ = (H₂, Φ_2), see Tab. 2.1] leads to a significant underestimate of the Coulomb effect if the electron is initially close to the ionic core. In the CCSFA, the evaluation of the Coulomb potential is performed on the approximate solution of the SFA. As a consequence, on a short time scale after ionization, the evaluation of the Coulomb interaction is performed on a position which is close to the real trajectory [Hamiltonian (1.14)] and therefore close to the core. We also observe that the dotted curve (SFA, $P_x^{\text{SFA}} = 0$) never agrees with the crosses [reference Hamiltonian (1.14)], even at very high intensity. This is a microscopic signature of the Coulomb asymmetry. In particular, we observe that the Coulomb asymmetry persists even for high intensity. For $I \gtrsim 10^{15} \text{ W} \cdot \text{cm}^{-2}$, we observe that the dashed green curve [Eq. (3.1)] agrees well with the dashed black curve (CCSFA) and the crosses [reference Hamiltonian (1.14)].

Result 2: Persistence of the Coulomb asymmetry

In Eq. (3.1), for very high intensities, or equivalently for very small Keldysh parameter, the correction to the T-trajectory final momentum in the CCSFA becomes

$$\lim_{E_0 \to \infty} \frac{\Delta \mathbf{p}}{(E_0/\omega)} = -\frac{\omega \pi Z \, \mathbf{e}_x}{(2I_p)^{3/2} \sqrt{\xi^2 + 1}},\tag{3.2}$$

which is valid for high ellipticity. The offset angle measured in an attoclock experiments is for high intensity

$$\lim_{E_0 \to \infty} \Theta = \pi - \tan^{-1} \frac{\xi (2I_p)^{3/2}}{\omega \pi Z}.$$

The larger the intensity, the closer to the core the electron is initiated, and thus the T-trajectory remains deflected by the ionic core. In addition, in the reference Hamiltonian (1.14), the larger the intensity, the larger the laser-atom interaction $\mathbf{r} \cdot \mathbf{E}(t)$ and the Coulomb potential contribution $V(\mathbf{r})$ at the tunnel exit. Therefore, the competition between the Coulomb potential and laser interaction is always present even at high intensity, as shown in Ref. [19]. Consequently, the Coulomb asymmetry persists even at very high intensity. Notice that these results do not depend on the shape of the laser pulse, and are still true for ultra short pulses.

In summary, as expected for large ellipticities (i.e., close to CP), there is a very good agreement between the CCSFA [Eqs. (2.3)] and the reference model [Hamiltonian (1.14)] for all intensities. Indeed, for large ellipticities, the electron initial drift momentum is also large, and the Coulomb potential acts significantly on the electron trajectory for a short time after ionization. The Coulomb interaction causes the deflection of the T-trajectory after ionization. For intensities $I \gtrsim 8 \times 10^{13}$ W · cm⁻², the GC model also captures this effect well.

3.1.1.2 Links with the Keldysh-Rutherford model

In Ref. [29], the Keldysh-Rutherford (KR) model [29] is introduced for the offset angle of the PMD when the intensity of the laser is weak and the Coulomb potential is short (see Θ in Fig 3.2). The KR model is based and derived on the intuition that the electron is scattered by the Coulomb potential after the ionization according to the Rutherford formula [151]. In Ref. [29], the potential vector is given by $\mathbf{A}(t) =$ $(E_0/\omega) \cos^4(\omega t/2T)[\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \sin(\omega t)]$ and the laser field is given by $\mathbf{E}(t) \approx E_0 \cos^4(\omega t/2T)[\mathbf{e}_x \sin(\omega t) - \mathbf{e}_y \cos(\omega t)]$. The peak amplitude of the laser field is reached at time t = 0. Here, we show that the KR provides similar results than the GC model, while only the GC model is based on first principles.

We compute the scattering angle Θ (also referred to as the offset angle) of the T-trajectory for $V(\mathbf{r}) = -Z/|\mathbf{r}|$ (hard Coulomb potential). In the KR model, the scattering angle is given by [29]

$$\Theta_{\rm KR} \approx \frac{\omega^2}{E_0} \frac{Z}{I_p}$$

In Ref. [29], the ADK theory is considered for determining the initial conditions of the electron after tunneling. In the ADK theory, the T-trajectory is given by $\mathbf{r}_{ADK}^T \approx -(I_p/E_0)\mathbf{e}_x$, $\mathbf{p}_{ADK}^T = \mathbf{0}$ [see Eq. (1.23)] and ionizes at time t = 0, at the peak amplitude of the laser field. At t = 0, the laser field and the vector potential are $\mathbf{E}(0) = \mathbf{e}_y E_0$ and $\mathbf{A}(0) = -\mathbf{e}_x E_0/\omega$, respectively. We perform the second order GC change of coordinates $\mathbf{G}_2 = (H_2, \Phi_2)$ (see Tab. 2.1). The initial conditions of the GC of the T-trajectory are

$$\bar{\mathbf{r}}_{\mathrm{ADK}}^{T} = -(E_0/\omega^2)(1+\gamma^2/2)\mathbf{e}_y, \bar{\mathbf{p}}_{\mathrm{ADK}}^{T} = (E_0/\omega)\mathbf{e}_x,$$



Figure 3.4: Projection of the photoelectron momentum distributions (PMDs) along p_z for $I = 8 \times 10^{13}$ W · cm⁻², Z = 1 and $\xi = 0.4$ of the reference Hamiltonian (1.14), the SFA [39, 154], the CCSFA [62] and the GC model. The CCSFA and the GC curves overlap. The shaded region corresponds to $|p_z| < 0.05$ a.u. The momentum p_z is scaled by E_0/ω .

where $\gamma = \sqrt{2I_p}\omega/E_0$ is the Keldysh parameter. The energy of the GC is $\mathcal{E}_T = |\mathbf{\bar{p}}_{ADK}^T|^2/2 - Z/|\mathbf{\bar{r}}_{ADK}^T|$ and its angular momentum perpendicular to the polarization plane is $\ell_T = \mathbf{\bar{r}}_{ADK}^T \times \mathbf{\bar{p}}_{ADK}^T \cdot \mathbf{e}_z$ ($\ell_T > 0$). We now use the polar-nodal coordinates in order to determine the asymptotic configuration of the GC, corresponding to the asymptotic configuration of the electron. The initial angle in the polar-nodal coordinates is $\theta_0 = -\pi/2$, the initial radius is $r_0 = (E_0/\omega^2)(1 + \gamma^2/2)$, the initial radial momentum is $p_{r,0} = 0$, and the constants of motion are $p_{\theta} = |\ell_T|$, $p_{\nu} = \ell_T$ and $\nu = 0$. The offset angle corresponds to the asymptotic angle in the polar-nodal coordinates, i.e., $\Theta = \theta$ when $r \to \infty$. Therefore, using Eq. (2.24), one obtains the offset angle $\Theta = \sin^{-1}(1 + 2\mathcal{E}_T p_{\theta}^2/Z^2)^{-1/2} = \sin^{-1}[E_0^3(1 + \gamma^2/2)/(\omega^4 Z) - 1]^{-1}$. In the limit of a weak laser field and a strong ion-electron interaction, one has $\gamma^2/2 \gg 1$ and therefore $\Theta \approx \sin^{-1}[\omega^2 Z/(E_0 I_p)]$. In the limit of small offset angle

$$\Theta \approx \frac{\omega^2}{E_0} \frac{Z}{I_p},$$

which is the same as the one obtained with the KR theory. We find indeed a close relation between the KR model and the GC model since the "half-scattering" angle is built into the GC model, and corresponds to the particular case for which $p_{r,0} = 0$. In the KR model, the initial position is evaluated directly at the position of the electron instead of the GC position, and therefore is more similar to the GC model at the first order.

3.1.2 Long time-scale dynamics

For lower ellipticities, we show that important properties of the system arising from long time-scale processes, in particular Coulomb-driven recollisions and Rydberg state creation, are well described by the GC model. To illustrate this, we consider an intensity $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$ ($\gamma \sim 1.6$) and an ellipticity $\xi = 0.4$.

3.1.2.1 Analysis of the ionized electron momentum

Figure 3.4 shows the projection of the photoelectron momentum distribution on the perpendicular momentum p_z . For the reference Hamiltonian (1.14), the distribution presents a cusp-like peak at zero perpendicular momentum. In Ref. [150], a similar shape of the distribution along the perpendicular momentum measured in experiments and with CTMC calculations has been reported. In the SFA, the drift momentum is conserved, and therefore the distribution does not change with time. As a result, the asymptotic distribution is Gaussian, in contrast with the results with the reference Hamiltonian (1.14) and in experiments [150]. The distribution of the CCSFA and the GC models overlap. In agreement with the observations of Ref. [150], this cusp is due to the long-range Coulomb interaction between the ionized electron and the core

Next, we focus on the part of the PMDs for which $|p_z| < 0.05$ a.u. represented in grey in Fig. 3.4. Figure 3.5 shows the PMDs computed with CTMC simulations of the reference Hamiltonian (1.14), the SFA



Figure 3.5: Photoelectron momentum distributions (PMDs) for $|p_z| < 0.05$ in logarithmic scale for $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, Z = 1 and $\xi = 0.4$ of the reference Hamiltonian (1.14), the SFA [39, 154], the CCSFA [62] and the GC model. The upper (resp. lower) black dot is the T-trajectory final momentum for each model (resp. its symmetric momentum with respect to the origin). The momenta are scaled by E_0/ω .

[Eqs. (2.2)] [39, 154], the CCSFA [Eqs. (2.3)] [62] and the GC model [$G_2 = (H_2, \Phi_2)$, see Tab. 2.1]. The Ttrajectory final momentum is shown with a black dot for each model. The PMDs are mainly composed of two clouds centered around the T-trajectory final momentum. The two clouds are roughly symmetric with respect to the origin according to the symmetry ($\mathbf{r}, \mathbf{p}, t$) $\mapsto (-\mathbf{r}, -\mathbf{p}, t + T/2)$ of the reference Hamiltonian (1.14) for a constant laser envelope (f = 1) which is also preserved by the initial conditions [see Eq. (1.25)] and the reduced models.

For the reference Hamiltonian (1.14), the PMD in the leftmost panel of Fig. 3.5 exhibits three significant features: The asymmetry with respect to the \mathbf{e}_y -axis, the relatively high density of electrons with nearzero momentum –corresponding to near-zero energy photoelectrons [169]–, and the tails for high momentum (regions for $|p_x| > 1$). In order to interpret these features, we compare this PMD with those of the three reduced models. The CTMC approach we use does not take into account any effects due to the absorption of photons or interference after ionization, which lead for example to the rings in ATI [122]. These would of course be included in a time-dependent Schrödinger equation (TDSE) calculation, but also in other kinds of semiclassical calculations.

In the PMD of the SFA [39, 154] (second panel of Fig. 3.5 from the left), the two clouds are symmetric with respect to the \mathbf{e}_y -axis, there is a lack of near-zero energy photoelectrons, and there are no tails for high momentum. Therefore, the asymmetry of the clouds, the presence of near-zero energy photoelectrons and the tails for high momentum observed in the PMD of Hamiltonian (1.14) are a consequence of the Coulomb potential, which is expected to be significant here since the characteristic time of the ionized trajectories is long compared to one laser cycle.

In the PMD of the CCSFA [62] (third panel of Fig. 3.5 from the left), the two clouds are asymmetric with respect to the \mathbf{e}_y -axis. As discussed in the previous section, after ionizing, the electron trajectories deviate because of the Coulomb interaction: This asymmetry is the Coulomb asymmetry. With the CCSFA, however, we observe that the distribution is very low around the origin of momentum space, i.e., there is still a lack of near-zero energy photoelectrons. Indeed, the drift momentum of the near-zero energy photoelectrons is low and the conditions on the validity of the CCSFA are not met. We notice that the integrals we compute numerically for determining the correction to the final momentum of the electron [Eqs. (2.3)] do not always converge. Obviously, the integrals diverge if for instance $\mathbf{p}_0 - \mathbf{A}(t_0) = \mathbf{0}$. Also, for small drift momentum, it is challenging to obtain numerically converged integrals. Finally, we observe tails for $|p_x| > 1$ in the PMD of the CCSFA like in the PMD of Hamiltonian (1.14).

In the PMD of the GC model (rightmost panel of Fig. 3.5), the clouds are asymmetric with respect to the \mathbf{e}_y -axis. After ionizing, the electron trajectories are deflected by the Coulomb force exerted on their GC. One advantage of this model is that the final momentum of the electron has an explicit expression for $V(\mathbf{\bar{r}}) \approx -1/|\mathbf{\bar{r}}|$, and as a consequence the computations of the CTMCs are as fast as the computation of the CTMCs of the SFA. Moreover, this model does not rely on computing integrals that may or may not converge. In addition, we observe that the asymmetric clouds are connected to the origin of the momentum space, showing that the near-zero energy photoelectrons which ionize directly are well captured by this model. We distinguish the near-zero energy photoelectrons which ionize directly from the near-zero energy electrons induced by rescattering; the latter are the cause of the low-energy structure (LES) [24, 84] in the photoelectron energy spectra. However, the absence of tails in the GC model suggests that the tails observed in the reference model and the CCSFA are the contributions of rescattered electrons [82, 114].

Hence, the asymmetry observed in the PMD of the reference Hamiltonian (1.14) is also captured by the reduced models of the CCSFA and the GC. This asymmetry is due to the deviation of the electrons or their GC originating from the Coulomb interaction. In addition, near-zero-energy photoelectrons are captured by the GC model. The tails in the PMDs are due to the rescattering of electrons that have experienced a recollision [82], in which the electron comes close to the ionic core and is rescattered due to the competitive forces between the laser and the Coulomb interaction. This short time-scale process is well known and well described by the CCSFA (see, e.g., Refs. [82, 84, 85, 114]).

3.1.2.2 Analysis of the initial conditions

We investigate the initial conditions of the electron after tunnel-ionization to interpret and understand the origin of the near-zero energy photoelectrons. Figure 3.6 shows the final energy of the electron as a function of its initial conditions after tunneling for $I = 8 \times 10^{13}$ W·cm⁻² and $\xi = 0.4$ for the reference Hamiltonian (1.14), the SFA [Eqs. (2.2)] [39, 154], the CCSFA [Eqs. (2.3)] [62] and the GC model [G₂ = (H₂, Φ_2), see Tab. 2.1]. The space of initial conditions is restricted to $p_{\parallel} = p_{z,0} = 0$, which is the most probable initial longitudinal and perpendicular momentum.

For the reference Hamiltonian (1.14) (upper panel of Fig. 3.6), we observe two grey regions of initial conditions where the electron final energy is negative, i.e., in which the electron is trapped in Rydberg states [127]. The color corresponds to the final energy of photoelectrons which have reached the detector. Enclosed by the grey regions, we observe that there are ionized electrons whose energy depends extremely sensitively on the initial conditions, as a signature of the rescattering process (see also the uppermost panel of Fig. 2.1). The boundaries of the grey regions are surrounded by regions of near-zero energy photoelectrons which ionize directly. The part of the grey region with small p_{\perp} (lower part of the left grey regions) is in a region where the ionization rate is high. As a consequence, a significant number of electrons which ionize directly reach the detector with near-zero energy, as observed in the leftmost panel of Fig. 3.5.

For the SFA [39, 154], the final momentum of the electron is given by its initial drift momentum $\mathbf{p}_0 - \mathbf{A}(t_0)$ since it is constant in time. As a consequence, the electron final energy is

$$\mathcal{E}^{\text{SFA}} = \frac{1}{2} \left| \mathbf{p}_0 - \mathbf{A}(t_0) \right|^2.$$
(3.3)

In the SFA (second panel from the top of Fig. 3.6), only two initial conditions lead to near zero-energy electrons, located at $p_{\perp} = -(E_0/\omega)\xi/\sqrt{\xi^2 + 1}$ and $\omega t_0 = \pi$, and at $p_{\perp} = -(E_0/\omega)/\sqrt{\xi^2 + 1}$ and $\omega t_0 = 3\pi/2$, represented by red dots in Fig 3.6. These initial conditions are located where the ionization rate is one or several orders of magnitude lower than the maximum ionization rate. The consequence is a lack of near-zero energy photoelectrons in the PMD for the SFA observed in Fig. 3.5.

For the CCSFA [62] (third panel from the top of Fig. 3.6), we observe the same patterns as for the SFA. The initial conditions of the near-zero energy photoelectrons which ionize directly for the CCSFA are located in the same region of low ionization rate as for the SFA. Here again, the consequence is the lack of near-zero energy electrons for the CCSFA observed in Fig. 3.5. However, we observe in the CCSFA a region with an abrupt change of energy with initial conditions across the dark colored path connecting the two red dots, absent in the SFA. This path is also present in the reference Hamiltonian (1.14). It is due to the rescattering process, i.e., the correction of the momentum of the CCSFA due to a recollision in the SFA (see second panel from the top of Fig. 2.1). Furthermore, this path separates near-zero energy photoelectrons induced by rescattering –responsible for the LES in the photoelectron energy spectra [24, 84]– from high energy photoelectrons. The CCSFA has been used to describe these short time-scale processes (see for instance Refs. [82, 84, 85, 114]).

The final energy of the electron using the GC model is given by

$$\mathcal{E} = \frac{1}{2} \left| \mathbf{p}_0 - \mathbf{A}(t_0) \right|^2 + V \left(\mathbf{r}_0 - \mathbf{E}(t_0) / \omega^2 \right).$$
(3.4)

The GC energy allows us to clearly distinguish two types of trajectories: Trajectories with $\mathcal{E} > 0$ and $\mathcal{E} < 0$. The set of initial conditions for which $\mathcal{E} < 0$ is referred to as the rescattering domain. Comparing the lowest panels of Fig. 2.1 and Fig. 3.6, this definition of the rescattering domain includes the Rydberg state creations, the Coulomb-driven recollisions and the subcycle recollisions most weighted by the PPT ionization rate. Notice that this definition misses a piece of the light purple band between the two rescattering domains in Fig. 2.1, which carries a lower weight according to the PPT ionization rate for all ellipticities. In the top panel of Fig. 3.6, we observe that the condition $\mathcal{E} < 0$ determines well the grey region of initial conditions in the reference Hamiltonian (1.14). The initial conditions for which the electron final energy is zero in the SFA are contained inside this region. The Coulomb potential creates this region in which the GC motion is bounded, which allows the electron to come back to the ionic core and to rescatter after multiple laser cycles, or to



Figure 3.6: Electron final energy as a function of the initial conditions $(t_0, p_{\perp}, p_{\parallel} = p_{z,0} = 0)$ for $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, Z = 1 and $\xi = 0.4$ of the reference Hamiltonian (1.14), the SFA [39, 154], the CCSFA [62] and the GC model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1). In grey-colored regions, the electron final energy is negative. The white dashed lines are contours of constant ionization rate $W_{\text{PPT}}(t_0, p_{\perp}\mathbf{e}_{\perp}(t_0))$ for $W_{\text{PPT}}/\max(W_{\text{PPT}}) = 10^{-1}$, 10^{-5} and 10^{-15} , from bottom to top. The red dots correspond to the initial conditions for which the electron final energy in the SFA is zero, i.e., $\mathcal{E}^{\text{SFA}} = 0$ [see Eq. (3.3)]. The solid black lines correspond to the initial conditions for which the GC energy is zero, i.e., $\mathcal{E} = 0$ [see Eq. (3.4)]. The black dashed line corresponds to the initial conditions for which the GC angular momentum is zero and the initial radial momentum is negative, i.e., $\mathbf{L} = \mathbf{0}$ and $p_r(t_0) < 0$, where $\mathbf{L} = \bar{\mathbf{r}} \times \bar{\mathbf{p}}$ and $p_r = \bar{\mathbf{p}} \cdot \bar{\mathbf{r}}/|\bar{\mathbf{r}}|$. The momentum and the energy are scaled by E_0/ω and $U_p = E_0^2/4\omega^2$, respectively.

be trapped into Rydberg states, scenarios analyzed in Sec. 3.2. The boundaries of this rescattering domain correspond to the initial conditions for which the electron final energy is zero, i.e., $\mathcal{E} = 0$. We observe that the inclusion of the Coulomb potential pushes down the near-zero-energy photoelectrons to regions in momentum space for which the ionization rate is higher. As a consequence, we observe a significant number of near-zero energy photoelectrons in the PMD of the GC model. Moreover, we notice that $\mathcal{E} = \mathcal{E}^{\text{SFA}} + V(\mathbf{r}_0 - \mathbf{E}(t_0)/\omega^2)$, and since the Coulomb potential is strictly negative, it is evident that electrons lose energy because of the Coulomb interaction, i.e., that electrons are subjected to Coulomb focusing.

3.1.2.3 Types of trajectories

In order to understand the origin of the sensitivity to initial conditions observed in the rescattering domain, we analyze the different types of trajectories. Figure 3.7a shows the scattering angle of the electron, whose trajectory is obtained from the reference Hamiltonian (1.14), as a function of the initial conditions (t_0, p_{\perp}) for $\xi = 0.4$. The scattering angle corresponds to the angle between momentum of the ionized electron \mathbf{p} at infinity and the major polarization axis (\mathbf{e}_x -axis). In Figs. 3.7b–e, the dark blue curves are the trajectories of the electron of Hamiltonian (1.14), with initial conditions indicated by the corresponding markers in Fig. 3.7a. The light blue curves are the GC trajectories of Hamiltonian $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1). For Figs. 3.7c–e (as well for Fig. 3.8d–e and Fig. 3.10d), the GC is initialized far from the ionic core (for $|\mathbf{r}| \gtrsim 2E_0/\omega^2$), during the plateau, in the domain of validity of the GC model (see Sec. 3.3.2.4 for a study of the discrepancy between the GC and the electron trajectory).

Figure 3.7b shows a subcycle recollision. The initial condition of this trajectory is near the condition for which the GC angular momentum is $\mathbf{L} = \bar{\mathbf{r}} \times \bar{\mathbf{p}} \approx \mathbf{0}$ and the initial GC radial momentum is negative, and corresponds to the light purple region in the uppermost panel of Fig. 2.1. Right after ionization, the GC trajectory is (mostly) straight, brings the electron to the core, and the electron recollides. The recollision occurs in a time scale shorter than one laser cycle, referred to as a subcycle recollision. We notice that if the electron tunnel-ionizes further away from the ionic core, the same conditions (near zero GC angular momentum and negative initial radial momentum) could lead to a multiple laser-cycle recollision. Looking at the third panel from the top of Fig. 2.1, we observe that this type of recollisions, for which $\bar{\mathbf{r}}(t_0) \cdot \bar{\mathbf{p}}(t_0) < \mathbf{0}$ and $\bar{\mathbf{r}}(t_0) \times \bar{\mathbf{p}}(t_0) \approx \mathbf{0}$, are well predicted by the CCSFA.

Figure 3.7c shows a *direct ionization*. The initial condition of this trajectory is in a regular region, for which the GC energy is positive $\mathcal{E} > 0$. The GC trajectory is unbounded, and leaves the ionic core region. The electron also leaves the ionic core region, driven by its GC.

Figure 3.7d shows a *Coulomb-driven recollision*. The initial condition of this trajectory is in one of the main chaotic regions, for which the GC energy is negative $\mathcal{E} < 0$, corresponding to the colored layers in the uppermost panel of Fig. 2.1. The GC trajectory is bounded. As a consequence, the electron returns to the ionic core, driven by its GC, and recollides with the ionic core. After rescattering, the GC energy jumps to another energy level (see Fig. 2.5), and the electron could ionize.

Figure 3.7e shows a *Rydberg state creation*. The initial condition of this trajectory is in the grey area, for which the GC energy is negative $\mathcal{E} < 0$. The GC trajectory is bounded. However, contrary to the Coulombdriven recollision (Fig. 3.7d), the laser pulse ends before the occurrence of the recollision. The Rydberg state creation corresponds to a frustrated Coulomb-driven recollision. The laser pulse duration plays an important role in determining the ratio between Coulomb-driven recollisions and Rydberg state trapping (see Sec. 3.2.1).

We observe that, in the four types of trajectories, two of them cannot be predicted by the SFA. While direct ionization and one-laser-cycle rescattering (Figs. 3.7c and 3.7b, respectively) are, at least qualitatively, predictable by the SFA, Coulomb-driven recollisions and Rydberg state creation (Figs. 3.7d and 3.7e, respectively) are predictable only when the Coulomb potential is taken into account. In the next section, we analyze Coulomb-driven recollisions and Rydberg state creation in more detail.

3.2 Coulomb-driven recollisions and Rydberg state creation

3.2.1 Ionization time dependence

Figures 3.8b and 3.8c show the final scattering angle of the ionized electron as a function of its initial conditions $(\omega t_0, p_{\perp})$, for an ionization that takes place at the beginning of the pulse and at the end of the pulse, respectively (see Fig. 3.8a). Figures 3.8d and 3.8e show two trajectories with the same initial momentum and the same laser phase, but with two distinct ionization times (separated by six laser cycles).



Figure 3.7: (a) Scattering angle of the electron as a function of the initial conditions after tunneling $(t_0, p_\perp, p_{\parallel} = p_{z,0} = 0)$ for $I = 8 \times 10^{13} \text{W} \cdot \text{cm}^{-2}$, Z = 1, and $\xi = 0.4$. The white dashed lines are the contours of constant ionization rate $W_{\text{PPT}}(t_0, p_\perp \mathbf{e}_\perp(t_0))$ for $W_{\text{PPT}}/\max(W_{\text{PPT}}) = 10^{-1}$, 10^{-5} and 10^{-15} , from bottom to top. The red dots correspond to the initial conditions for which $\mathcal{E}^{\text{SFA}} = 0$ [see Eq. (3.3)]. The solid black line corresponds to the initial conditions for which $\mathcal{E} = 0$ [boundaries of the rescattering domain for the GC model, see Eq. (3.4)]. The black dashed line corresponds to the initial conditions for which $\mathcal{E} = 0$ [and $p_r(t_0) < 0$, where $\mathbf{L} = \bar{\mathbf{r}} \times \bar{\mathbf{p}}$ and $p_r = \bar{\mathbf{p}} \cdot \bar{\mathbf{r}}/|\bar{\mathbf{r}}|$. Grey areas show the conditions for which the electron is trapped into Rydberg states. (b–e) Dark and light blue curves are the electron and its GC trajectory, respectively. The initial condition of each trajectory is associated with a marker represented in (a). These trajectories represent a typical: (b) subcycle recollision, (c) direct ionization, (d) Coulomb-driven recollision, and (e) Rydberg state creation. Panels (b) and (c) have positive GC energy, while (d) and (e) have negative GC energy. Blue shaded panels indicate the cases with recollisions. The momentum and position are scaled by E_0/ω and E_0/ω^2 , respectively.

Since the phase is the same, the GC trajectories (light blue curves) in Figs. 3.8d and 3.8e are the same. Since the GC energy of these trajectories is negative, the GC trajectory is bounded.

In Fig. 3.8d, for which the electron ionizes at the beginning of the plateau, we observe that the electron oscillates around the bounded GC trajectory, which drives the electron back to the ionic core. After about



Figure 3.8: (a) Electric field components and amplitude as a function of ωt . The grey regions indicate the ionization time for which the final scattering angle is computed in (b) and (c). (b–c) Final scattering angle of the ionized electron as a function of the initial conditions $(\omega t_0, p_\perp, p_{\parallel} = p_{z,0} = 0)$ for $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, Z = 1, and $\xi = 0.4$ for the reference Hamiltonian (1.14). The white dashed lines are the contour plot of the ionization rate $W_{\text{PPT}}(t_0, p_\perp \mathbf{e}_\perp(t_0))$ for $W_{\text{PPT}}/\max(W_{\text{PPT}}) = 10^{-1}$, 10^{-5} and 10^{-15} , from bottom to top. The solid black line corresponds to the initial conditions for which $\mathcal{E} = 0$ [boundaries of the rescattering domain for the GC model, see Eq. (3.4)]. The dark grey region corresponds to the initial conditions for which the electron is trapped into a Rydberg state at the end of the pulse. (d–e) Dark and light blue curves are the electron and its GC trajectory, respectively. The initial conditions of the trajectories in (d) and (e) are indicated by circles in (b) and (c), respectively. The trajectories in (d) and (e) are initialized at the same laser phase, but (d) is a Coulomb-driven recollision and (e) is a Rydberg state creation. The momentum and the position are scaled by E_0/ω and E_0/ω^2 , respectively.

four oscillations around the GC trajectory, the electron comes back to the ionic core. At this time, the GC energy jumps to another energy level due to the combined Coulomb and laser interaction, and the electron ionizes. This is a Coulomb-driven recollision.

In Fig. 3.8e, we observe that the electron oscillates as well around the bounded GC trajectory, which drives the electron back towards the core. However, when the electron is still far from the ionic core, the electric field is turned off, and the electron is trapped into a Rydberg state. The Rydberg state in which the electron is trapped corresponds almost to the Rydberg state of its GC.

In other words, for both trajectories of Fig. 3.8d and 3.8e, the electron oscillates around the same GC trajectory. The difference between these two trajectories is the remaining time $10T - t_0$ before the laser field is turned off. In Fig. 3.8d, the electron has enough time to undergo a close encounter with the ionic core $(|\mathbf{r}| < E_0/\omega^2)$ before the electric field is turned off, when in Fig. 3.8e, the electric field turns off sooner, while the electron is still far from the ionic core $(|\mathbf{r}| > E_0/\omega^2)$. The close encounter with the ionic core distinguishes the Coulomb-driven recollision from the Rydberg state creation. The scenarios of Coulomb-driven recollision and Rydberg state creation are closely related, since in both cases, the electron oscillates around a negative-energy GC.

Looking at the excursion time per laser cycle $\Delta t/T$ of the GC model depicted in the lowest panel of Fig. 2.1 and Fig. 3.9a, we observe similar layered patterns as for the reference Hamiltonian (1.14) (see uppermost panel of Fig. 2.1, Figs. 3.6a, 3.7a and 3.8b–c). These layered patterns correspond to trajectories which spend multiple laser cycles far from the origin before returning to the ionic core, such as the one depicted in Fig. 3.7d (Coulomb-driven recollision). Each layer is associated with a range of $\Delta t/T$ around an integer number, where, for decreasing ionization time for $\omega t_0 < \pi$, $\Delta t/T$ associated with each layer increases.

In order to picture roughly the conditions for which the Coulomb-driven recollisions occur, we first approximate the potential by a hard-Coulomb potential, i.e., $V(\bar{\mathbf{r}}) \approx -Z/|\bar{\mathbf{r}}|$, reducting this GC model to a Kepler problem (see also Appendix B). Then, we consider the period of the orbit per laser cycle $T_q/T = Z\omega/(2|\mathcal{E}|)^{3/2}$ (using $V(\bar{\mathbf{r}}) \approx -1/|\bar{\mathbf{r}}|$), where T_g is referred to as the GC orbit period in what follows. Figure 3.9b shows the period of the GC orbit per laser cycle $T_g/T = Z\omega/(2|\mathcal{E}|)^{3/2}$ as a function of the initial conditions in the largest rescattering domain. The grey regions correspond to the regions where the GC perihelion [see Eq. (2.25)] —the closest distance between the GC orbit and the ionic core- is greater than E_0/ω^2 or where $t_0 + T_g > T_f$. Figure 3.9c shows the GC distance from the ionic core $|\bar{\mathbf{r}}(t)|$ as a function of time per laser cycle of a sample of initial conditions indicated with the markers in Fig. 3.9b, and the distance from the ionic core $|\Pi(\Phi_2^{-1}(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t)))|$ of the corresponding reconstructed trajectories. We see that the color levels associated with the GC orbit period T_g agree well with the color levels associated with the excursion time Δt in Fig. 3.9a. Indeed, in Fig. 3.9c, we observe that the larger the period of the GC orbit followed by the electron, the larger its excursion time. As a consequence, the GC orbit period T_g is a good observable to estimate the excursion time of the electron Δt . In addition, the GC orbit period of the trajectory associated with the leftmost marker in Fig. 3.9b is such that $t_0 + T_g > T_f$. The electron does not undergo recollision and ends up trapped in a Rydberg state since it comes back to the ionic core after the end of the laser pulse. Therefore, electrons undergoing Coulomb-driven recollisions are typically driven by GC orbits such that $T_g < T_f - t_0$.



Figure 3.9: Recollisions in the GC model for $I = 8 \times 10^{13}$ W · cm⁻², $\xi = 0.4$ and Z = 1. (a) $\Delta t/T$ as a function of the initial conditions $(t_0, p_{\perp}, p_{\parallel} = p_{z,0} = 0)$ (Zoom of the lowest panel of Fig. 2.1 on the largest rescattering domain), where Δt is the smallest time interval such that $|\Phi_2^{-1}(\bar{\mathbf{r}}(t_0 + \Delta t), \bar{\mathbf{p}}(t_0 + \Delta t))| = 5$ a.u., with $t_0 + \Delta t < T_f$ (laser pulse duration $T_f = 10T$). The white and dark grey regions are where this condition is never met, and where the GC energy is positive (white region) and negative (grey region). (b) $T_g/T = \omega/(2|\mathcal{E}|)^{3/2}$ (see text). The dark grey region is where the GC perihelion [see Eq. (2.25)] is greater than the quiver radius $E_0/\omega^2 \approx 14$ a.u., or where $t_0 + T_g \geq T_f$. The inset is a zoom. The crosses are the location of the GC circular orbits (see Sec. 3.2.2). (a–b) The red dots and the black thick dashed curves are the same as in Fig. 3.6a. The dark dashed lines are contours of constant ionization rate $W_{\rm PPT}(t_0, p_{\perp}\mathbf{e}_{\perp}(t_0))$ for $W_{\rm PPT}/\max(W_{\rm PPT}) = 10^{-1}$, 10^{-5} and 10^{-15} , from bottom to top. (c) The lines with markers are the GC trajectories $|\bar{\mathbf{r}}(t)|$ with initial conditions plotted in panel (b) with the corresponding marker and color, and the light grey lines are the reconstructed trajectories $\Phi_2^{-1}(\bar{\mathbf{r}}(t), \bar{\mathbf{p}}(t))$. In the light grey region, the laser field is turned off. Momenta are scaled by E_0/ω .

Result 3: Coulomb-driven recollision and Rydberg state creation

Coulomb-driven recollisions and the Rydberg state creations are observed when the electron is driven by a GC with a negative energy $\mathcal{E} < 0$. The Coulomb-driven recollisions and the Rydberg state creation are in the rescattering domain.

Coulomb-driven recollision The electron is likely to undergo a Coulomb-driven recollision if it oscillates around a GC with a positive initial GC radial momentum $p_r(t_0) = \bar{\mathbf{p}}(t_0) \cdot \bar{\mathbf{r}}(t_0) |\bar{\mathbf{r}}(t_0)| > 0$, a GC orbital period such that $T_g = 2\pi Z/(2|\mathcal{E}|)^{3/2} < T_f - t_0$ and a GC perihelion smaller than the quiver radius. Notice that the condition that the perihelion of the GC orbit is smaller than the quiver radius is similar to $\mathbf{L} \approx \mathbf{0}$. As a consequence, all recollisions are likely driven by small absolute values of the GC angular momentum.

Rydberg state creation The electron is likely to be trapped in a Rydberg state if it oscillates around a GC with either an orbital period greater than the laser pulse duration $T_g > T_f - t_0$ or a perihelion greater than the quiver radius, i.e., a large GC angular momentum. Section 3.2.2 shows that the latter process is robust due to the existence of center-saddle periodic orbits which are weakly unstable.

3.2.2 Long plateau durations

In the lowest panel of Fig. 2.1 and Fig. 3.9a, we notice some grey regions in the upper and lower part of the rescattering domain for which the GC orbit period is such that $T_g < T_f - t_0$. However, in these regions, the electron does not recollide because the GC perihelion is large (greater than $E_0/\omega^2 \approx 14$ a.u.), as it is shown in Fig. 3.10a. As a consequence, there exists no time Δt such that $|\Pi(\Phi(\bar{\mathbf{r}}(t_0 + \Delta t), \bar{\mathbf{p}}(t_0 + \Delta t)))|$ is small, i.e., it is unlikely the electron recollides. This is also a scenario we observe in the reference model (1.14), in which the electron spins around the core for multiple laser cycles without recolliding.

For long plateau durations $(T_p = 100T, T_f = T_p + 2T)$ and an ionization time at the beginning of the laser pulse $(t_0 \ll T_p)$, we expect that electrons oscillating around a negative near-zero energy GC (for which the GC orbit period is such that $T_g > T_f - t_0$) and electrons with a large GC perihelion [see Eq. (2.25)] (GC perihelion greater than E_0/ω^2 that prevents the electron from rescattering) create Rydberg states. In Fig. 3.10b, we observe indeed a pink thin layer of electrons creating Rydberg states, with a near-zero-energy GC such as the dark blue trajectory depicted in Fig. 3.10d. In addition, we observe two regions of initial conditions with smaller values of final energy for which the electrons are trapped in Rydberg states after having remained in the vicinity of the ionic core, for which the GC perihelion is larger than the quiver radius, as shown for the dark blue trajectory of the reference Hamiltonian in Fig. 3.10e. However, by comparing Fig. 3.10a and Fig. 3.10b, we observe that not all the electrons with a GC perihelion larger than the quiver radius are captured into Rydberg states. Here, we show how some electrons remain trapped while others do not.

As observed in Fig. 3.10c, the filled region of initial conditions leading to electrons trapped in Rydberg states with a large GC perihelion is roughly regular. Figure 3.10e shows in dark blue a typical trajectory of Hamiltonian (1.14) initiated inside this regular region. We observe that this trajectory turns around the core multiple times without being rescattered by the ionic core. As a consequence, the GC energy of this electron remains negative and roughly constant throughout the laser pulse duration. When the laser field is turned off, its GC energy is still negative and the electron is trapped in a Rydberg state. Near the initial conditions of this trajectory, there is a center-saddle periodic orbit of the reference model (1.14) which exhibits the same pattern as this trajectory. This center-saddle periodic orbit is depicted in thick dark blue in Fig. 3.10e. In its neighborhood, the periodic orbit is center in one plane and saddle in a transverse plane defined by the eigenvectors of the monodromy matrix associated with the complex and real eigenvalues, respectively. Hence, there are two-dimensional invariant tori surrounding the periodic orbit in the center direction. The saddle direction is weakly unstable (its eigenvalue is ~ 1.4) and the orbit period is large (period of 30*T*), which implies that the unstable direction pushes slowly the electron away from each invariant torus. Consequently, trajectories in the vicinity of this periodic orbit remain close to it for relatively long times, even for long laser pulses.



Figure 3.10: The parameters are $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, $\xi = 0.4$, Z = 1 and plateau duration $T_p = 100T$. (a) GC perihelion [see Eq. (2.25)] in the rescattering domain depicted in the space of initial conditions $(t_0, p_{\perp}, p_{\parallel} = p_{z,0} = 0)$. (b) Final negative energies of the electron trajectories of the reference Hamiltonian (1.14), where (c) is a zoom around the trapping region. The white color in (a–c) denotes an electron not trapped at the end of the laser pulse. (d),(e) Trajectories for the initial conditions indicated with a diamond in (c) and a triangle in (b), respectively. The dark blue and cyan curves are the electron trajectory of the reference Hamiltonian (1.14) and the GC trajectory, respectively. The thick dark blue curve in (e) is a center-saddle periodic orbit very close to the region depicted in (c). The cyan crosses in (a) and (b) are the initial conditions of the clockwise (upper cross) and anticlockwise (lower cross) circular GC orbits. The cyan curve in (e) is the GC clockwise circular orbit, whose initial conditions are very close to the trapping region depicted in (c). Momenta and distance are scaled by E_0/ω and E_0/ω^2 , respectively.



Figure 3.11: Rydberg state creation (RSC) probability as a function of the laser ellipticity ξ for $I = 8 \times 10^{13}$, 3×10^{14} , and 8×10^{14} W \cdot cm⁻², and Z = 1. The RSC probability is defined as the ratio of the RSC yield \mathbb{Y} to the ionized electron yield $N = \int_0^{T_f} dt_0 \int_{-\infty}^{\infty} d^3 \mathbf{p}_{ADK} W_{PPT}(t_0, \mathbf{p}_{PPT})$. The thick solid and dotted curves are our prediction with the GC model \mathbb{Y}_{GC}/N [Eq. (3.7)], and the SFA \mathbb{Y}_{SFA}/N , respectively. The thin curves with stars and crosses are the CTMC simulations of the reference Hamiltonian (1.14) using $T_p = T$ and $T_p = 8T$, respectively. The filled areas show the estimate of the Coulomb-driven recollision probability for $T_p = 8T$. Red circles show the probability of RSC at the critical ellipticity ξ_c given by Eq. (3.13). Left panel: Only \mathbb{Y}_{SFA} is normalized such that it agrees at $\xi = 0$ with the CTMC simulations of the reference Hamiltonian (1.14) for $T_p = T$ (thin lines with star markers). Right panel: All predictions are normalized such that it is one at $\xi = 0$.

In Fig. 3.10a, we observe that when the GC perihelion is large (greater than E_0/ω^2), the recollisions are unlikely to happen as mentioned earlier. In these two regions of large GC perihelion, there are two cyan crosses indicating the initial conditions for which the GC orbit is circular. The initial conditions of these circular orbits are $p_{\perp} = \mathbf{A}(t_0) \cdot \mathbf{e}_{\perp}(t_0) \pm Z\omega^2/|\mathbf{E}(t_0)|\cosh\tau_0(t_0)$ with $\omega t_0 = n\pi$ and $n \in \mathbb{N}$. They are close to the regular region in Figs. 3.10b–c. The circular orbit of the GC is depicted in cyan in Fig. 3.10e. We observe that the cyan curve provides the leading behavior of the averaged trajectory of the center-saddle periodic orbit in thick dark blue. The energy of the GC circular orbits (clockwise and anticlockwise) is given by $\mathcal{E} = -Z\omega^2\sqrt{\xi^2 + 1}/(2E_0\cosh\tau)$ and their perihelion by $Z/(2|\mathcal{E}|)$.

In summary, there is a region of initial conditions for which the GC perihelion is larger than the quiver radius E_0/ω^2 , preventing the electron to recollide with the core. Instead, the electron is trapped in a Rydberg state. We showed that this process is robust because in the neighborhood of these initial conditions, there are center-saddle periodic orbits with weakly unstable directions that keep the electron in the vicinity of the core.

3.2.3 Rate of Rydberg state creation

Next, we investigate the rate of Rydberg state creation as a function of the laser ellipticity. A Rydberg state is created if the electron energy is negative at the end of the laser pulse. In the SFA, the condition of Rydberg state creation $\mathcal{E}^{SFA} = 0$ [see Eq. (3.3)] is a one-dimensional curve $(t_0, \mathbf{p}_0^*(t_0))$ in a four-dimensional space (t_0, \mathbf{p}_{PPT}) , with $\mathbf{p}_{PPT}^*(t_0) = \mathbf{A}(t_0)$. As a consequence, the probability of Rydberg state creation is in fact zero. In Refs. [127, 96], the yield of Rydberg state creation is given by $\mathbb{Y}_{SFA} = \int_0^{T_f} dt_0 W_{PPT}(t_0, \mathbf{p}_0^*(t_0))$.

Figure 3.11 shows the Rydberg state creation probability as a function of the laser ellipticity from CTMC simulations of the reference Hamiltonian (1.14) (thin solid curves with markers) and the SFA prediction (dotted curves) $\mathbb{Y}_{\text{SFA}}/N$ with $N = \int_0^{T_f} dt_0 \int_{-\infty}^{\infty} d^3 \mathbf{p}_{\text{PPT}} W_{\text{PPT}}(t_0, \mathbf{p}_{\text{PPT}})$ the yield of ionized electrons. In Ref. [96], The SFA prediction is normalized such that it agrees at $\xi = 0$ with the CTMC simulations of the reference Hamiltonian (1.14) for $T_p = T$ (thin lines with star markers). Notice that only the prediction of the SFA is artificially normalized. For the SFA prediction (dotted curves), we observe a good agreement with the reference model at high ellipticity for all intensities and at low ellipticity for high intensity. However, there is a large discrepancy at low ellipticity for low and intermediate intensities, i.e., for $I \leq 5 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. For such intensities, the rescattering domain where Rydberg states arise is wide compared to the gradient of the ionization rate as observed in the top panel of Fig. 3.6 and Fig. 3.7a. As a consequence, the SFA prediction that Rydberg states arise from the center of the rescattering domain is not accurate.

On the contrary, in Fig. 3.6, we see that the GC model is a good approximation for evaluating the size

of the rescattering domain where the Rydberg states are created. In the GC model, a Rydberg state can be created only if the GC energy is negative $\mathcal{E} < 0$. As an approximation, we neglect the cases for which the electron undergoes a Coulomb-driven recollision according to the GC model.

Result 4: Rate of Rydberg state creation

The GC prediction of the yield of Rydberg state creation is given by

$$\mathbb{Y}_{\mathrm{GC}} = \int_{\Omega_R} W(t_0, \mathbf{p}_0) \,\mathrm{d}t_0 \mathrm{d}^3 \mathbf{p}_0,\tag{3.5}$$

where $\Omega_R = \{t_0 \in [0, T_f], \mathbf{p}_0 \in \mathbb{R}^3 \mid \mathcal{E} < 0\}$ is the set of initial conditions such that the GC energy \mathcal{E} is negative [see Eq. (3.4)], and $W(t_0, \mathbf{p}_0)$ is the ionization rate as a function of the initial conditions.

According to Sec. 3.2.1, an electron populating the rescattering domain either undergoes a Coulomb-driven recollision or is trapped in a Rydberg state. In order to minimize Coulomb-driven recollisions, we compare the GC prediction with CTMC simulations of the reference model for $T_p = T$. Figure 3.11 shows the GC prediction of Rydberg state creation probability (solid curves) \mathbb{Y}_{GC}/N . We observe an excellent agreement between the results of the simulation of the reference model (1.14) for $T_p = T$ and the GC prediction for all ellipticities and intensities plotted here. In particular, on the right panel of Fig. 3.11, where all the predictions are normalized to one at $\xi = 0$, we observe that the solid curves and the stars overlap. For increasing intensity, the volume of the rescattering domain decreases, as shown in the next section. Hence, at high intensity, the ionization rate varies on large scales compared to the size of the rescattering domain, and the ionization rate is almost constant in the rescattering domain. Therefore, for high intensity, $\mathbb{Y}_{SFA} \propto \mathbb{Y}_{GC}$ as we observe in Fig. 3.11 for $I = 8 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$.

3.3 The shape of the rescattering domain and its experimental implications

3.3.1 Analysis of the shape of the rescattering domain

After ionization, the GC energy of the electron is given by Eq. (3.4). Substituting $\mathbf{A}(t_0) = [\mathbf{A}(t_0) \cdot \mathbf{e}_{\parallel}(t_0)]\mathbf{e}_{\parallel}(t_0) + [\mathbf{A}(t_0) \cdot \mathbf{e}_{\perp}(t_0)]\mathbf{e}_{\perp}(t_0)$ in Eq. (3.4), the rescattering domain defined by $\mathcal{E} < 0$ is the ensemble of initial conditions $(t_0, \mathbf{p}_{\text{PPT}})$ such that

$$\left[p_{\parallel} - p_{\parallel}^{\star}(t_0)\right]^2 + \left[p_{\perp} - p_{\perp}^{\star}(t_0)\right]^2 + p_{z,0}^2 < \Delta p(t_0)^2, \tag{3.6}$$

where $\Delta p(t_0) = \sqrt{2|V(\bar{\mathbf{r}}(t_0))|}$, $\mathbf{p}_0^{\star}(t_0) = p_{\parallel}^{\star}(t_0)\mathbf{e}_{\parallel}(t_0) + p_{\perp}^{\star}(t_0)\mathbf{e}_{\perp}(t_0)$, hence $p_{\parallel}^{\star}(t_0) = \mathbf{A}(t_0) \cdot \mathbf{e}_{\parallel}(t_0)$ and $p_{\perp}^{\star}(t_0) = \mathbf{A}(t_0) \cdot \mathbf{e}_{\perp}(t_0)$. Here, $\bar{\mathbf{r}}(t_0) = -\mathbf{E}(t_0)\cosh\tau_0(t_0)/\omega^2$ [see Eqs. (2.18) and (1.25)]. For a given ionization time t_0 , the rescattering domain is a sphere centered at $\mathbf{p}^{\star}(t_0)$ and of radius $\Delta p(t_0)$ in momentum space. The yield of Rydberg state creation in the GC model [see Eq. (3.5)] becomes

$$\mathbb{Y}_{\rm GC} = \int_0^{T_f} \mathrm{d}t_0 \ \Delta p(t_0)^3 \int_0^1 \mathrm{d}\rho \ \rho^2 \int_0^{2\pi} \mathrm{d}\vartheta \int_0^{\pi} \mathrm{d}\theta \ W_{\rm PPT}(t_0, \mathbf{p}_0^{\star}(t_0) + \rho \Delta p(t_0)\mathbf{e}(t_0, \vartheta, \theta)) \sin \theta, \tag{3.7}$$

where $\mathbf{e}(t_0, \vartheta, \theta) = \mathbf{e}_{\parallel}(t_0) \cos \vartheta \sin \theta + \mathbf{e}_{\perp}(t_0) \sin \vartheta \sin \theta + \mathbf{e}_z \cos \theta$. Equation (3.7) is used to compute the yield of Rydberg state creation of Fig. 3.11, and the integrals are performed numerically.

Figures 3.12a and 3.12b show the boundaries of the rescattering domain in the space $(t_0, p_{\parallel}, p_{\perp}, p_{z,0} = 0)$ for $\xi = 0.2$ and $\xi = 0.7$. To see how the shape of the rescattering domain evolves as a function of the parameters, we focus on the conditions $p_{\parallel} = p_{z,0} = 0$ for which the ionization rate is maximum. For low ellipticity, the surface $p_{\parallel} = p_{z,0} = 0$ and the rescattering domain intersect in approximately ellipsoidal subdomains, while for high ellipticity, they intersect in a band.

3.3.1.1 Close to LP

First, we consider the second order Taylor expansion of the shape of the rescattering domain for $p_{\parallel} = p_{z,0} = 0$ as a function of the ellipticity in the plane (t_0, p_{\perp}) close to LP $(\xi \ll 1)$. For low ellipticity, the rescattering domain is approximately a set of ellipses, with two subsets: ellipses at the peak laser amplitude [around



Figure 3.12: Shape of the rescattering domain for $I = 8 \times 10^{13}$ W \cdot cm⁻², Z = 1 and $\xi = 0.2$ [close to LP, (a) and (c)], and $\xi = 0.7$ [close to CP, (b) and (d)]. (a), (b) Boundary of the rescattering domain as a function of the initial conditions $(t_0, p_{\parallel}, p_{\perp}, p_{z,0} = 0)$. The color is the logarithm of the PPT ionization rate normalized by its maximum. The black lines are the boundaries of the rescattering domain for $p_{\parallel} = p_{z,0} = 0$. (c),(d) Slice of the initial conditions $p_{\parallel} = p_{z,0} = 0$ [shaded blue planes in (a) and (b)]. Only the dominant orders in ξ are depicted. Momenta are scaled by E_0/ω .

 $\omega t_0 = n\pi$, with $n \in \mathbb{N}$, and ellipses at the lowest laser amplitude [around $\omega t_0 = (n + 1/2)\pi$].

For $p_{\parallel} = p_{z,0} = 0$, the local minima of the final electron energy [see Eq. (3.4)] are located at $\omega t_0^{\star} = n\pi/2$ and $p_{\perp}^{\star} = p_{\perp}^{\star}(t_0^{\star})$ for $n \in \mathbb{N}$. The local minima of the GC energy are the red dots depicted in Fig. 3.12c. In Eq. (3.6), we fix $p_{\parallel} = p_{z,0} = 0$ and we Taylor expand with respect to $t_0 - t_0^{\star}$. We obtain that the rescattering domain for $p_{\parallel} = p_{z,0} = 0$ can be written in the form

$$\frac{(p_{\perp} - p_{\perp}^{\star})^2}{\Delta p_{\perp}^2} + \frac{(t_0 - t_0^{\star})^2}{\Delta t_0^2} < 1,$$
(3.8)

where terms of order $(t_0 - t_0^*)^4$ and higher are neglected. Consequently, the subsets of rescattering domain in the plane (t_0, p_{\perp}) defined by $p_{\parallel} = p_{z,0} = 0$ are approximately ellipses and are centered around the local minima of the GC energy (t_0^*, p_{\perp}^*) . The expressions for p_{\perp}^* , Δp_{\perp} and Δt_0 depend on whether the ellipse is at the peak laser amplitude or at the lowest laser amplitude.

Rescattering domains at the lowest laser amplitude: After Taylor expanding Eq. (3.6) with respect to t_0 and ξ around the local minima $\omega t_0^{\star} = (n + 1/2)\pi$ and $\xi = 0$, respectively, one gets (at the third order in the Taylor expansion) $p_{\perp}^{\star} \approx (E_0/\omega)(1 - \xi^2/2)$,

$$\Delta p_{\perp} \approx (E_0/\omega)\xi_c c_{\gamma} \left(1 - \frac{\xi^2}{4\gamma^2}\right),$$

$$\omega \Delta t_0 \approx \xi \xi_c c_{\gamma},$$

where $c_{\gamma} = \sqrt{\gamma} (1 + \gamma^2)^{1/4} / \sinh^{-1} \gamma$, ξ_c is defined in Eq. (3.13), and we have used $\tau \approx \sinh^{-1} \gamma$. Hence, at low ellipticities, the area of these ellipses is proportional to ξ and consequently very small. For LP, the area of these ellipses is zero. In addition, at low ellipticities, these ellipses have a low weight given by the ionization rate, so their influence is negligible.

Rescattering domains at the peak laser amplitude: After Taylor expanding Eq. (3.6) with respect to t_0 and ξ around the local minima $\omega t_0^* = n\pi$ and $\xi = 0$, respectively, one gets (at the third order in the Taylor expansion) $p_{\perp}^* \approx \xi(E_0/\omega)(1-\xi^2/2)$,

$$\Delta p_{\perp} \approx (E_0/\omega)\xi_c C_{\gamma} \left[1 + \frac{\xi^2}{4(1+\gamma^2)}\right], \qquad (3.9a)$$

$$\omega \Delta t_0 \approx \xi_c C_\gamma \left[1 + \xi^2 \frac{7 + 6\gamma^2}{4(1 + \gamma^2)} \right], \qquad (3.9b)$$

where $C_{\gamma} = \gamma/\sinh^{-1}\gamma$ and we have used $\tau \approx \sinh^{-1}\gamma$. Here, the area of the ellipses is non-zero for LP, and because these ellipses are highly weighted by the ionization rate, they have a strong influence in the phenomena related to the rescattering domain such as, for instance, Rydberg state creation. We observe that for increasing intensity, these elliptical domains shrink towards their centers for which the GC energy is minimal (red dots in Fig. 3.12), which correspond also to the SFA conditions for which the electron final energy is zero [see Eq. (3.3)]. The Z-dependence in Δp_{\perp} and $\omega \Delta t_0$ is in ξ_c only, at first orders in the ellipticity. The area of the ellipse increases linearly for increasing Z.

3.3.1.2 Close to CP

Next, we consider the second order Taylor expansion of the shape of the rescattering domain for $p_{\parallel} = p_{z,0} = 0$ as a function of the ellipticity in the plane (t_0, p_{\perp}) close to CP $(1 - |\xi| \ll 1)$. For ellipticity close to 1, the rescattering domain is approximately a band between two lines. We write Eq. (3.6) in the form

$$p_{\perp}^{-}(t_0) < p_{\perp} < p_{\perp}^{+}(t_0),$$

with $p_{\perp}^{\pm}(t_0) = p_{\perp}^{\star}(t_0) \pm [p_{\parallel}^{\star}(t_0)^2 + \Delta p(t_0)^2]^{1/2}$. By Taylor expanding this expression to the first order (the second and third order expansions are too lengthy and do not provide additional relevant information to the discussion) with respect to $1 - |\xi|$ around $\xi = 1$, one gets that the lines surrounding the rescattering domain are

$$p_{\perp}^{\pm}(t_0) \approx \frac{E_0}{\sqrt{2\omega}} \left[\cos(2\omega t_0) \frac{\xi - 1}{2} + 1 \pm \xi_1 \right],$$
 (3.10)



Figure 3.13: Photoelectron momentum distribution along the minor polarization axis \mathbf{e}_y as a function of the ellipticity for $I = 1.2 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$ and Ar $(I_p = 0.58 \text{ a.u.} \text{ and } Z = 1)$ and $\gamma \sim 1$. The color scale is the experimental data of Ref. [104]. The dotted and dashed black lines are the T-trajectory of the SFA and the CCSFA, respectively. The cross markers and red solid lines are the T-trajectory of the reference Hamiltonian (1.14) and the GC model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1), respectively. Momenta are scaled by E_0/ω .

where $\xi_1 = (\sqrt{Z}\omega^2/E_0^{3/2})(\gamma^2 + 1/2)^{-1/4}$ and we have used $\tau \approx \sinh^{-1}\sqrt{2}\gamma$. Hence, Coulomb-driven recollisions and Rydberg state creation after tunneling are likely when the lowest boundary line of the rescattering domain (see Fig. 3.12) approaches the regions of initial conditions with high ionization rate, i.e., $p_{\perp}^-(t_0) \lesssim \mathbf{p}_{\text{ADK}}^T \cdot \mathbf{e}_{\perp}(t_0)$. Fixing $\xi = 1$ and using Eq. (3.10), one gets

$$E_0^{3/2} \lesssim \sqrt{Z}\omega^2 \frac{\sinh^{-1}(\sqrt{2\gamma})}{\sqrt{2\gamma(\gamma^2 + 1/2)^{1/4}}}.$$

The term on the right-hand side of the inequality decreases for increasing γ . For $\gamma \ll 1$, the inequality becomes $I \leq 2\sqrt{Z} \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$. However, the condition $\gamma \ll 1$ implies that $I_p \ll 0.1$ a.u. in order for the electron to undergo a Coulomb-driven recollisions or be trapped in a Rydberg state at this frequency. We observe it is unlikely that the electron undergoes a Coulomb-driven recollision or is trapped in a Rydberg state for nearly-CP pulses, if the ionization takes place during the plateau. Most probably, this electron undergoes direct ionization.

3.3.2 Implication of the shape of the rescattering domain

In this section, we investigate the physical phenomena related to the shape of the rescattering domain, and we compare the results with experimental data. For instance, when the laser ellipiticity ξ varies, the rescattering domain moves in phase space and as a consequence the PMDs change shape. In Fig. 3.13, we show the experimental measurements from Ref. [104], of the final momentum distribution of the electron along the minor polarization axis \mathbf{e}_y as a function of the ellipticity ξ for $I = 1.2 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, Ar ($I_p = 0.58 \text{ a.u.}$) and $\gamma \sim 1$. The experimental measurements of the final momentum along the minor polarization axis (color scale) show a distribution peaked around zero for small ellipticity. As the ellipticity increases, we observe a bifurcation of the peak of the distribution at a critical ellipticity $\xi_c \approx 0.25$, for which the distribution is no longer peaked around zero. After the bifurcation (for $\xi > \xi_c$), the peaks of the distribution move further apart for increasing ellipticity. In Ref. [104], a semiclassical theory is developed and is in agreement with the experimental measurements. It is also shown that the initial conditions of the most probable trajectory of the theory in Ref. [104] are relatively close to the initial conditions of the most probable trajectory in PPT. Here, we show that this bifurcation can be reproduced and understood by the analysis of the T-trajectory only.

In Fig. 3.13, we also show the \mathbf{e}_y -component of the T-trajectory final momentum P_y computed using the SFA $\mathbf{P}^{\text{SFA}} = \mathbf{e}_y \xi(E_0/\omega) \sinh \tau / (\tau \sqrt{\xi^2 + 1})$ (dotted lines), the CCSFA from Eq. (2.3) (dashed lines), the

reference Hamiltonian (1.14) (crosses) and the GC prediction [see Eq. (3.12)] (solid lines). The prediction of the reference Hamiltonian (1.14) is depicted only if the ionization is direct, i.e., if it has not undergone any recollisions and has not been trapped in Rydberg states. The GC prediction is depicted only when the GC energy is positive. Otherwise, the GC energy is negative and the electron does not reach the detector according to the GC model. We observe an excellent agreement between the experimental results from Ref. [104], the reference Hamiltonian [Hamiltonian (1.14)] and the GC prediction.

In a nutshell, for $\xi < \xi_c$, the T-trajectory is inside the rescattering domain. The GC motion is most often bounded, and as a consequence the electron undergoes recollisions or is trapped in a Rydberg state. When the ellipticity increases, the rescattering domain and the initial conditions of the T-trajectory change. At the critical ellipticity ξ_c , the T-trajectory is on the boundary of the rescattering domain, i.e., its GC energy is zero. For $\xi > \xi_c$, the GC motion is unbounded, and the electron ionizes without recollision. Therefore, the bunches in the PMDs after the bifurcation (as observed in Fig. 3.5) are mainly composed of direct ionizations. Right after the bifurcation, a ridge structure can be seen for a certain range of laser parameters and atoms [114, 114]. The ridge structure is composed of near-zero-energy electrons induced by rescattering, and the bifurcation with ellipticity can be used to isolate these electrons from the electrons ionized directly [114, 43].

3.3.2.1 Critical ionization time

In LP fields, for $p_{\perp} = p_{z,0} = 0$ which reduces to a one-dimensional (d = 1) model, the SFA predicts that if an electron ionizes after a peak laser amplitude, i.e., at $t_0 > t_0^*$ ($\omega t_0^* = n\pi$ where $n \in \mathbb{N}$), it undergoes a recollision [39], while if it ionizes before this peak, i.e., at $t_0 < t_0^*$, it ionizes directly. In the top panel of Fig. 3.6 and in Figs. 3.7a and 3.8a–b, we observe that this critical time $\omega t_0 = n\pi$ predicted by the SFA is lower if the Coulomb potential is taken into account, and according to the discussion in Sec. 3.2.1, the electron potentially comes back to the ionic core even if it ionizes before the peak of the laser field.

According to the GC model, using Eqs. (3.8) and (3.9) for $p_{\perp} = p_{z,0} = 0$, the left boundaries of the rescattering domain are given by $\omega t_c = n\pi - \xi_c C_{\gamma}$. If the electron ionizes at $t_0 < t_c$, the electron ionizes directly. If the electron ionizes at $t_0 > t_c$, the electron is in the rescattering domain. According to the discussion in Sec. 3.2, the electron either populates Rydberg states or undergoes a recollision. In particular, if an electron ionizes before the peak of the laser field and recollides, it is mainly because of the Coulomb interaction and the bounded motion of its GC that brings the electron back to the core. If the electron ionizes after the peak of the laser field, its GC initial radial momentum is negative (and its angular momentum is zero for d = 1), and as a consequence the electron recollides.

The same arguments are extended to estimate t_c for low ellipticity and $\xi \leq \xi_c$. We fix the initial momentum at its most probable value given by $(p_{\parallel} = \mathbf{p}_{ADK}^T \cdot \mathbf{e}_{\parallel}(t_0), p_{\perp} = \mathbf{p}_{ADK}^T \cdot \mathbf{e}_{\perp}(t_0), p_{z,0} = \mathbf{p}_{ADK}^T \cdot \mathbf{e}_z)$ and we let the ionization time t_0 free. At low ellipticity $\mathbf{p}_{ADK}^T \approx \mathbf{0}$, and if $\omega t_0 = \omega t_0^*$ the trajectory is approximately at the center of the rescattering domain (see Fig. 3.12c). As a consequence, there exist intervals of ionization time t_0 for which the initial conditions are inside the rescattering domain, but also because of the shape of the rescattering domain (see Fig. 3.12c), there are intervals of ionization times t_0 for which the initial conditions are outside the rescattering domain. The critical time t_c is the ionization time for which $(t_c, \mathbf{p}_{ADK}^T)$ is on the left boundary of the rescattering domain. In Eq. (3.8), we transform the inequality into an equality and we fix $p_{\perp} = \mathbf{p}_{ADK}^T \cdot \mathbf{e}_{\perp}(t_0) \approx \xi(E_0/\omega)(1 - \gamma/\sinh^{-1}\gamma)$ [see Eq. (1.32)]. Then, using Eqs. (3.9) up to the second order in ξ , the critical time t_c is given by

$$\omega t_c \approx \omega t_0^\star - C_\gamma \sqrt{\xi_c^2 - \xi^2},$$

with $C_{\gamma} = \gamma/\sinh^{-1}\gamma$ and ξ_c defined Eq. (3.13). Also, we have seen in Sec. 3.2 that if the ionization takes place after the peak of the laser field, the GC radial momentum is negative and the electron tends to recollide with the ionic core. Hence, the ionization time t_0 for direct ionization is $\omega t_0 \in \omega t_c - [0, \pi/2]$. This is in agreement with the CTMC simulations of Ref. [105].

3.3.2.2 Critical ellipticity

Next, we consider the bifurcation with respect to the laser parameters. We consider the T-trajectory given by the initial conditions (1.32) and $\omega t_0 = \pi$. For LP ($\xi = 0$), the T-trajectory is inside the rescattering domain. As a consequence, the GC energy of the T-trajectory is negative, and the electron is either trapped in a Rydberg state, or undergoes a recollision. For increasing ellipticity, $p_{\perp}^{\star}(t_0)$ increases and $\mathbf{p}_{\text{ADK}}^T \cdot \mathbf{e}_{\perp}(t_0)$ decreases. At ellipticity $\xi = \xi_c$, the initial condition of the T-trajectory ($\omega t_0 = \pi, \mathbf{p}_{\text{ADK}}^T$) crosses the boundary of the rescattering domain. For $\xi > \xi_c$, the T-trajectory is outside the rescattering domain, and the electron ionizes without recolliding. The initial condition of the GC of the T-trajectory is determined by combining Eqs. (2.18) and Eqs. (1.32), and reads

$$\bar{\mathbf{r}}_{\text{PPT}}^T = \mathbf{e}_x \frac{E_0}{\omega^2 \sqrt{\xi^2 + 1}} \cosh \tau, \quad \bar{\mathbf{p}}_{\text{PPT}}^T = \mathbf{e}_y \frac{\xi E_0}{\omega \sqrt{\xi^2 + 1}} \frac{\sinh \tau}{\tau}.$$

Since Hamiltonian H_2 is time-independent and rotationally invariant, the GC energy \mathcal{E}_T and angular momentum $\ell_T = \mathbf{e}_z \cdot \bar{\mathbf{r}}_{\text{PPT}}^T \times \bar{\mathbf{p}}_{\text{PPT}}^T$ of the T-trajectory are conserved and given by

$$\mathcal{E}_T = \frac{\xi^2 E_0^2}{2\omega^2 (\xi^2 + 1)} \frac{\sinh^2 \tau}{\tau^2} - \frac{Z\omega^2 \sqrt{\xi^2 + 1}}{E_0 \cosh \tau}, \qquad \ell_T = \frac{\xi E_0^2}{\omega^3 (\xi^2 + 1)} \frac{\sinh 2\tau}{2\tau}, \tag{3.11}$$

with $V(\bar{\mathbf{r}}_{PPT}^T) \approx -Z/|\bar{\mathbf{r}}_{PPT}^T|$. When the electric field is turned off, we assume that the final momentum of the T-trajectory and the final momentum of its GC are equal, with $P_x = \sqrt{2\mathcal{E}_T} \cos \Theta$ and $P_y = \sqrt{2\mathcal{E}_T} \sin \Theta$, where its scattering angle is given by [see Eq. (2.24)]

$$\Theta = \pi/2 + \sin^{-1} \left(2\mathcal{E}_T \ell_T^2 / Z^2 + 1 \right)^{-1/2}.$$

As a consequence,

$$P_x = -\sqrt{2\mathcal{E}_T} \left(2\mathcal{E}_T \ell_T^2 / Z^2 + 1 \right)^{-1/2}, \qquad (3.12a)$$

$$P_y = \sqrt{2\mathcal{E}_T} \left[1 - \left(2\mathcal{E}_T \ell_T^2 / Z^2 + 1 \right)^{-1} \right]^{1/2}.$$
 (3.12b)

Equations (3.12) are used to compute P_x and P_y of the GC.

Result 5: Bifurcation in the PMDs

The energy of the GC of the electron after ionization is given in Eq. (3.11). There exists a bifurcation at $\xi = \xi_c$ such that $\mathcal{E}_T = 0$. Assuming that $\xi_c \ll 1$, the critical ellipticity is

$$\xi_c \approx \frac{\sqrt{2Z\omega^2}}{E_0^{3/2}} \frac{\sinh^{-1}\gamma}{\gamma(1+\gamma^2)^{1/4}}.$$
(3.13)

The critical ellipticity ξ_c indicates the drop-off of the recollision probability as a function of the ellipticity.

- If $\xi < \xi_c$, the energy of the GC of the T-trajectory is negative, the GC motion is bounded, and the electron is trapped in a Rydberg states or undergoes recollision.
- If $\xi > \xi_c$, the energy of the GC of the T-trajectory is positive, the GC motion is unbounded, and the electron ionizes without recolliding.

Close to the bifurcation, for $\xi \approx \xi_c$ and using $\tau \approx \sinh^{-1} \gamma$,

$$\mathcal{E}_T \approx (\xi - \xi_c) 4 \mathrm{U}_p \xi_c \gamma^2 / (\sinh^{-1} \gamma)^2, \qquad (3.14a)$$

$$P_x \approx -(\xi - \xi_c)^{1/2} \sqrt{2\xi_c} (E_0/\omega) (\gamma/\sinh^{-1}\gamma),$$
 (3.14b)

$$P_{y} \approx (\xi - \xi_{c}) 2\sqrt{2} (E_{0}/\omega) (\gamma/\sinh^{-1}\gamma). \qquad (3.14c)$$

The critical exponents of the bifurcation predicted by the GC model for P_x and P_y are 0.5 and 1, respectively, i.e., $P_x \sim (\xi - \xi_c)^{1/2}$ and $P_y \sim (\xi - \xi_c)$. In the PMDs, the bifurcation in P_x signals the appearance of Coulomb asymmetry as a function of the ellipticity, while the bifurcation in P_y shows the breakdown of Coulomb focusing as a function of the ellipticity. We observe that Coulomb asymmetry appears at the same time as Coulomb focusing begins to recede.

3.3.2.3 Comparison with experiments

In Fig. 3.14, we show the final momentum of the T-trajectory \mathbf{P} as a function of the ellipticity ξ computed using the SFA (dotted lines), the CCSFA from Eq. (2.3) (dashed lines), the reference Hamiltonian (1.14) (crosses) and the GC from Eqs. (3.12) (solid lines). The T-trajectory final momentum of the reference Hamiltonian (1.14) is not depicted if it is trapped in a Rydberg state or undergoes rescattering. In the lower-left panel, the hexagrams are the experimental data of \mathbf{P} reproduced from Ref. [93].

For $I = 8 \times 10^{13}$ W · cm⁻², He ($I_p = 0.9$ a.u.) and $\gamma \sim 0.6$ (top panels of Fig. 3.14), the T-trajectory of the reference Hamiltonian (1.14) corresponds to a direct ionization at the critical ellipticity $\xi_c \approx 0.25$, and reaches the detector without undergoing rescattering for $\xi > \xi_c$. The critical ellipticity is in agreement with the prediction $\xi_c \approx 0.26$ of Eq. (3.13). On the left panel, we observe a good agreement between the T-trajectory final momentum **P** of the reference Hamiltonian (1.14) (thin curves with crosses) and that of the GC model (thick solid curves) for the entire range of ellipticities $\xi > \xi_c$.



Figure 3.14: Final momentum of the T-trajectory $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$ as a function of the ellipticity ξ for Z = 1. Top panels: $I = 8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, He ($I_p = 0.9 \text{ a.u.}$) and $\gamma \sim 1.6$. Middle panels: $I = 1.2 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, Ar ($I_p = 0.58 \text{ a.u.}$) and $\gamma \sim 1$. Bottom panels: $I = 8 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, He ($I_p = 0.9 \text{ a.u.}$) and $\gamma \sim 0.6$. The hexagrams are the experimental data reproduced from Ref. [93]. In all panels: the dotted and dashed black lines are the T-trajectory of the SFA and the CCSFA, respectively. The thin (with crosses) and solid curves are the T-trajectory of the reference Hamiltonian (1.14) and the GC model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1), respectively. The components of the final momentum of the T-trajectory P_x and P_y are depicted in blue and red, respectively. The critical ellipticity ξ_c is at the intersection between the grey and white regions and corresponds to the largest ellipticity for which the T-trajectory of the critical ellipticity. We indicate the scaling of \mathbf{P} of the reference model (1.14) in the neighborhood of the bifurcation. Momenta are scaled by E_0/ω .

For $I = 1.2 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, Ar $(I_p = 0.58)$ and $\gamma \sim 1$ (middle panels of Fig. 3.14), the T-trajectory of the reference Hamiltonian (1.14) becomes a direct ionization at $\xi_c \approx 0.19$ while the GC prediction [see Eq. (3.13)] is $\xi_c \approx 0.24$. There is a small disagreement between the critical ellipticity of the reference model and the prediction of Eq. (3.13). However, there is a good agreement of the GC critical ellipticity with the experimental measurements of Ref. [104] of $\xi_c \approx 0.24$ as observed in Fig. 3.13. Furthermore, there is a good agreement between the T-trajectory final momentum **P** of the reference Hamiltonian (1.14) (thin curves with crosses) and that of the GC model (thick solid curves) for $\xi \gtrsim 0.3$. However, we observe a small disagreement between P_x of the reference Hamiltonian (1.14) (thin curves with crosses) and the GC prediction for all ellipticities. This discrepancy is related to the observations made in Fig. 3.3 and whose origin is discussed below.

For $I = 8 \times 10^{14}$ W · cm⁻², He ($I_p = 0.9$) and $\gamma \sim 0.6$ (lower panels of Fig. 3.14), the T-trajectory of the reference Hamiltonian (1.14) becomes a direct electron at $\xi_c \approx 0.05$. The critical ellipticity is in agreement with the prediction $\xi_c \approx 0.07$ of Eq. (3.13). In addition, these values agree well with the critical ellipticity $\xi_c \approx 0.08$ of the experiments [93] (hexagrams). There is again a good agreement between the Ttrajectory final momentum **P** of the reference Hamiltonian (1.14) (thin curves with crosses) and that of the GC model (thick solid curves) for $\xi \gtrsim 0.1$. However, we observe a disagreement between P_x of the reference Hamiltonian (1.14) (thin curves with crosses) and the GC prediction in the entire ellipticity range. We notice that for decreasing Keldysh parameters, the disagreement between P_x of the reference Hamiltonian (1.14) and the GC model increases, as observed in the lower panel of Fig. 3.3.

On the right panels of Fig. 3.14, we observe a good agreement between the exponents of P_x of the reference Hamiltonian (1.14) at the bifurcation and the prediction 0.5 of Eq. (3.14). However, the exponent of P_y at the bifurcation is much smaller than the exponent 1 predicted by Eq. (3.14).

In the left panels of Fig. 3.14, we observe excellent agreement between the T-trajectory final momentum of the reference Hamiltonian (1.14) (thin curves with crosses) and that of the CCSFA (dashed curves) after the bifurcation when the electron final energy is large.

3.3.2.4 T-trajectory analysis

Here, we show that the origin of the disagreements between the T-trajectory of the reference Hamiltonian (1.14) and the GC T-trajectory –the disagreement of P_x for small Keldysh parameters, or the disagreement with the critical exponents of P_y in the neighborhood of the bifurcation– are related to an underestimate of the Coulomb interaction by the GC model for a short time after ionization. In contrast, we show that the CCSFA agrees well with the solution of the reference Hamiltonian (1.14) for $\xi \gg \xi_c$ while it cannot capture correctly the phenomena related to the bifurcation.

In Fig. 3.15, the red dash-dotted, cyan solid and black dashed curves are the T-trajectory of Hamiltonian (1.14) after the change of coordinates, i.e., $\Phi_2(\mathbf{r}(t), \mathbf{p}(t))$, the GC model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1), and the CCSFA given by Eqs. (2.3), respectively. The thick dark blue curves are the T-trajectory of the reference Hamiltonian (1.14). Associated with each trajectory, we also show the GC energy, for each model, as a function of time per laser cycle t/T. The GC energy for each model consists in computing $H_2(\Phi_2(\mathbf{r}(t), \mathbf{p}(t)))$. Where the GC energy of the reference model is conserved, the GC model (whose GC energy is conserved) is valid.

For $\gamma \sim 1.6$ (see Fig. 3.15a–d), the electron ionizes far from the ionic core $(|\mathbf{r}_{PPT}^T| \sim E_0/\omega^2)$. For $\xi = 0.25$ and $\xi = 0.7$, respectively, we see the variations of the GC energy of the T-trajectory of Hamiltonian (1.14) (dash-dotted curve) are small, a signature of the validity of the GC model and an absence of rescattering. When the GC energy of Hamiltonian (1.14) becomes constant, it is only about 0.02 a.u. above the GC model prediction. As a consequence, we observe a good agreement between the trajectories of Hamiltonian (1.14) and the GC model trajectories in Fig. 3.15a and Fig. 3.15b. In particular, at $\xi = 0.25$, we observe the T-trajectory of Hamiltonian (1.14) is trapped in a Rydberg state, a feature which is reproduced by the GC model (cyan solid curve), but not well reproduced by the CCSFA (dashed black curve). Indeed, the Coulomb interaction remains significant for a long time after ionization during Rydberg state creation, and the conditions for the validity of the CCSFA are not met.

For $\gamma \sim 1$ (see Fig. 3.15e-h), the electron ionizes closer to the ionic core $(|\mathbf{r}_{PPT}^T| \sim 0.4E_0/\omega^2)$. For $\xi = 0.15$, the electron T-trajectory of Hamiltonian (1.14) (dash-dotted curve) and the GC model are trapped in Rydberg states. However, there is a large discrepancy between the trajectories. Indeed, in Fig. 3.15g, we observe that the dash-dotted red curve varies after ionization, indicating that the electron rescatters for a short time after ionization. For $\xi = 0.7$, the same happens in terms of energy (see Fig. 3.15h), and we see that the GC trajectory does not agree well with the T-trajectory of Hamiltonian (1.14) (see Fig. 3.15f). When the energy of Hamiltonian (1.14) becomes constant at $t \approx T/2$, it is larger than the GC energy prediction of



Figure 3.15: (a,b,e,f,i,j) T-trajectory in the polarization plane (x, y) for Z = 1. The thick dark blue curves are the T-trajectory of the reference Hamiltonian (1.14). The red dash-dotted, cyan solid and dashed black curves are the T-trajectory of Hamiltonian (1.14) with the change of coordinates $\Phi_2(\mathbf{r}(t), \mathbf{p}(t))$, the GC model $G_2 = (H_2, \Phi_2)$ (see Tab. 2.1) and the CCSFA (2.3), respectively. (c,d,g,h,k,l) Energy H_2 as a function of $(t - t_0)/T$, with $t_0 = T/2$, associated with each model. Right panels: $\xi = 0.7$. (a,c), (e,g) and (i,k) (the grey background panels are those for $\xi < \xi_c$) $\xi = 0.25$, $\xi = 0.15$ and $\xi = 0.05$, respectively. (a–d) are for $I = 8 \times 10^{13}$ W · cm⁻², He ($I_p = 0.9$ a.u.) and $\gamma \sim 1.6$ (same parameters as the top panels of Fig. 3.14). (e–h) are for $I = 1.2 \times 10^{14}$ W · cm⁻², Ar ($I_p = 0.58$ a.u.) and $\gamma \sim 1$ (same parameters as the middle panels of Fig. 3.14). (i–l) are for $I = 8 \times 10^{14}$ W · cm⁻², He ($I_p = 0.9$ a.u.) and $\gamma \sim 0.6$ (same parameters as the lower panels of Fig. 3.14). The dots indicate the origin, and the circles $|\mathbf{r}| = E_0/\omega^2$. The distances are scaled by E_0/ω^2 .

0.15 a.u. In Fig. 3.15e or Fig. 3.15f, we observe that after ionization, the initial electron distance from the core is $|\mathbf{r}_{\rm PPT}| \sim 0.4E_0/\omega^2$, while the GC initially at a distance $|\bar{\mathbf{r}}(t_0)| \sim 1.4E_0/\omega^2$ from core. Since in the GC model, the Coulomb interaction is evaluated at the GC position only, when the electron is closer to the core than predicted by the GC model, as is the case after ionization for $\gamma \leq 1.6$, the Coulomb interaction is underestimated in the GC model: the closer the electron to the ionic core, the more underestimated the Coulomb interaction.

For $\gamma \sim 0.6$ (see Figs. 3.15i–l), the electron ionizes even closer to the ionic core $(|\mathbf{r}_{PPT}| \sim 0.15E_0/\omega^2)$. For $\xi = 0.05$ and $\xi = 0.07$, there are also discrepancies between the cyan and red curves. We observe that the energy of the T-trajectory of Hamiltonian (1.14) (red dash-dotted curve in Figs. 3.15k and 3.15l) varies a lot for a short time after ionization (about 0.2T). Here again, the electron rescatters after ionization. In Fig. 3.15l, when the red dash-dotted curve becomes constant, the energy is above the GC prediction only by 0.02 a.u. However, this agreement is only coincidental since the T-trajectories of Hamiltonian (1.14) and of the GC disagree significantly due to the increase in energy of the rescattering. We observe that this increase in energy after ionization is well captured by the CCSFA.

In each panel, we observe an excellent agreement between the CCSFA and the T-trajectory of Hamiltonian (1.14) for a short time after ionization, i.e., $0 < t - t_0 \leq T$, when the hypotheses of the CCSFA are met. This method is effective for short time-scale dynamics or phenomena [80, 82, 114]. This agreement persists for longer times if the electron leaves quickly the ionic core region like in Ref. [62] or for large ellipticity (see Sec. 3.1.1), i.e., if its drift momentum is initially large.

3.4 Conclusions

In sum, we have investigated the role of the Coulomb potential in atoms subjected to strong laser fields, in particular, on its influence on short and long time scales. To do so, we have considered three reduced models of the reference Hamiltonian (1.14), namely the SFA [Eqs. (2.2)], the CCSFA [Eqs. (2.3)], and the GC model $[G_2 = (H_2, \Phi_2)$, see Tab. 2.1]. The analysis of these three reduced models allowed us to shed light on the manifestations of the Coulomb potential in various ionization processes. In the SFA, there are two types of trajectories: subcycle recollisions and direct ionizations. However, even when the intensity is very large, i.e., when the conditions of the SFA are met, the Coulomb interaction still makes its presence known for long time-scale phenomena. In particular, even at very high intensities, the Coulomb asymmetry persists as seen in Fig. 3.3 and discussed in Sec. 3.1.1. The Coulomb interaction brings with it a variety of additional types of trajectories, such as Coulomb-driven recollisions and Rydberg states. We have shown in Sec. 3.2 that these two processes are intimately related, and can be interpreted and predicted by the GC model.

During step (ii) of the recollision scenario, we have shown that the electron oscillates around the GC trajectory. In phase space, the GC trajectory lies on a curve of constant energy $\mathcal{E} = H_2(\bar{\mathbf{r}}, \bar{\mathbf{p}})$. If $\mathcal{E} > 0$, the GC motion is unbounded. In this case, it is likely the electron recollides if its GC angular momentum is near zero and its initial radial momentum is negative (like in Fig. 3.7b). Otherwise, the electron ionizes directly without recollision (like in Fig. 3.7c). If $\mathcal{E} < 0$, the GC motion is bounded. In this case, there exists at least one time at which the electron turns back towards the ionic core. Then, the electron may or may not recollide before the laser field is turned on. If the electron does recollide (like in Figs. 3.7d and 3.8d), the GC energy jumps to a new energy level. If the laser field is turned off before the electron recollides (as in Figs. 3.7e and 3.8e), the electron ends up in a Rydberg state.

The GC model does not capture the rescattering effects close to the ionic core but the CCSFA can since it is a rather short time-scale phenomenon [82, 84, 85, 114]. As observed in Fig. 3.15, the variations of energy of the reference model (1.14) can be well described by the CCSFA for short time scales. After rescattering, the electron potentially ionizes if its GC energy becomes positive (such as in Fig. 3.7d). Therefore, the CCSFA and the GC models are clearly complementary. The CCSFA is adapted for describing short timescale processes such as rescattering while the GC model is more suited for describing long time-scale processes such as Coulomb-driven recollisions and the creation of Rydberg states.

Finally, we have shown that changing ellipticity changes the yield of the different type of trajectories after quantum tunneling. In particular, for increasing ellipticities, the rescattering domain for which $\mathcal{E} < 0$, in which Coulomb-driven recollisions and Rydberg states are created, moves to regions with low ionization rate. Therefore, the yield of Rydberg state creation decreases drastically for increasing ellipticity, as observed in the left panel of Fig. 3.1. Also, we have shown that the behavior of the T-trajectory changes drastically for increasing ellipticities. Close to LP fields ($\xi = 0$), the GC energy of the T-trajectory is negative and the electron likely recollides with the core. At the critical ellipticity ξ_c , the GC energy of the T-trajectory changes sign, and its GC motion becomes unbounded. The electron likely undergoes direct ionization for $\xi > \xi_c$. Therefore, at $\xi = \xi_c$, the yield of Rydberg states is much smaller than the rate of Rydberg states at $\xi = 0$. Hence, the drastic decrease of the yield of Rydberg states and the bifurcation in the PMDs are due to the interplay between the population of classical states by the quantum tunneling, and the subsequent classical motion of the electron impacted by the Coulomb potential.

Chapter 4

Recollision mechanism at high ellipticities and its significance in HHG and NSDI: Role of the laser envelope

In absence of laser envelope, for f = 1, the laser field in Eq. (1.10) is $\mathbf{E}(t) = E_0[\mathbf{e}_x \cos(\omega t + \varphi_{\text{CEP}}) + \mathbf{e}_y \xi \sin(\omega t + \varphi_{\text{CEP}})]$, and the vector potential is $\mathbf{A}(t) = E_0[-\mathbf{e}_x \sin(\omega t + \varphi_{\text{CEP}}) + \mathbf{e}_y \xi \cos(\omega t + \varphi_{\text{CEP}})]$. For a given ionization time t_0 , in the SFA, the motion of the electron is given by

$$\mathbf{r}(t) = \mathbf{r}(t_0) + [\mathbf{p}(t_0) - \mathbf{A}(t_0)](t - t_0) + [\mathbf{E}(t) - \mathbf{E}(t_0)]/\omega^2.$$
(4.1)

At ionization, the electron exits the potential barrier with a very low initial momentum [see Sec. 1.2.2]. If $\mathbf{p}(t_0) = \mathbf{0}$, for $\xi = 0$, the electron leaves the ionic core with a drift momentum $-\mathbf{A}(t_0) = \hat{\mathbf{x}}(E_0/\omega) \sin(\omega t_0)$, and returns close to the core at time t_r , i.e., $\mathbf{r}(t_r) \approx \mathbf{r}(t_0)$, due to the laser oscillations $\mathbf{E}(t)/\omega^2$. For $\xi > 0$, strictly speaking the electron never comes back to the core, i.e., there are no times t_0 and t_r such that $\mathbf{r}(t_r) = \mathbf{r}(t_0)$ in Eq. (4.1) because of the non vanishing drift momentum $-\mathbf{A}(t_i)$ in the direction transverse



Figure 4.1: Single and double ionization probability curves as a function of the laser intensity in CP fields $(\xi = 1)$ from experimental measurements reproduced from (left panel) Ref. [55] and (right panel) Ref. [61]. Left panel: Target atoms He and Ne, the wavelength of the laser is $\lambda = 614$ nm. The curves are the normalized prediction of SDI given by the ADK ionization rare [see Eq. (1.24)]. Right panel: Target atom Mg, the wavelength of the laser is $\lambda = 800$ nm.

CHAPTER 4. RECOLLISION MECHANISM AT HIGH ELLIPTICITIES

to the electric field, which pushes the electron away from the core [39]. In this framework, the electron can only recollide when the laser field is linearly polarized (LP).

The recollision picture can, however, be extended to near LP-fields by taking into account that after ionization, the initial momentum along the transverse direction to the laser field p_{\perp} is distributed [8, 10] as $\propto \exp(-p_{\perp}^2 \sqrt{2I_p}/E_0)$ [see Sec. 1.2.2], i.e., the initial momentum is not exactly non-zero. The initial transverse momentum compensates the initial drift momentum of the electron after ionization and recollisions become possible [119, 97]. At the critical ellipticity ξ_c given in Eq. (3.13), the recollision probability drops by a factor 3 compared to its value for $\xi = 0$. For ellipticities $\xi > \xi_c$, the initial momentum $p_{\perp} \approx \xi E_0/\omega$ necessary to compensate the drift momentum is poorly weighted by the ionization rate and the probability that the electron returns to the core drops off. In Sec. 3.3.1.2, it is shown that even the ion-electron interaction is not sufficient to bring back the electron to the core after ionizing in CP fields with f = 1. Moreover, If the electron returns to the core in an elliptically polarized laser field, its difference of energy in the SFA is given by

$$\Delta \mathcal{E} = \kappa \mathbf{U}_p \frac{1 - \xi^2}{\xi^2 + 1}.$$
(4.2)

with $0 \le \kappa \le 3.17$. Hence, the recolliding electrons, if any, do not bring back enough energy from the laser field at high ellipticities to trigger HHG or NSDI. The three-step model predicts that the high frequency part of the HHG spectra or the double ionization induced by recollisions is suppressed with elliptically polarized lights. It is noted in Ref. [39] that

"For circularly polarized light, [Eq. (4.1)] indicates that the electron trajectory never returns to the vicinity of the ion. Consequently, electron-ion interactions will not be important".

The double ionization of atoms can occur sequentially or non-sequentially. The sequential double ionization (SDI) is the most intuitive mechanism for double ionization. It occurs when two electrons are teared off the core in sequence, independently of each other. In contrast, the nonsequential double ionization (NSDI), occurs when the double ionization of the atom is induced or impacted by electron-electron correlations. For instance, NSDI can be induced by recollisions [39, 73, 110]. Intuitively, the SDI probability is the product between the probability the first electron ionizes, and the probability the second electron ionizes [154]. The probability a single electron ionizes is given by the ADK ionization rate [see Eq. (1.24)]. The resulting SDI probability are smoothly increasing for increasing laser intensity. It corresponds to the curves in the left panel of Fig. 4.1. In the left panel of Fig. 4.1, the experimental measurements of the double ionization probability of He (\Box) and Ne (Δ) subjected to CP fields rather follow the SDI probabilities. Hence, in CP fields of wavelength $\lambda = 614$ nm, the double ionization of He and Ne is likely sequential, i.e., it does not involves recollisions. This is in agreement with the absence of recollisions in CP fields predicted by the three-step model [39]. However, in the right panel of Fig. 4.1, the experimental measurements of the double ionization probability of Mg (\circ) subjected to CP fields exhibits a knee structure for increasing laser intensity, in contrast with the SDI probability curves. In Ref. [61], it is mentioned that "The persistence of enhanced double ionization for circular polarization contradicts simple rescattering theories and opens the possibility for studying other ionization mechanisms". The hypothesis is

"[...] it is possible for the rapid departure of the first electron to create a change in the potential for the remaining electron that is sufficient to shake it into excited ionic states. [...] Shakeup exhibits no strong ellipticity dependence and hence does not preclude the production of enhanced double ionization for circular polarization".

However, the enhanced double ionization, i.e., the knee structure, is theoretically identified to be induced by recollisions in Ref. [111], in conflict with the prediction of the three-step model [39].

Questions

- Under which conditions experimental manifestations of recollisions in CP and near-CP fields can be observed ?
- How can the electron gain a significant amount of energy compared to U_p in CP fields in order to trigger HHG and NSDI ?

4.1. SCENARIO OF ENVELOPE-DRIVEN RECOLLISIONS



Figure 4.2: Typical recollision with a high return energy: From the ground state to the return for a 2-4-2 laser envelope. Left panel: Trajectory in the rotating frame (\tilde{x}, \tilde{y}) . The white and red circles are the position of the electron at the ionization and return time, respectively. The color shows the value of the zero-velocity surface during the plateau. Right panels: The trajectory projected along the position and energy of Hamiltonian (4.9) at time $t_0 \approx 0.3T$, $T_u = 2T$ (ramp-up duration) and $t_r \approx 2.7T$ (from left to right). The grey surface is the classical forbidden region for $\tilde{y} = 0$ [$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) < \mathcal{Z}(\tilde{\mathbf{r}}, t)$]. The light blue curve is the zero-velocity surface at time t = 0. The saddle point is the local maximum of the zero-velocity surface for negative \tilde{x} . Distances and energies are in a.u.

Plan All the analysis in the three-step model in the SFA is done in absence of pulse envelope. We find that the presence of an envelope drastically affects the conclusions of the three-step model. In this chapter, we identify a highly probable recollision channel with large return energy by accounting for the effects of the pulse envelope f(t). The recollisions taking this channel are referred to as envelope-driven recollisions. This recollision channel is particularly effective for nearly circular polarizations, since the conventional recollision channel disappears. Figure 4.2 shows the three steps of a typical recollision in CP fields following this channel, and seen in the rotating frame. The competition between the Coulomb force and the laser field makes this recollision channel highly probable by creating a channel of ionization early after the laser field is turned on. Just as the electron is outside the core region, the amplitude of the vector potential is small, and therefore the sideways drift of the electron can be compensated by its momentum. We show that this recollision channel and the return of the electron can be understood using the SFA, in opposition with the conventional threestep model for which the laser envelope is constant. Finally, we show this recollision channel can be used to produce HHG with atoms driven by high elliptically polarized laser fields, and is also responsible for the enhanced double ionization observed in experimental and theoretical measurements for CP fields [61, 111]. The existence conditions of this recollision channel agrees well with the enhanced double ionization for CP fields observed experimentally for specific target atoms and laser wavelengths. In addition, we notice that, under reasonable conditions, recollisions can take place in the Attoclock setup, where recollisions are always assumed to be nonexistent [53, 152].

Publication

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4.1 Scenario of envelope-driven recollisions

In this section, we describe the ionization mechanism of the envelope-driven recollisions. In particular, we show how the Coulomb potential induces highly probable ionizations early after the laser field is turned on. We describe quantum mechanically the field-free atom in the rotating frame (RF), in which the laser field is unidirectional. We consider the laser field given by Eq. (1.10). We show that, while the energy of the ground state of the electron is degenerate with respect to the quantum magnetic number in the laboratory frame, it is non-degenerate in the rotating frame. Then, we determine the time the electron over-the-barrier ionizes as a function of the initial energy of the electron in the ground state by treating classically the dynamics of the electron. The time when the electron ionizes allows us to draw conditions for which the electron does or does not return to the core. The return energy of the recolliding electrons is analyzed in the SFA. We find

that, for highly elliptically polarized laser fields, the envelope-driven recollisions mainly gain energy from the variations of the envelope.

4.1.1 Field-free atom in the rotating frame (RF)

We consider the atom in the ground state of energy $-I_p$. In Sec. 1.1.2, we describe the field-free atom in the LF. The Hamiltonian \hat{H} of the field free atom is given by Eq. (1.5) and the energy of the ground state energy of the atom $\Psi(\mathbf{r})$ is given by Eq. (1.6). The potential is rotationally invariant, and therefore the wave function of the electron can be written in terms of the radial wave function $R_n(|\mathbf{r}|)$ and spherical harmonics $Y_l^m(\mathbf{r}/|\mathbf{r}|)$ such that $\Psi(\mathbf{r}) = R_n(|\mathbf{r}|)Y_l^m(\mathbf{r}/|\mathbf{r}|)$. The principal, angular and magnetic quantum numbers are n, l and m, respectively. The angular and magnetic quantum numbers are such that $l = 0, \ldots, n-1$ and $m = -l, \ldots, l$, respectively. We consider the unitary transformation $\tilde{\Psi}(\mathbf{r}) = \hat{U}(t)\Psi(\mathbf{r})$ from the LF to the RF. The transformation is such that $\tilde{\Psi}(\tilde{\mathbf{r}}) = \Psi(\mathbf{r})$ where $\tilde{\mathbf{r}} = \mathbf{R}_z(\omega t + \varphi_{\text{CEP}})\mathbf{r}$ and $\mathbf{R}_z(\omega t + \varphi_{\text{CEP}})$ is the rotation matrix along \mathbf{e}_z (see Appendix A). The unitary operator associated to this transformation is

$$\hat{U}(t) = \exp\left[i\left(\omega t + \varphi_{\text{CEP}}\right)\hat{L}_z\right],\tag{4.3}$$

where $\hat{L}_z = \hat{\mathbf{r}} \times \hat{\mathbf{p}} \cdot \mathbf{e}_z$ is the angular momentum along \mathbf{e}_z . We substitute the transformation in the Schrödinger equation $\hat{H}\Psi = i\partial\Psi/\partial t$ and we identify the Hamiltonian in the RF such that $\hat{H}\tilde{\Psi} = i\partial\tilde{\Psi}/\partial t$. The relation between the Hamiltonian in the RF and LF is

$$\hat{\tilde{H}} = \hat{U}\hat{H}\hat{U}^{\dagger} - i\hat{U}\frac{\partial\hat{U}^{\dagger}}{\partial t}.$$
(4.4)

We apply the left- and right-hand side of Eq. (4.4) on $\tilde{\Psi}$. We use the relation $\hat{U}\partial\hat{U}^{\dagger}/\partial t = -(\partial\hat{U}/\partial t)\hat{U}^{\dagger}$. The energy of the electron in the RF is denoted $-\tilde{I}_p$, and is given by the eigenvalue problem

$$\tilde{H}\tilde{\Psi}(\tilde{\mathbf{r}}) = -\tilde{I}_p\tilde{\Psi}(\tilde{\mathbf{r}}). \tag{4.5}$$

We substitute the relation $\tilde{\Psi} = \hat{U}\Psi$, and the eigenvalue problems given by Eq. (1.6) and (4.5) in Eq. (4.4). Using $(\partial \hat{U}/\partial t)\hat{U}^{\dagger}\tilde{\Psi} = i\omega\hat{U}\hat{L}_{z}\Psi = i\omega m\tilde{\Psi}$ where $\hat{L}_{z}\Psi = m\Psi$. Finally, we obtain

$$\tilde{I}_p = I_p + \omega m. \tag{4.6}$$

Therefore, the energy of the electron in the RF depends on the magnetic quantum number $m \in \mathbb{Z}$. If m = 0, the wave function of the electron is invariant under rotations along \mathbf{e}_z , and as a consequence, the wave function is unchanged with respect to the unitary transformation (4.3). If $m \neq 0$, the wave function of the electron is no longer invariant under rotations along \mathbf{e}_z , and as a consequence, the electron in the RF might be larger than in the LF. The difference of energy of the ground state of the electron in the RF and in the LF is given by $m\omega$.

4.1.2 Over-the-barrier ionization: Role of the target species and the laser wavelength

We subject the field-free atom, described in the previous section, to an intense laser field elliptically polarized. The dynamics of the electron is treated classically in the RF. The initial energy of the electron in the RF is $\tilde{I}_p = I_p + m\omega$. We study the over-the-barrier ionization of the electron. We show that the ionization time of the electron strongly depends on the target species and the laser wavelength. In addition, we show that the ionization time of the electron is crucial for the conditions under which it comes back to the parent ion.

4.1.2.1 Classical Hamiltonian of the electron subjected to a strong laser field in the RF

We consider a single-active electron atom driven by a strong laser field elliptically polarized in the laboratory frame (LF) $\mathbf{E}(t) = E_0 f(t)/\sqrt{\xi^2 + 1} \left[\mathbf{e}_x \cos(\omega t + \varphi_{\text{CEP}}) + \mathbf{e}_y \xi \sin(\omega t + \varphi_{\text{CEP}}) \right]$. In the LF, the Hamiltonian is given by Eq. (1.14). We use the soft-Coulomb potential [78] to describe the ion-electron interaction $V(\mathbf{r}) = -Z(|\mathbf{r}|^2 + a^2)^{-1/2}$. We perform the canonical change of coordinates ($\tilde{\mathbf{r}}, \tilde{\mathbf{p}}$) = $\mathbf{R}_z(\omega t + \varphi_{\text{CEP}})(\mathbf{r}, \mathbf{p})$ to map the phase-space coordinates of the electron from the LF to the RF, where $\mathbf{R}_z(\omega t + \varphi_{\text{CEP}})$ is the rotation

matrix along \mathbf{e}_z [see Appendix A]. The absolute value of the position of the electron and its canonical conjugate momentum are conserved, i.e., $|\mathbf{r}| = |\tilde{\mathbf{r}}|$ and $|\mathbf{p}| = |\tilde{\mathbf{p}}|$. In the RF, the Hamiltonian reads

$$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = \frac{|\tilde{\mathbf{p}}|^2}{2} - \omega \tilde{\mathbf{r}} \times \tilde{\mathbf{p}} \cdot \tilde{\mathbf{e}}_z + V(\tilde{\mathbf{r}}) + \tilde{\mathbf{r}} \cdot \tilde{\mathbf{E}}(t).$$
(4.7)

The ion-electron potential in the RF $V(\tilde{\mathbf{r}})$ does not depends on time since V is rotationally invariant, which is in general the case for atoms. The term $-\omega \tilde{\mathbf{r}} \times \tilde{\mathbf{p}} \cdot \tilde{\mathbf{e}}_{z}$ is the Coriolis force. In analogy with the quantum mechanical treatment, this term comes from the term $i\hat{U}\partial\hat{U}^{\dagger}/\partial t$ in Eq. (4.4), and appears due to the inertial force acting on the electron when moving to the RF. In the RF, the elliptically polarized laser field reads

$$\tilde{\mathbf{E}}(t) = \frac{E_0 f(t)}{\sqrt{\xi^2 + 1}} \left\{ \tilde{\mathbf{e}}_x + (\xi^2 - 1) \left[\tilde{\mathbf{e}}_x \sin^2(\omega t + \varphi_{\text{CEP}}) + \tilde{\mathbf{e}}_y \sin(\omega t + \varphi_{\text{CEP}}) \cos(\omega t + \varphi_{\text{CEP}}) \right] \right\}.$$
(4.8)

We substitute the expression of the laser field (4.8) in Hamiltonian (4.7). For high elliptically polarized laser fields, $1 - \xi^2 \ll 1$, and therefore the term proportional to $(1 - \xi^2)$ acts as a perturbation of the CP case for which $\xi = 1$. The unperturbed Hamiltonian, corresponding to the CP case for which $\xi = 1$, reads

$$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = \frac{|\tilde{\mathbf{p}}|^2}{2} - \omega \tilde{\mathbf{r}} \times \tilde{\mathbf{p}} \cdot \tilde{\mathbf{e}}_z + V(\tilde{\mathbf{r}}) + \tilde{\mathbf{r}} \cdot \tilde{\mathbf{e}}_x \frac{E_0 f(t)}{\sqrt{2}}.$$
(4.9)

The direction of the electric field is along $\tilde{\mathbf{e}}_x$ for all time. Hamiltonian (4.9) does not depend explicitly on the CEP. As a consequence, for instance, the double ionization probability computed using rotationally invariant distribution of the initial conditions (or rotationally invariant initial wave function in quantum mechanical computations) does not depend on the CEP, as observed in Ref. [171]. If the laser envelope is f = 1, Hamiltonian (4.9) does not depend on time $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = \mathcal{K}$, and \mathcal{K} is the *Jacobi constant* [77]. Next, we consider the CP case and Hamiltonian (4.9).

4.1.2.2 Zero-velocity surface

Using $\tilde{\mathbf{r}} \times \tilde{\mathbf{p}} \cdot \tilde{\mathbf{e}}_z = \tilde{\mathbf{e}}_z \times \tilde{\mathbf{r}} \cdot \tilde{\mathbf{p}}$, Hamiltonian (4.9) can be written in the form

$$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = \frac{1}{2} |\tilde{\mathbf{p}} + \omega \tilde{\mathbf{r}} \times \tilde{\mathbf{e}}_z|^2 - \frac{\omega^2}{2} |\tilde{\mathbf{r}} \times \tilde{\mathbf{e}}_z|^2 + V(\tilde{\mathbf{r}}) + \tilde{\mathbf{r}} \cdot \tilde{\mathbf{e}}_x \frac{E_0 f(t)}{\sqrt{2}}.$$

As a consequence, the phase-space variables are such that $\hat{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) \geq \mathcal{Z}(\tilde{\mathbf{r}}, t)$ for all time t. The surface $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = \mathcal{Z}(\tilde{\mathbf{r}}, t)$ is obtained for $\dot{\tilde{\mathbf{r}}} = \tilde{\mathbf{p}} + \omega \tilde{\mathbf{r}} \times \tilde{\mathbf{e}}_z = \mathbf{0}$ and is called the *zero-velocity surface* [71]. The expression of the zero-velocity surface reads

$$\mathcal{Z}(\tilde{\mathbf{r}},t) = -\frac{\omega^2}{2} \left| \tilde{\mathbf{r}} \times \tilde{\mathbf{e}}_z \right|^2 + V(\tilde{\mathbf{r}}) + \tilde{\mathbf{r}} \cdot \tilde{\mathbf{e}}_x \frac{E_0 f(t)}{\sqrt{2}}.$$
(4.10)

Figure 4.3 shows the zero-velocity surface for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, f = 1 and $z = p_z = 0$ in the plane (\tilde{x}, \tilde{y}) . The zero-velocity surface $\mathcal{Z}(\tilde{\mathbf{r}}, t)$ is composed of a well near the origin due to the ion-electron interaction $V(\tilde{\mathbf{r}})$, a dome around $\tilde{\mathbf{e}}_x E_0 / \sqrt{2}\omega^2$ which corresponds to the global maximum of the zero-velocity surface, and a saddle point (indicated by a red dot). In the adiabatic approximation, in which the laser envelope is constant, these three points correspond to three fixed points of the dynamics for which $\dot{\tilde{\mathbf{r}}} = \tilde{\mathbf{p}} = \mathbf{0}$ (see Sec. 5.3.3). The coordinates of the saddle point are denoted $\tilde{\mathbf{r}}^* = \tilde{x}^* \tilde{\mathbf{e}}_x$ and $\tilde{\mathbf{p}}^* = \omega \tilde{x}^* \tilde{\mathbf{e}}_y$, where \tilde{x}^* is the negative solution of the equation

$$-\omega^2 \tilde{x}^* + \frac{\partial V}{\partial \tilde{x}}|_{\tilde{\mathbf{r}}^*} + E_0 f(t)/\sqrt{2} = 0.$$
(4.11)

The energy of the saddle point is denoted $\mathcal{Z}^{\star}(t) = \mathcal{Z}(\tilde{\mathbf{r}}^{\star}, t)$. The zero-velocity surface corresponds to the classical forbidden region of the electron, and acts as a barrier on the electron dynamics as observed in the inset of Fig. 4.3. If the laser envelope is f = 1 and the electron is inside the well near the origin with $\mathcal{K} < \mathcal{Z}^{\star}$, the electron is topologically bounded by the zero-velocity surface, i.e., it is trapped and cannot ionize.

4.1.2.3 Ionization time of the electron

We consider that the laser field is turned on at time t = 0. For $t \leq 0$, the location of the maximum of the zero-velocity surface is denoted $\tilde{\mathbf{r}}^*$. We assume that $V(\tilde{\mathbf{r}}) \approx -Z/|\tilde{\mathbf{r}}|$ (hard-Coulomb potential approximation).


Figure 4.3: Zero-velocity surface $\mathcal{Z}(\tilde{\mathbf{r}}, t)$ given by Eq. (4.10) for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, f = 1 and $V(\tilde{\mathbf{r}}) = -1/|\tilde{\mathbf{r}}|$. The circle shows the location of the saddle point $(\tilde{x}^*, 0, \mathcal{Z}^*(t))$. The plane corresponds to a surface $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) = -0.7$ a.u. in the plane (\tilde{x}, \tilde{y}) . The colored areas correspond to the allowed positions of the electron.

Substituting this assumption in Eq. (4.10) and looking for the location of the maximum of the zero-velocity surface, we obtain $|\tilde{\mathbf{r}}^{\star}| = (Z/\omega^2)^{1/3}$. The maximum of the zero-velocity surface before the laser field is turned on is

$$\mathcal{Z}^{\star}(0) = \mathcal{Z}(\mathbf{r}^{\star}, 0) = -\frac{3}{2} (Z\omega)^{2/3}.$$
(4.12)

Therefore, the zero-velocity surface, which bounds topologically the electron motion, is under the zero-energy surface before the laser field is turned on. In addition, before the laser field is turned on, i.e., for f(0) = 0, the energy of the electron in the RF is $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, 0) = -\tilde{I}_p$ with \tilde{I}_p given by Eq. (4.6). We distinguish two cases:

- If $I_p < -\mathcal{Z}^*(0)$, the electron is not topologically bounded by the zero-velocity surface. However, it is dynamically bounded by invariant tori.
- If $\tilde{I}_p > -\mathcal{Z}^{\star}(0)$, the electron is topologically bounded by the zero-velocity surface and dynamically bounded by invariant tori.

We denote t_0 the ionization time of the electron. For $0 \leq t \leq t_0$, the electron is near the origin and $|\mathbf{r}| \sim 1$. As a consequence, the time-dependent term in Hamiltonian (4.9) is small and the energy of the electron varies slowly compared to the duration of the pulse. Therefore, we assume that $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) \approx -\tilde{I}_p$ for $0 \leq t \leq t_0$. For increasing time, the envelope increases and the energy of the saddle point decreases as $\mathcal{Z}^*(t) \approx \mathcal{Z}^*(0) - f(t)E_0\omega^{-2/3}$ for $f(t) \ll 1$. When the saddle point comes below the electron, the electron is no longer topologically bounded (in the adiabatic picture), and it can ionize. Likely, the electron ionizes through the saddle point. We assume that the breakdown of the invariant tori, which bound the electron dynamically, occurs right after the energy of the electron is above the energy of the saddle point of the zero-velocity surface. Under this approximation, the ionization time is:

- If $\tilde{I}_p < -\mathcal{Z}^{\star}(0)$, the electron is below the top of the zero-velocity surface before the laser is turned on. The electron ionizes right after the laser is turned on, and $t_0 = 0$.
- If $\tilde{I}_p > -\mathcal{Z}^*(0)$, the electron is above the top of the zero-velocity surface before the laser is turned on. The ionization occurs when the energy of the saddle point is the same as the energy of the electron, and t_0 is solution of $\mathcal{Z}^*(t_0) = -\tilde{I}_p$.

We notice that the electron can ionize earlier by tunneling through the classical forbidden region. However, we expect the rate of electrons ionizing through the barrier to be negligible compared to the rate of electrons ionizing over the barrier. In the second case, when $\tilde{I}_p > -\mathcal{Z}^*(0)$, the ionization time is defined by $\mathcal{Z}^*(t_0) = -\tilde{I}_p$. This condition occurs when the laser field reaches a specific effective amplitude $E^* = E_0 f(t_0)/\sqrt{2}$. At time t_0 , such that $\mathcal{Z}^*(t_0) = -\tilde{I}_p$, the saddle point of the zero-velocity surface is located at

$$x^{\star} = \left(\frac{2Z}{\omega^2}\right)^{1/3} \rho(\mu), \tag{4.13}$$

with $\rho(\mu) = (-1 + \sqrt{\mu + 1})^{1/3} - (1 + \sqrt{\mu + 1})^{1/3}$ and $\mu = 2\tilde{I}_p^3(Z\omega)^{-2}/27$, and where we have considered $V(\tilde{\mathbf{r}}) \approx -Z/|\tilde{\mathbf{r}}|$. Notice that the location of the saddle point when its energy is the same as the energy of the electron does not depend on the intensity of the laser. The effective amplitude of the laser when the electron ionizes, $E^* = E_0 f(t_0)/\sqrt{2}$, is solution of the equation $-\tilde{I}_p = -\omega^2 x^{*2}/2 + Z/x^* + x^* E^*$ (where we have used $x^* < 0$).

Result 6: Ionization time of the electron in a CP field

The time when the electron of energy $I_p = I_p + m\omega$ (with *m* the magnetic quantum number of the electron) over-the-barrier ionizes from atoms driven by CP fields is given by

$$t_0 = f^{-1} \left(\sqrt{2} E^* / E_0 \right), \tag{4.14}$$

where f^{-1} is the inverse function of the laser envelope. The effective amplitude of the laser when the electron ionizes is $E^{\star} = E_0 f(t_0)/\sqrt{2}$. If $E^{\star} > E_0/\sqrt{2}$, the electron cannot ionize over the barrier. The expression of the effective amplitude of the laser is given by

$$E^{\star} = \max\left\{0, \left(\frac{Z\omega^4}{4}\right)^{1/3} \frac{1+2\rho(\mu)^3}{\rho(\mu)^2}\right\},\tag{4.15}$$

with $\rho(\mu) = (-1 + \sqrt{\mu + 1})^{1/3} - (1 + \sqrt{\mu + 1})^{1/3}$ and $\mu = 2\tilde{I}_p^3(Z\omega)^{-2}/27$. The effective amplitude of the laser when the electron ionizes is the same for all intensities. In contrast, the ionization time of the electron varies for varying intensity. If the energy of the electron is above the top of the zero-velocity surface before the laser field is turned on, $E^* = 0$.

4.1.2.4 Breakdown of recollisions in CP fields

Just as the electron is outside the core region, around the saddle point of the zero-velocity surface, the effective amplitude of the laser field $E^* = E_0 f(t_0)/\sqrt{2}$ is given by Eq. (4.15) as a function of the parameters of the atom and the laser. At time t_0 , the vector potential of the laser is such that $|\mathbf{A}(t_0)| \approx E^*/\omega$. The larger $f(t_0)$, the larger the vector potential at ionization. Therefore, the larger $f(t_0)$, the larger the necessary initial momentum $|\mathbf{p}(t_0)|$ to compensate the sideways drift of the electron [see Eq. (4.1)]. However, at time t_0 , the electron is close to the zero-velocity surface, and as a consequence, its initial momentum is small compared to E_0/ω . To be able to compensate the sideways drift of the electron, the ionization must take place for small effective amplitude, i.e., early after the laser field is turned on. The ionization time t_0 plays a crucial role in the condition for which the electron can come back to the core.

We assume that the electron ionizes through the saddle point. Under this hypothesis, at time t_0 , the position and the momentum of the electron are $\tilde{\mathbf{r}}(t_0) = \tilde{x}^* \tilde{\mathbf{e}}_x$ and $\tilde{\mathbf{p}}(t_i) = \omega \tilde{x}^* \tilde{\mathbf{e}}_y$, respectively, where \tilde{x}^* is given in Eq. (4.13). In Sec. 3.2, we show that the energy of the GC of the electron \mathcal{E} can be used to distinguish electrons which do and do not come back to the core by taking into account the ion-electron interaction. If $\mathcal{E} < 0$, the GC motion is bounded, and the electron comes back to the core. If $\mathcal{E} > 0$, the GC motion is unbounded, and the electron ionizes directly. In order to determine the configuration of the GC of the electron at time t_0 , we use the second order GC model $G_2 = (H_2, \Phi_2)$. In the RF, the coordinates of the GC at time t_0 are related to the electron coordinates with

$$\tilde{\tilde{\mathbf{r}}}(t_0) = \tilde{\mathbf{r}}(t_0) - \tilde{\mathbf{e}}_x E^* / \omega^2 = \left[\tilde{x}^* - E^* / \omega^2 \right] \tilde{\mathbf{e}}_x, \tilde{\tilde{\mathbf{p}}}(t_0) = \tilde{\mathbf{p}}(t_0) - \tilde{\mathbf{e}}_y E^* / \omega = \omega \left[\tilde{x}^* - E^* / \omega^2 \right] \tilde{\mathbf{e}}_y.$$

The energy of the GC is conserved in time until the electron returns to the core. The energy of the GC is given by $\mathcal{E} = |\tilde{\mathbf{p}}(t_0)|^2/2 - Z/|\tilde{\mathbf{r}}(t_0)|$, where we have used the conservation of the norm of the GC coordinates when moving from the LF to the RF. By using the definition of the saddle point along $\tilde{\mathbf{e}}_x$ given by Eq. (4.11), we obtain $\omega^2 \tilde{x}^* - E^* = -Z/\tilde{x}^{*2}$. We substitute the later equation in the energy of the GC, and we obtain

$$\mathcal{E} = \left(\frac{\omega^2}{2}\right)^{1/3} \frac{1 - 8\rho(\mu)^6}{4\rho(\mu)^4},\tag{4.16}$$

with $\rho(\mu) = (-1 + \sqrt{\mu + 1})^{1/3} - (1 + \sqrt{\mu + 1})^{1/3}$ and $\mu = 2\tilde{I}_p^3(Z\omega)^{-2}/27$. If $\mathcal{E} > 0$, the trajectory of the GC is unbounded and the electron goes away from the core without recolliding. If $\mathcal{E} < 0$, the trajectory of the guiding center is bounded and the electron can come back to the core, driven by its GC. Using Eq. (4.16), the condition for the electron returns $\mathcal{E} < 0$ implies that $8\rho(\mu)^6 > 1$. Since $\rho(\mu)$ is negative, $\rho(\mu) < -1/\sqrt{2}$.



Figure 4.4: Phase diagram of the recollisions in CP as a function of the ionization potential of the atom I_p and the wavelength of the laser λ . The solid curves are the critical ionization potential given in Eq. (4.17). The lower and upper solid curves correspond to atoms with outer shell electrons of angular quantum numbers l = 0and l = 1, respectively. The dashed grey curve corresponds to the critical ionization potential of Ref. [58]. The white and yellow markers show parameters for experimental measurements of double ionization. The white symbols are where a knee structure is absent in the double ionization probability as a function of the laser intensity. The yellow symbol is where a knee structure is seen in the double ionization probability as a function of the laser intensity. The experimental measurements for He (\Box) and Ne (Δ) at $\lambda = 614$ nm are reported in Ref. [55], and shown in the left panel of Fig. 4.1. The experimental measurements for Mg (\circ) at $\lambda = 780$ nm are reported in Ref. [61], and shown in the right panel of Fig. 4.1.

Thus, the parameter μ satisfies $\mu < \mu^*$ with $\rho(\mu^*) = -1/\sqrt{2}$. The solution of $\rho(\mu^*) = -1/\sqrt{2}$ is given by $\mu^* = (-97 + 140\sqrt{2})/216 \approx 0.47$. Using the definition $\mu = 2\tilde{I}_p^3(Z\omega)^{-2}/27$, we obtain $\tilde{I}_p^3 < \mu^*(27/2)(Z\omega)^2$.

Result 7: Critical ionization potential for recollisions in CP fields

Recollisions in CP fields manifest for specific conditions [65, 61, 111, 81]. The energy of the initial state of the electron is denoted $-I_p$. We obtain that:

- If $I_p < I_c$, the electron can recollide in CP fields. The electron ionizes early enough after the laser field is turned on, and the sideways drift of the electron can be compensated by its momentum when it is outside the core region.
- If $I_p > I_c$, the electron cannot recollide in CP fields. The effective amplitude of the laser when the electron ionizes E^* [see Eq. (4.15)] is too large, and the sideways drift of the electron cannot be compensated by its momentum when it is outside the core region.

The critical ionization potential separating the recolliding from the non-recolliding electronic states in CP fields is given by

$$I_c = \eta (Z\omega)^{2/3} + \omega l, \qquad (4.17)$$

where all quantities are in atomic units, $\eta \approx 1.85$ and $l \in \mathbb{N}$ is the angular quantum number of the initial state of the electron. For instance, for a laser wavelength $\lambda = 780$ nm, the critical ionization potential is $I_c \approx 7.4$ eV. As a consequence, all electrons initiated in an excited state of Hydrogen can recollide when subjected to CP fields of wavelength $\lambda = 780$ nm.

Figure 4.4 shows the phase diagram of the electron in the plane (λ, I_p) . The solid lines are the critical ionization potential given by Eq. (4.17) for l = 0 (lower curve) and l = 1 (upper curve). For instance, we observe that for $\lambda > 780$ nm, recollisions are likely to be observed only for Mg (\circ) and Li, in agreement with the enhanced double ionization probability observed in the experimental measurements reported in Ref. [61], depicted in the right panel of Fig. 4.1. In addition, the energy of all excited states of H (the energy of the first excited state of H is -3.4 eV) are above the critical ionization potential I_c given by Eq. (4.17). In the Attoclock setup with H, recollisions can manifest if there are excited H atoms in the sample [152]. If the laser wavelength is decreased, recollisions can be observed for other target species. This is in agreement with the numerical measurements of Ref. [36]. For $\lambda = 614$ nm, we observe that recollisions are unlikely in He (\Box) and Ne (Δ), in agreement with the absence of enhanced double ionization in the experimental measurements reported in Ref. [55], and depicted in the right panel of Fig. 4.1. For $\lambda = 800$ nm, we observe that recollisions are unlikely in Ke (\diamond), in agreement with the absence of enhanced double ionization in the experimental measurements reported in Ref. [55].

4.1.3 Return energy of the recolliding electron

We consider the excursion of recolliding electrons ionizing at time t_0 and returning at time t_r in intense laser fields. We show that for a constant laser envelope f = 1, the electron does not bring back energy to the core during its excursion in the continuum, even when the ion-electron interaction is taken into account. Then, we consider a slowly varying laser envelope to study the conditions under which the electron gains energy in highly elliptically polarized laser fields using the SFA. We find that for highly elliptically polarized laser pulses, the recolliding electrons mainly gain energy from the variations of the laser envelope, in contrast to low elliptically polarized laser pulses where the recolliding electrons mainly gain energy from the oscillations of the laser field induced by the carrier-envelope frequency.

4.1.3.1 Constant laser envelope

We consider the case for which the envelope of the laser is constant, f = 1. The electron ionizes close to the core at time t_0 and returns close to the core at time t_r . That means

$$\mathbf{r}(t_0) \approx \mathbf{r}(t_r) \approx \mathbf{0}.\tag{4.18}$$

Hamiltonian (4.9) is conserved in time, $\mathcal{K} = \tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}})$, and as a consequence, $\tilde{H}(\tilde{\mathbf{r}}(t_0), \tilde{\mathbf{p}}(t_0)) = \tilde{H}(\tilde{\mathbf{r}}(t_r), \tilde{\mathbf{p}}(t_r))$, i.e., the energy of the electron in the RF is the same when it ionizes outside the ionic core region and when it returns to the core. We substitute Eq. (4.18) in $\tilde{H}(\tilde{\mathbf{r}}(t_0), \tilde{\mathbf{p}}(t_0)) = \tilde{H}(\tilde{\mathbf{r}}(t_r), \tilde{\mathbf{p}}(t_r))$. After simplification, we obtain

$$\Delta \mathcal{E} = \frac{|\mathbf{p}(t_r)|^2}{2} - \frac{|\mathbf{p}(t_0)|^2}{2} \approx 0, \tag{4.19}$$

where we have used $|\mathbf{p}| = |\tilde{\mathbf{p}}|$. We denote $\Delta \mathcal{E}$ the difference of energy of the recolliding electron when it returns to the core and when it ionizes. Therefore, even if the ion-electron interaction is taken into account, the recolliding electron does not bring back energy to the core from its excursion in CP fields with constant laser envelope due to the Jacobi constant. We notice that the SFA prediction given by Eq. (4.2) is consistent with Eq. (4.19).

4.1.3.2 Energy boost of the electron

If the laser envelope is taken into account, Hamiltonian (4.9) depends explicitly on time, and it is no longer conserved. As a consequence, the energy of the recolliding electron in the RF can also vary during its excursion, as observed in Fig. 4.2. In order to assess the energy difference $\Delta \mathcal{E}$ of the envelope-driven recollisions, we consider a slowly varying laser envelope such that $|\partial^j f/\partial t^j| \ll \omega^j$ for $j \in \mathbb{N}^*$. In the SFA, at first order, the difference of return energy of the electron is given by

$$\Delta \mathcal{E} \approx \frac{2U_p}{\xi^2 + 1} \left[f(t_r)^2 - f(t_0)^2 \right].$$
(4.20)

If the electron ionizes and returns when the laser field is f = 1, we observe that the electron does not gain energy, in agreement with Eq. (4.2) and (4.19). In contrast, if the electron ionizes at time t_0 and returns at time t_r such that $f(t_0)^2 < f(t_r)^2$, the electron gains energy. For instance, this is the case if the electron ionizes during the ramp-up of the laser field and returns at the peak amplitude of the laser field. The maximum energy the electron can bring back to the core, in the slowly varying envelope approximation, is $2U_p/(\xi^2+1)$. This is the energy boost of an electron ionizing at $t_0 = 0$ (when $f(t_0) = 0$) and returning to the core at the peak amplitude of the laser, when $f(t_r) = 1$. In sum, in order to bring back a large amount of energy to the core from the laser field, electrons must ionize early after the laser field is turned on. The conditions under which the energy of the electron can be compensated by its momentum when it ionizes. As a consequence, electrons which ionize early after the laser field is turned to intense and CP laser fields with high return energy.



Figure 4.5: Distribution of the return energy of the electron for $I = 1.4 \times 10^{15} \text{ W} \cdot \text{cm}^{-2}$, $\xi = 1$ and a 2-4-2 trapezoidal laser envelope. We use a microcanonical generation of the initial conditions such that $H(\mathbf{r}, \mathbf{p}) = -0.1$ a.u. before the laser field is turned on. The return energy is computed with $\mathcal{E}_r = |\mathbf{p}(t_r)|^2/2$ at time t_r such that $|\mathbf{r}(t_r)| = 5$ a.u. The solid and dashed black lines overlap and correspond to the return energy and the difference energy of the recolliding electron in the SFA for $t_0 = 0$.

Figure 4.5 shows the distribution of return energy of the electron initiated with an energy $H(\mathbf{r}, \mathbf{p}) = -0.1$ a.u. (see Appendix D.1.2 for detail) as a function of the return time t_r , for a trapezoidal envelope 2–4–2, i.e., $T_u = 2T$, $T_p = 4T$ and $T_d = 2T$. The recolliding trajectories are detected using distance criteria with

thresholds R_{\min} and R_{\max} . The electron ionizes at time at time t_0 such that $|\mathbf{r}(t_0)| < R_{\min}$, travels outside the core at a distance $|\mathbf{r}(t)| > R_{\max}$, and then returns at time t_r such that $|\mathbf{r}(t_r)| < R_{\min}$. The parameter R_{\min} corresponds roughly to the core region. The parameter R_{\max} ensures the electron goes far enough from the core to be considered as a recollision. We choose $R_{\min} = 5$ a.u. and $R_{\max} = 6$ a.u.. The ionization time t_0 and the return time t_r could be defined such as $|\mathbf{r}(t_0)| = R_{\min}$ and $|\mathbf{r}(t_r)| = R_{\min}$. Here, we choose to define them as the time the electron has changed direction, i.e., the time its radial momentum changes sign. The ionization time t_0 (resp. return time t_r) is the last time (resp. first time) when $|\mathbf{r}(t_0)| < R_{\min}$ (resp. $|\mathbf{r}(t_r)| < R_{\min}$) such that

$$\mathbf{r}(t_0) \cdot \mathbf{p}(t_0) = 0, \quad \mathbf{r}(t_r) \cdot \mathbf{p}(t_r) = 0.$$
(4.21)

In this way, we ensure to identify the ionization and return times as the time when the electron is the closest from the ionic core before and after ionization, respectively. If the electron does not return to the core, t_i is the ionization time of the electron.

In Fig. 4.5, the return energy of Hamiltonian (1.14) with initial conditions $H(\mathbf{r}, \mathbf{p}) = -0.1$ a.u. is compared with the return energy \mathcal{E}_r (solid line) and the difference energy $\Delta \mathcal{E}$ (dashed line) of the electron in the SFA for $t_0 = 0$ and a trapezoidal envelope 2–4–2 [see Eq. (4.22) and (4.23)]. For each return time, we observe an excellent agreement between the maximum of the distribution of Hamiltonian (1.14) and the SFA prediction. As a consequence, the SFA can be used to determine the return energy of these electrons. In addition, we observe that the solid and dashed black lines overlap. Therefore, the initial energy $\mathcal{E}_0 = \mathcal{E}_r - \Delta \mathcal{E}$ (energy at time t_0) of the electrons which return to the core is very small compared to U_p . Because the electrons ionize early after the laser field is turned on, their energy is sufficient to compensate their sideways drifts.

4.1.3.3 Extension to EP laser fields

For $\xi \neq 1$, the recollision picture is not so clear since the saddle point of the zero-velocity surface moves in time even for fixed laser envelope. However, the scenario works in the same way: The electron initiated with a sufficiently small ionization potential ionizes early after the laser field is turned on, with a small initial momentum, which compensates the initial vector potential in Eq. (4.1), such that $\mathbf{p}(t_i) - \mathbf{A}(t_i) \approx \mathbf{0}$. Then, the electron travels in the continuum. In the SFA, using the conditions $\mathbf{r}(t_0) = \mathbf{r}(t_r)$ for an electric field given by Eq. (1.10) and for an arbitrary laser envelope, the return energy of the electron is

$$\mathcal{E}_{r}(t_{0},t_{r}) = \frac{E_{0}^{2}}{2} \int_{t_{0}}^{t_{r}} \mathrm{d}t_{1} \int_{t_{0}}^{t_{r}} \mathrm{d}t_{2} \left\{ f(t_{1})f(t_{2})\cos[\omega(t_{1}-t_{2})] - 2 \int_{t_{0}}^{t_{2}} \mathrm{d}t_{3} f(t_{1})f(t_{3})\frac{\cos[\omega(t_{1}-t_{3})]}{t_{r}-t_{0}} + \int_{t_{0}}^{t_{1}} \mathrm{d}t_{3} \int_{t_{0}}^{t_{2}} \mathrm{d}t_{4} f(t_{3})f(t_{4})\frac{\cos[\omega(t_{3}-t_{4})]}{(t_{0}-t_{r})^{2}} \right\} + \frac{E_{0}^{2}(\xi^{2}-1)}{2(\xi^{2}+1)} \left[\int_{t_{0}}^{t_{r}} \mathrm{d}t_{1} f(t_{1})\sin(\omega t_{1}+\varphi) - \int_{t_{0}}^{t_{r}} \mathrm{d}t_{1} \int_{t_{0}}^{t_{1}} \mathrm{d}t_{2} f(t_{2})\frac{\sin(\omega t_{2}+\varphi)}{t_{r}-t_{0}} \right]^{2}.$$
(4.22)

One can verify that if f = 1, the maximum return energy of the electron is the well-known $3.17 U_p/(\xi^2 + 1)$. The difference of energy of the electron during its excursion outside the core region is

$$\Delta \mathcal{E}(t_0, t_r) = \frac{E_0^2}{2} \int_{t_0}^{t_r} dt_1 \int_{t_0}^{t_r} dt_2 \int_{t_0}^{t_2} dt_3 f(t_1) \left\{ f(t_2) \frac{\cos\left[\omega(t_1 - t_2)\right]}{t_2 - t_0} - 2f(t_3) \frac{\cos\left[\omega(t_1 - t_3)\right]}{t_r - t_0} \right\} + \frac{E_0^2(\xi^2 - 1)}{2(\xi^2 + 1)} \int_{t_0}^{t_r} dt_1 f(t_1) \sin(\omega t_1 + \varphi) \times \int_{t_0}^{t_r} dt_2 \int_{t_0}^{t_2} dt_3 \left[f(t_2) \frac{\sin(\omega t_2 + \varphi)}{t_2 - t_0} - 2f(t_3) \frac{\sin(\omega t_3 + \varphi)}{t_r - t_0} \right].$$
(4.23)

For slowly varying laser envelope, the laser envelope is such that $|\partial^j f(t)/\partial t^j| \ll \omega^j$, where $j \in \mathbb{N}^*$. We use the small parameter ϵ for bookkeeping purposes, such that $f(t) = f(\epsilon t)$ and such as it is used in Sec. 2.2.2.5. In Eq. (4.23), we perform integration by parts to obtain a Taylor series in ϵ . At the lowest order, the energy gained by the electron is

$$\Delta \mathcal{E} \approx 2 U_p \left[f(t_r)^2 - f(t_i)^2 \right] + 2 U_p (1 - \xi^2) \left[g(t_r, t_i) - g(t_i, t_r) \right], \tag{4.24}$$



Figure 4.6: HHG spectrum for $\xi = 0, 0.25, 0.5, 0.75$ and 1 and $I = 8(\xi^2 + 1) \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. The vertical lines correspond to the radiated frequencies $2.3 \text{U}_p/(\xi^2 + 1) + I_p$ (left) and $3.17 \text{U}_p/(\xi^2 + 1) + I_p$ (right). Upper left: Initial state initiated as $\Psi(\mathbf{r}, 0) = [\psi_0(\mathbf{r}) + \psi_1(\mathbf{r})]/\sqrt{2}$. Upper right: Initial state initiated as $\Psi(\mathbf{r}, 0) = [3\psi_0(\mathbf{r}) + c_1\psi_1(\mathbf{r})]/\sqrt{10}$. Lower panel: Comparison of the spectrum in LP and CP for an initial state with a small and large portion of excited state.

with

$$g(t_1, t_2) = f(t_1)^2 \cos(\omega t_1 + \varphi) \left[\cos(\omega t_1 + \varphi) - 2 \frac{\sin(\omega t_1 + \varphi)}{\omega(t_1 - t_2)} \right]$$
$$+ 2f(t_1)f(t_2) \frac{\sin(\omega t_2 + \varphi_{\text{CEP}})\cos(\omega t_1 + \varphi_{\text{CEP}})}{\omega(t_1 - t_2)}.$$

For slowly varying envelope, in Eq. (4.24), we observe two main terms contributing to the energy boost of the electron during its excursion outside the core. The first term in the right-hand side of Eq. (4.24) is $2U_p \left[f(t_r)^2 - f(t_i)^2\right]$, and depends on the laser envelope only. In contrast, the last term in the right-hand side of Eq. (4.24) is proportional to $2U_p(1-\xi^2) \left[g(t_r,t_i) - g(t_i,t_r)\right]$, and depends on the CEP φ_{CEP} and the carrier-envelope frequency ω . At high ellipticities, $1-\xi^2 \ll 1$, and the last term in the right-hand side of Eq. (4.24) is very small compared to the first term. In this case, the electron mostly gains energy through the variations of the laser envelope. In contrast, at low ellipticities, $1-\xi^2 \approx 1$, and the magnitude of the last term in the right-hand side of Eq. (4.24) is very small compared to the magnitude of the first term. In this case, the electron mostly gains energy through the oscillations of the laser.

4.2 Applications to HHG

The high harmonic generation (HHG) of atomic or molecular gases subjected to intense infra-red laser fields heralded a new era in science and technology due to its ability to unravel the electron dynamics on its own spatial and temporal scales [59, 162, 4] and its role in realizing table-top ultrashort light sources [132, 153, 35]. Controlling HHG from the parameters of the driving laser represents a formidable challenge both on the experimental [15] and theoretical [44] sides with considerable amount of research efforts for the past decades. In this section, we use envelope-induced recollisions to produce HHG with an atom subjected to a highly elliptically polarized laser pulse.

4.2.1 Production of HHG with high EP laser fields

The radiation of the electron is induced by its dipole acceleration

$$\langle \mathbf{a}(t) \rangle = -\int \mathrm{d}\mathbf{r} \, |\Psi(\mathbf{r},t)|^2 \frac{\partial V(\mathbf{r})}{\partial \mathbf{r}},$$
(4.25)

where $\Psi(\mathbf{r}, t)$ is the total wave function of the electron. According to the semi-classical scenario of recollisions [100, 9], harmonic emission occurs for a linear combination of two eigenstates: One portion of the wave function is in the ground state $\psi_0(\mathbf{r})$ and another portion is in the recolliding state $\psi_1(\mathbf{r}, t)$. The total wave function is of the form [100] $\Psi(\mathbf{r}, t) = c_0\psi_0(\mathbf{r}) + c_1\psi_1(\mathbf{r}, t)$. Roughly speaking, following Ref. [88], we assume that the energy of the ground state $-I_p$ and the return energy of the recolliding electron \mathcal{E}_r are eigenvalues of the Hamiltonian operator in the eigenstates ψ_0 and ψ_1 , respectively. In this case, the wave function reads $\Psi(\mathbf{r}, t) \approx c_0 \exp(iI_p t)\psi_0(\mathbf{r}) + c_1 \exp(-i\mathcal{E}_r t)\psi_1(\mathbf{r})$. We substitute the latter equation in Eq. (4.25), and we obtain

$$\langle \mathbf{a}(t) \rangle = -c_0^* c_1 \exp\left[-i(\mathcal{E}_r + I_p)\right] \int d\mathbf{r} \,\psi_0^*(\mathbf{r}) \psi_1(\mathbf{r}) \frac{\partial V(\mathbf{r})}{\partial \mathbf{r}} + \text{c.c.}.$$
(4.26)

Therefore, the dipole acceleration oscillates at a frequency $I_p + \mathcal{E}_r$. When the recolliding wave packet of energy \mathcal{E}_r interferes with the portion of the wave function in the ground state, the electron recombines in the ground state of energy $-I_p$. Due to energy conservation, a photon of frequency

$$\Omega = I_p + \mathcal{E}_r,$$

is emitted. The production of high harmonics is observed if $\mathcal{E}_r \sim U_p$. The simplified expression for the dipole acceleration derived in Ref. [88] and given by Eq. (4.26) highlights the important ingredients for the production of HHG. The production of HHG is possible only if:

- A portion of the ionized wave packet returns to the core, i.e., $c_1 \neq 0$.
- The ground state of the electron is populated when the recolliding wave packet returns to the core, i.e., $c_0 \neq 0$.
- The recolliding electrons gain energy from the laser field in order that $\mathcal{E}_r \sim U_p$.

We recall that envelope-induced recollisions are observed for electrons with ionization potential such that $I_p < I_c$. When this condition is fulfilled, a portion of the ionized wave packet can return to the core. For high ellipticities, the electron gains energy from the laser field only if it ionizes early after the laser field is turned on. In this case, according to Eq. (4.24), for highly elliptically polarized laser fields, the electron can gain a large amount of energy (of order $2U_p$) through the variations of the laser envelope. However, this also implies that the electrons of high return energy depopulate completely the core region early after the laser field is turned on, and therefore the ground state of the electron is depopulated when the recolliding electrons return to the core. In order to overcome this difficulty, the atom is initiated in a superposition of states [109, 131].

We consider the initial wave function in a superposition of states computed using the imaginary time propagation and the Gram-Schmidt orthonormalization. The initial wave function is $\Psi(\mathbf{r}, 0) = c_0\psi_0(\mathbf{r}) + c_1\psi_1(\mathbf{r})$, where $\psi_0(\mathbf{r})$ and $\psi_1(\mathbf{r})$ are the ground state and the first excited state of the atom, respectively, c_0 and c_1 are constants. We use the soft Coulomb potential given by Eq. (1.8) for a = 0.26 and Z = 1. The energy of the ground state is $-I_0 \approx -24.6$ eV and the energy of the excited state is $-I_p \approx -5.7$ eV (softening parameter a = 0.26, close to He). The ionization potential of the excited state is such that $I_p < I_c$ $[I_c = 7.4 \text{ eV}$ with Eq. (4.17)], and therefore we expect the electrons in the excited state to undergo envelopeinduced recollisions after ionization. Figure 4.6 shows the intensity spectrum of the dipole acceleration $\langle \mathbf{a}(t) \rangle$ given by Eq. (4.25). The HHG intensity spectrum $|\mathbf{R}_{\text{HHG}}(\Omega)|^2$ is computed with

$$\mathbf{R}_{\rm HHG}(\Omega) = \mathcal{F}\left[\langle \mathbf{a}(t) \rangle W_{\rm HHG}(t)\right](\Omega),$$

where \mathcal{F} is the Fourier transform (see Appendix A). We use a Hanning window $W_{\text{HHG}}(t) = \sin(\pi t/T_f)^2$ for the HHG intensity spectrum, where $T_f = 10 \times 2\pi/\omega$ (ω the laser frequency) is the total integration time. The wave function of the electron $\Psi(\mathbf{r}, t)$ at time t is obtained from the two-dimensional TDSE

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = \left[-\frac{\Delta}{2} + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t)\right]\Psi(\mathbf{r},t).$$
(4.27)

The electric field in the LF is $\mathbf{E}(t) = E_0 f(t) / \sqrt{\xi^2 + 1} [\hat{\mathbf{x}} \cos(\omega t) + \hat{\mathbf{y}} \xi \sin(\omega t)]$. The laser envelope is trapezoidal 2–4–2. The wave function at time t is obtained using the real time propagation and the splitting operator method [11]. In Fig. 4.6, the intensity of the laser is such that $U_p/(\xi^2 + 1)$ is at the same harmonic for all ellipticities, i.e., $I = 8(\xi^2 + 1) \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$.

In the upper panels of Fig. 4.6, we observe a cutoff at the 100th harmonic which corresponds to the electrons with return energy about $3.17 U_p/(\xi^2 + 1)$ for $\xi = 0$, 0.25 and 0.5. For larger ellipticities, the strength of the 80-160th harmonics decreases significantly, and a second cutoff appears at the 80th harmonic corresponding to a return energy about $2.3 U_p$. We notice that this second cutoff is also observed at lower ellipticities. We observe that the peak intensity in the plateau region of the HHG spectrum does not change with respect to the ellipticity, in contrast to the predictions of the conventional three-step model [39]. This is observe that the influence of the portion of excited states in the initial wave function. In the lower panel of Fig. 4.6, we observe that the influence of the portion of excited states in the initial wave function influences weakly the peak intensity in the plateau region of the HHG spectrum.

4.2.2 Time-frequency analysis: Dominance of the envelope-driven recollisions for highly elliptic laser polarizations

Figure 4.7 shows the time-frequency analysis of $\mathbf{a}(t)$ used for computing the HHG spectra of Fig. 4.6. The time-frequency analysis $|\mathbf{R}_{\text{TFA}}(\Omega)|^2$ of the dipole acceleration is computed with



$$\mathbf{R}_{\mathrm{TFA}}(\Omega, \tau) = \mathcal{F}\left[\langle \mathbf{a}(t) \rangle W_{\mathrm{TGA}}(t-\tau)\right](\Omega),$$

Figure 4.7: Time-frequency analysis of the dipole acceleration $\mathbf{a}(t)$ at different ellipticities for a trapezoidal envelope 2-4-2. The parameters are the same as in the upper left panel of Fig. 4.6. The solid and dashed curves overlap and correspond to the return energy and energy difference of the electron ionizing at time $t_i = 0$ and returning at time t_r in the SFA.



Figure 4.8: HHG spectrum as a function of the ramp-up duration T_u/T in logarithmic scale for LP (top panel) and CP (lower panel). The laser envelope is trapezoidal of ramp-up duration T_u , plateau duration $6T - T_u$, and ramp-down duration 2T. The dashed and solid red lines are the SFA prediction of the energy difference and return energy, respectively.

where \mathcal{F} is the Fourier transform (see Appendix A). We use a window function $W_{\text{TFA}}(t) = \cos(\pi t/T)^4$ for -T/2 < t < T/2 and zero otherwise, with $T = 0.25 \times 2\pi/\omega$. In Fig. 4.7, we depict the return energy (solid grey curves) and the energy difference (dashed grey curves) of an electron ionizing at time $t_0 = 0$ as a function of the return time computed in the SFA. We tested numerically that the depicted curves are robust with respect to the ionization time. No qualitative difference is observed on the dashed and solid curves for $t_0 \lesssim 0.6T$. There is an excellent agreement between the grey curve and the maximum of the distribution of the time-frequency analysis. For $\xi = 0$, we observe that the time frequency analysis is dominated by the conventional recollision channel in which the electrons ionizes and return during the plateau [39, 100, 147, 163] with a maximum return energy around $3U_p$. We observe a minor contribution of the envelope-induced recollision channel, described in this chapter, with a maximum return around $2U_p$. For $\xi = 0.5$, we observe a major contribution of the envelope-induced recollision channel and a minor contribution of the conventional recollision channel [39, 100, 147, 163]. Around $\xi = 0.25$, the conventional recollision channel disappears [119] because of the large drift momentum of the electron at ionization [39]. Only the envelope-induced recollision channel, described in this chapter, persists for larger ellipticities. Also, the solid and dashed grey curves overlap almost completely for all return time and for all ellipticities, implying that the electron energy at ionization is very small.

In addition, we observe that the classical calculations of the distribution of the return energy as a function of the return time in Fig. 4.5 and the quantum calculation of the time-frequency analysis of the dipole acceleration in the upper panel of Fig. 4.7 exhibit the same patterns. The classical calculation in Fig. 4.5 is performed using a microcanonical distribution of the initial conditions with $H(\mathbf{r}, \mathbf{p}, 0) = -0.1$ a.u. The electrons ionize over the barrier early after the laser field is turned on. The agreement between the full classical and quantum calculations show that the nature of the envelope-induced recollisions is essentially classical.

4.2.3 Influence of the duration of the ramp-up: Control of the HHG cutoff

The importance of the small electron energy at ionization (or small initial velocity) is demonstrated in Fig. 4.8 which shows the HHG spectrum as a function of the harmonic number and the ramp-up duration T_u . The envelope is trapezoidal, the ramp-up is at $t \in [0, T_u]$, the plateau at $t \in [T_u, 6T]$ and the ramp-down at $t \in [6T, 8T]$. The solid and dashed red curves in the lower panel are the maximum return energy and energy difference in CP in the SFA, respectively, of the electron ionizing at time $t_0 = 0$. For LP, the conventional recollision scenario is dominant, the spectrum follows the well-known cutoff law $\Omega \approx 3.17 U_p + I_p$, and we

observe no significant influence of the ramp-up duration T_u . For CP, first, we observe that the HHG spectrum is very low for $T_u \leq T$. At the same time, we observe that $\mathcal{E}_i = \mathcal{E} - \Delta \mathcal{E}$ is large. It implies that the energy of the electron at ionization necessary to compensate its sideways drift must be large. However, the initial energy of the electron after ionization is small, and therefore the drift momentum of the electron pushes it away from the core without recolliding. For $T_u > T$, we observe that the HHG cutoff oscillates as a function of the ramp-up duration. We observe a good agreement between the SFA prediction and the HHG cutoff. Hence, for highly elliptically polarized laser fields, the HHG cutoff depends drastically on the laser envelope.

4.3 Applications to NSDI

Manifestations of recollisions in CP fields have been first observed experimentally in the enhancement of double ionization [61] [see the right panel of Fig. 4.1]. In Ref. [61], it is noticed that

"[...] all ionization should proceed above the Coulomb barrier and hence there exists no barrier for a tunneling mechanism"

In this section, we show that the envelope-induced recollisions, whose first step is over-the-barrier ionization, correspond to the recollisions responsible of the enhanced double ionization in CP [61, 111].

4.3.1 Double ionization probability curves

We consider an atom with two active electrons labeled by k = 1, 2, with phase space variables $(\mathbf{r}_k, \mathbf{p}_k)$. The classical Hamiltonian of the system [111] in the dipole approximation in the LF reads

$$H(\{\mathbf{r}_k, \mathbf{p}_k\}_{k=1}^2, t) = \sum_{k=1}^2 \left[\frac{|\mathbf{p}_k|^2}{2} - \frac{2}{\sqrt{|\mathbf{r}_k|^2 + a^2}} + \mathbf{r}_k \cdot \mathbf{E}(t) \right] + \frac{1}{\sqrt{|\mathbf{r}_1 - \mathbf{r}_2|^2 + 1}}.$$
(4.28)

The term $(|\mathbf{r}_1 - \mathbf{r}_2|^2 + 1)^{-1/2}$ corresponds to the electron-electron interaction, and is the fundamental ingredient of NSDI [110, 111]. Here, we use purely classical calculations [see Ref. [130] for a comparison between purely classical and purely quantum numerical measurements on double ionization with Hamiltonian (4.28) for LP fields for d = 1]. The atom is initiated with $H(\{\mathbf{r}_k, \mathbf{p}_k\}_{k=1}^2, 0) = \mathcal{E}_g$ using microcanonical generation of the initial conditions [2] (see Appendix D.1.2 for detail), with \mathcal{E}_g the ground state energy of the two-active electron atom. The softening parameter of the ion-electron potential *a* is chosen to avoid self-ionization and non-emptiness of the ground state [108]. The softening parameters used for each atom are shown in Tab. 4.1.

	He	Ne	Mg	Ar	Xe
\mathcal{E}_1 (in a.u.)	-0.90	-0.79	-0.28	-0.58	-0.45
\mathcal{E}_2 (in a.u.)	-2.00	-1.51	-0.55	-1.02	-0.77
\mathcal{E}_g (in a.u.)	-2.90	-2.30	-0.83	-1.60	-1.22
a (in a.u.)	0.8	1	3	1.5	1.8

Table 4.1: Energy of the ground state and softening parameters used in the manuscript for Hamiltonian (4.28) to model Mg, He, Xe, Ar and Ne.

The upper panel of Fig. 4.9 shows the double ionization probability curves for the Mg atom subjected to a CP laser field. The envelope of the laser is trapezoidal with T_u the duration of the ramp-up, $T_p = 6T - T_u$ the duration of the plateau duration and $T_d = 2T$ the duration of the ramp-down. The solid curves are the double ionization probability as a function of the intensity I computed using a distance criterion $|\mathbf{r}_k| > 100$ a.u. for k = 1, 2. The dash-dotted curves are the NSDI probability curves as a function of the intensity I. The NSDI occurs when one electron recollides and the other electron is bounded at the return of the recolliding electron. The dashed curves are the SDI probability curves as a function of the intensity I. The SDI occurs when both electrons ionize without recollisions. For $T_u = 0$ (grey curves), we observe that the NSDI probability is zero, and therefore the conventional recollision channel is absent, in agreement with Ref. [39]. For $T_u \ge 2T$ (colored curves), we observe a knee structure in the double ionization probability curves as a function of the laser intensity, in agreement with the experimental measurements in the right panel of Fig. 4.1. The contribution of the knee structure is a signature of the recollisions [72, 111].

The lower panels of Fig. 4.9 show the photoelectron momentum distribution of the electron for $T_u = 2T$, and $I = 10^{14} \text{ W} \cdot \text{cm}^{-2}$ (left panel) and $I = 5 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$ (right panel). In the left panel, the NSDI is dominant. We observe that the distribution of radial final momentum is peaked around $p_{r,1} = p_{r,2}$. The



Figure 4.9: Double ionization probability of Mg ($\mathcal{E}_g = -0.85$ a.u., a = 3) as a function of the laser intensity I (solid thick curves) solution of Hamiltonian (4.28) for a trapezoidal envelope with ramp-up duration T_u , plateau duration $T_p = 6T - T_u$, and ramp-down duration $T_d = 2T$. The grey, blue, pink and purple curves are for $T_u = 0$, 2T, 3T and 4T, respectively. The thin dashed-dotted and dashed curves are the probability of NSDI and SDI, respectively. Notice that the NSDI probability for $T_u = 0$ (dashed-dotted grey curve) is below 10^{-4} . The lower panels are the correlated photoelectron radial momentum distributions with $p_{r,j} = \mathbf{r}_j \cdot \mathbf{p}_j / |\mathbf{r}_j|$ for j = 1, 2. Left and right panels are for $I = 10^{14}$ W \cdot cm⁻² and $I = 5 \times 10^{14}$ W \cdot cm⁻² (indicated by the black vertical lines in the top panel), respectively. The dashed white lines indicate the equal energy sharing between electrons $p_{r,1} = p_{r,2}$. Intensity is in W \cdot cm⁻² and momenta are scaled by E_0/ω .



Figure 4.10: Typical NSDI of Hamiltonian (4.28): From the ground state to the return for a 2-4-2 laser envelope. Left panel: Trajectories in the RF (\tilde{x}, \tilde{y}) . The grey levels show the value of the zero-velocity surface of the outer electron during the plateau. Right panels: Red and blue dots are the two electrons. The trajectories projected along the position and energy at the ionization time of the recolliding electron t_0 , at $T_u = 2T$ (ramp-up duration) and at the return time of the recolliding electron t_r . The grey surface is the classical forbidden region for $\tilde{y} = 0$ [$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) < \mathcal{Z}(\tilde{\mathbf{r}}, t)$]. The light blue curve is the zero-velocity surface at time t = 0. The saddle point is the local maximum of the zero-velocity surface for negative \tilde{x} . Distances and energies are in a.u.

electrons likely share equally their energy [110]. In the right panel, the SDI is dominant. We observe that the distribution of the final radial momentum is composed of two peaks. The energy of each electron is \mathcal{E} , given in Eq. (4.16), with effective charge Z = 1 for the first ionized electron and Z = 2 for the second ionized electron. A typical SDI of Hamiltonian (4.28) is depicted in Fig. 4.11. Notice that the effective laser amplitude when the first electron is ionized is smaller than the effective laser amplitude when the second electron is ionized. Therefore, the energy of the second ionized electron is larger than the energy of the first ionized electron.



Figure 4.11: Typical SDI of Hamiltonian (4.28): From the ground state to the return for a 2-4-2 laser envelope. Left panel: Trajectories in the RF (\tilde{x}, \tilde{y}) . The grey levels shows the value of the zero-velocity surface of the outer electron during the plateau. Right panels: The trajectories projected along the position and energy at the first ionization, at the second ionization and at $T_u = 2T$ (end of the ramp-up). The grey surface is the classical forbidden region for $\tilde{y} = 0$ [$\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}, t) < \mathcal{Z}(\tilde{\mathbf{r}}, t)$]. The light blue curve is the zero-velocity surface at time t = 0. The saddle point is the local maximum of the zero-velocity surface for negative \tilde{x} . Distances and energies are in a.u.

4.3.1.1 Inner-outer electrons approximation

In Fig. 4.10, at $t_0 < t < t_r$, the red (recolliding) electron leaves the ionic core region, while the blue electron stays at the bottom of its zero-velocity surface. We label the recolliding (outer, red) electron k = 1, and the bound (inner, blue) electron k = 2. During its excursion outside the ionic core region, the nucleus charge

seen by the outer electron is screened by the inner electron. There is $|\mathbf{r}_1 - \mathbf{r}_2|^2 \approx |\mathbf{r}_1|^2 \gg a^2$ and

$$-\frac{2}{\sqrt{|\mathbf{r}_1|^2+a^2}}+\frac{1}{\sqrt{|\mathbf{r}_1-\mathbf{r}_2|^2+1}}\approx-\frac{1}{\sqrt{|\mathbf{r}_1|^2+a^2}}.$$

As a consequence, during the excursion of the recolliding electron in the continuum, Hamiltonian (4.28) can be written in terms of the sum of two Hamiltonians independent of each other

$$H({\mathbf{r}_k, \mathbf{p}_k}_{k=1}^2, t) \approx H_1({\mathbf{r}_1, \mathbf{p}_1, t}) + H_2({\mathbf{r}_2, \mathbf{p}_2, t})$$

Hamiltonian H_1 and H_2 are the Hamiltonian of the outer (recolliding) and inner (bond) electron, respectively. The expression of Hamiltonians H_1 and H_2 are given by Eq. (1.14) with the effective charge of the soft-Coulomb potential given by Z = 1 for the outer electron and Z = 2 for the inner electron. The inner-outer electrons approximation allows us to analyze the dynamics of each electron independently, i.e., in the SAE approximation. The energy H_1 and H_2 in the RF (\tilde{H}_1 and \tilde{H}_2) correspond to the energy of each electrons depicted in Figs. 4.10 and 4.11.

4.3.2 Ionization and return times of the recolliding electron

Figure 4.12 shows the distribution of ionization and return times of the outer electron $(\mathbf{r}_1, \mathbf{p}_1)$ for two different ramp-up envelopes: $f(t) = t/T_u$ (upper panel), and $f(t) = \sin(\pi t/2T_u)^2$ (lower panel) for $T_u = 3T$.



Figure 4.12: Distribution of ionization times t_0 of the recolliding electron leading to NSDI of Hamiltonian (4.28) for Mg (a = 3). The inset is the distribution of return times t_r of the recolliding electron leading to NSDI. The red dashed curve is the theoretical prediction Eq. (4.14) for $I_p = 0.3$ a.u. (corresponding to Mg). Upper panel and lower panels are for linear $f(t) = t/T_u$ and sinusoidal $f(t) = \sin(\pi t/2T_u)^2$ ramp-up, respectively, with $T_u = 3T$, $T_p = 6T$ and $T_d = 8T$.

We assume that the energy of the recolliding electron is initially $H_1(\mathbf{r}_1, \mathbf{p}_1, 0) = -0.3$ a.u. (corresponding to Mg) and therefore $I_p = 0.3$ a.u. The red dashed curves in Fig. 4.12 are the prediction of the ionization time of the recolliding electron given by Eq. (4.14). We observe that there is an excellent agreement between the prediction of Eq. (4.14) and the distribution of Fig. 4.12. Therefore, the recollisions inducing the enhancement in the double ionization probability of Mg are due to trajectories which ionize over the barrier, before the laser field reaches its peak amplitude. They correspond to envelope-induced recollisions described in this chapter.

In the insets of Fig. 4.12, we show the distribution of return times of the recollisions leading to NSDI as a function of the intensity of the laser field. We observe that the local maxima of the distribution depend on the intensity of the laser. Figure 4.13 shows the distribution of return times of the recollisions leading to NSDI for different envelope shapes and ramp-up durations. The ramp-up duration $T_u = 3T$ corresponds to the distributions in the inset of Fig. 4.12 summed over the intensity. The vertical lines indicates $t_r = (j + 1/4)T$ with $j \in \mathbb{N}$. We observe that the local maxima of the distribution depend on the shape of the laser envelope and on the ramp-up duration of the field. This local maxima depend on the distribution of the electron coordinates at ionization.

4.3.3 Breakdown of NSDI as a function of the laser frequency and the target species

Figure 4.14 shows the maximum of the NSDI probability \mathbb{P}_{max} as a function of $\omega - \omega_c$ for Mg, Ar, Ne and He, where ω_c is the critical frequency. The maximum of the NSDI probability \mathbb{P}_{max} corresponds to the maximum of the dash-dotted curves in Fig. 4.9. Practically, the critical frequency ω_c is the frequency for which $\mathbb{P}_{\text{max}} \approx 10^{-5}$. From the prediction of Eq. (4.17), the critical frequency for Mg is $\omega_c \approx 0.06$, Ar is $\omega_c \approx 0.18$, Ne is $\omega_c \approx 0.28$, and He is $\omega_c \approx 0.34$. The quantitative discrepancy between the numerical results and Eq. (4.17) may be due to our purely classical calculations, for which the energy sharing before ionization, where we assume that $H_k(\mathbf{r}_k, \mathbf{p}_k, t) \approx \mathcal{E}_k$ before ionization, is not fulfilled. However, Eq. (4.17) and the purely classical calculations agree qualitatively. For increasing first ionization potential, the NSDI probability decreases.

4.4 Conclusions

In sum, we have demonstrated the conditions under which a large portion of electrons are both ionized and later return to their parent ion regardless of the ellipticity of the laser field. The recollisions taking this recollision channel are referred to as *envelope-driven recollisions*. To recollide with a high return energy, the electron needs to ionize early after the laser field is turned on to benefit from the energy boost generated by the CP field, and not to drift away from the core. The GC model derived in Chap. 2 allowed us to determine under which conditions the envelope-driven recollisions can be observed in highly elliptically polarized laser fields. We have shown that, to undergo envelope-driven recollisions in highly elliptically polarized laser fields, the ionization potential of the electron must be such that $I_p < I_c$, where I_c is the critical ionization potential given by Eq. (4.17). For highly elliptically polarized laser fields, during its excursion in the continuum, the electrons gain energy mostly from the variations of the envelope of the driving laser. During their excursion, the energy of the envelope-driven recollisions can increase of $2U_p$, allowing for a return energy $\mathcal{E}_r = \mathcal{E}_0 + \Delta \mathcal{E}$ in excess of $2U_p$. Also, we observe a good agreement between the return energy of the electron in the SFA and the return energy of Hamiltonian (1.14).

We have shown that the envelope-driven recollisions can be used to produce HHG with atoms subjected to highly elliptically polarized laser fields. The atom must be initiated in a superposition of ground and excited states. The time-frequency analysis revealed that the envelope-driven recollisions persist for high ellipticities, while the conventional recollision channel disappears. In addition, we have shown that the envelope-driven recollisions are responsible for the enhanced double ionization probability of Mg subjected to CP fields observed in experiments [61]. The existence conditions of the envelope-driven recollisions allowed us to determine the conditions under which enhanced double ionization can be observed in atoms subjected to CP fields (see Fig. 4.4).



Figure 4.13: Probability density function of the return time of the recollisions leading to NSDI, summed over the intensity range $I \in [10^{13}, 10^{15}] \text{ W} \cdot \text{cm}^{-2}$ (i.e., insets of Fig. 4.12 projected along the return time axis), for a ramp-up duration $\tau = 2T$ (blue), 3T (pink) and 4T (purple). Upper and lower panel are with linear and sinusoidal ramp-up, respectively. The vertical lines indicate $t_r = (j + 1/4)T$ with $j \in \mathbb{N}$.



Figure 4.14: Maximum of the non-sequential double ionization probability (PNSDI) as a function of $\omega - \omega_c$ and a trapezoidal laser envelope 2–4–2, i.e., $T_u = 2T$, $T_p = 4T$ and $T_d = 2T$. The results of the simulation for the critical laser frequency ω_c are: $\omega_c \approx 0.04$ (Mg), $\omega_c \approx 0.09$ (Ar), $\omega_c \approx 0.14$ (Ne), and $\omega_c \approx 0.26$ (He).

Chapter 5

Structure of the phase space of the recolliding electron in step (ii)

When atoms are subjected to intense and elliptically polarized laser fields, multiple temporal and spatial scales arise from the competition between the strong laser and Coulomb forces. As seen throughout this manuscript, the same electron trajectory can involve multiple spatial and temporal scales. For instance, in Fig. 2.3 which shows a typical trajectory of Hamiltonian (1.14) for d = 1, we observe that the electron goes relatively far from the core (see white regions in Fig. 2.3), involving long-time and large-spatial scales dynamics. Also, the electron comes close to the core for short times (see grey regions in Fig. 2.3), involving short-time and small-spatial scales dynamics. When a unique spatial and a unique temporal scale is involved, we have shown in the previous chapters that reduced models derived from perturbative methods can approximate, relatively accurately, the motion of the electron. However, for trajectories involving multiple temporal and spatial scales, such as for instance the trajectory depicted in Fig. 2.3, perturbative methods have great difficulty to approximate accurately their motion in one piece. In particular, the presence of multiple temporal and spatial scales in the system makes the dynamics chaotic.

An intuitive way for analyzing the trajectories of the electron involving multiple temporal and spatial scales consists in reconstructing the trajectories of the electron using an appropriate reduced model for each interval of time interval during which a unique temporal and a unique spatial scale is involved. This procedure,



Figure 5.1: Illustration of the Poincaré section Σ of Hamiltonian (5.14) (2D case) corresponding to the stroboscopic map $\mathbf{P} : (\mathbf{r}, \mathbf{p}) \mapsto \varphi_0^T(\mathbf{r}, \mathbf{p})$ in 2D. The black line is the periodic orbit \mathcal{O}_2 projected in the plane (y, p_y) . The black dots are the fixed point of the periodic orbit under the mapping \mathbf{P} . The blue surface is an invariant torus of \mathcal{O}_2 of frequency ν as a function of time. The red and blue thick lines are two invariant tori under the mapping \mathbf{P} . The thin red and blue lines are trajectories along the red and blue torus, respectively. The inset is a zoom around the invariant tori projected on the plane (y, p_y) .

for instance, is used in Refs. [133, 37, 98] to describe the rescattering of the electrons. In these models, the collisions between the ion and the electron are considered as hard-sphere collisions. Between two collisions, the motion of the electron is described by the SFA. However, these procedures allow to analyze the behavior of individual trajectories only. Another way for analyzing the trajectories of the electron involving multiple temporal and spatial scales consists in nonperturbative methods. In this chapter, we study the invariant structures in phase space, such as for instance fixed points and periodic orbits, from a purely classical and nonlinear dynamical analysis. Electrons follow paths in phase space drawn by invariant structures. Computing and representing the invariant structures in phase space allows us to understand and predict the global dynamical behavior of the electrons.

Questions

- Which are the relevant and important invariant structures in phase space driving the electrons in two and three dimensions ?
- How can we compute numerically and represent these high-dimensional structures in high-dimensional phase space ?

Plan In this chapter, we study the structure of the phase space of Hamiltonian (1.14) using nonperturbative methods. The envelope of the laser is chosen as f = 1. The ion-electron interaction is the soft-Coulomb potential (1.8), $V(\mathbf{r}) = -Z(|\mathbf{r}|^2 + a^2)^{-1/2}$. Hamiltonian is given by Eq. (1.14) and the electric field reads

$$\mathbf{E}(t) = \frac{E_0}{\sqrt{\xi^2 + 1}} \left[\mathbf{e}_x \cos(\omega t) + \mathbf{e}_y \xi \sin(\omega t) \right].$$
(5.1)

The CEP is set to zero without loss of generality. The dimension of the phase space is 2d+1, due to the time dependence of the system. Since the laser envelope is f = 1, Hamiltonian (1.8) is *T*-periodic, with $T = 2\pi/\omega$ the period of the laser field. We take advantage of the periodicity of the Hamiltonian to analyze the electron dynamics under the *Poincaré map* (also called *stroboscopic map*)

$$\boldsymbol{\mathcal{P}}: (\mathbf{r}, \mathbf{p}) \mapsto \varphi_0^T(\mathbf{r}, \mathbf{p}), \tag{5.2}$$

where $\varphi_0^T(\mathbf{r}, \mathbf{p})$ is the Hamiltonian flow from time t = 0 to time t = T. Under Poincaré maps, the dimension of the phase space is decreased. In particular, under the stroboscopic map (5.2), the dimension of the phase space of Hamiltonian (1.14) is 2d. In addition, under Poincaré maps, the dimension of the invariant structures in phase space is decreased. Figure 5.1 illustrates the dimension reduction of the invariant structures under the stroboscopic map (5.2). The dimension is d = 2, and the phase space is projected on (y, p_y, t) . The Poincaré sections Σ associated with the stroboscopic map (5.2) are the white planes at $t/T = \{0, 1, 2\}$. The black curve shows the periodic orbit \mathcal{O}_S (see Fig. 5.2b) as a function of time. Under the stroboscopic map (5.2), the periodic orbit \mathcal{O}_S is a fixed point, as shown in the inset of Fig. 5.1. The blue surface represents a twodimensional invariant torus as a function of time. Clearly, we observe it is *T*-periodic. Under the Poincaré map (5.2), the *T*-periodic two-dimensional invariant torus becomes a one-dimensional invariant torus (closed curve), as shown in the inset of Fig. 5.1. Hence, using Poincaré maps, such as for instance the stroboscopic map (5.2), allows us to reduce the dimension of the phase space and invariant structures, and in the meantime, we keep all the relevant information on the electron dynamics.

In Sec. 5.1, we study the LP fields case for d = 1. We take advantage of the existence of the invariant subspace in LP fields along the polarization axis to analyze the dynamics for d = 1. This allows us to identify two periodic orbits candidates for driving the recollisions in higher dimensions, namely \mathcal{O}_S and \mathcal{O}_A . In Sec. 5.2, we study the LP fields for d = 2 and d = 3. We identify another set of relevant periodic orbits \mathcal{O}_F . We find that the invariant structures associated to \mathcal{O}_S drive the electron dynamics, while the dimension of the invariant structure associated with \mathcal{O}_A and \mathcal{O}_F is too small to drive the electron dynamics. We compute and represent the invariant structure associated with \mathcal{O}_S : The stable and unstable manifolds of a family of invariant tori. In Sec. 5.3, we study the EP fields case. In particular, we study the evolution of the periodic orbits \mathcal{O}_A and \mathcal{O}_F under the variation of the ellipticity. Finally, we show that for CP fields, the periodic orbit \mathcal{O}_F corresponds to a fixed point in the RF.

Publication

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Figure 5.2: Periodic orbits and associate stable and unstable manifolds of Hamiltonian (5.3) (case d = 1) for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, a = 1 and Z = 1. (a) RPOs and the associate fixed point in the Poincaré map \mathcal{P} . (b) SPO \mathcal{O}_S and the associate fixed point in the Poincaré map \mathcal{P} . (c) Final distance as a function of the initial conditions $(x_0, p_{x,0})$ for an integration t = 100T, i.e., $|\Pi \varphi_0^{100T}(x_0, p_{x,0})|$ with Π the projector along \mathbf{e}_x . The red and grey lines are the stable and unstable manifolds of the fixed point of \mathcal{O}_S under \mathcal{P} , respectively. The black and light grey lines are the stable and unstable manifolds of the RPOs, respectively.

5.1 Linearly polarized fields for d = 1

In this section, we investigate the structure of the phase space of an electron in an atom driven by a LP laser field for d = 1 (case d = 1). The Hamiltonian of the electron reads

$$H(x, p_x, t) = \frac{p_x^2}{2} - \frac{Z}{\sqrt{x^2 + a^2}} + xE_0 \cos(\omega t).$$
(5.3)

Figure 5.2b shows a periodic orbit of Hamiltonian (5.3) denoted \mathcal{O}_S . Figure 5.2c shows the final distance of the electron (at time t = 100T) as a function of the initial conditions x_0 and $p_{x,0}$ at time t = 0. The final distance of the electron from the core highlights the nonlinearities due to the strong ion-electron and laser-electron interaction. In particular, the blue regions and the paths with sensitivity with respect to the initial conditions correspond to configurations in phase space which lead to recollisions. The dark blue region, near the origin of phase space, is a region where the electron motion is bounded, and does not ionize.

In Fig. 5.2c, the orange dot indicates the location of the fixed point of \mathcal{O}_S under the Poincaré map \mathcal{P} . The red and grey curves are the stable and unstable manifolds of the SPO. Stable and unstable manifolds are invariant structures in phase space. Trajectories on the stable (resp. unstable) manifold come to (resp. go away from) \mathcal{O}_S . We observe that the stable manifold of the RPOs (red curves) reproduces the pattern of recollisions highlighted by the blue levels. In addition, these curves surround the dark blue region, where electrons are bounded. In phase space, the stable and unstable manifolds structure the phase space, by acting as barriers of the dynamics, and guide the electrons.

In this section, we identify relevant and important periodic orbits of Hamiltonian (5.3). In particular, we identify a family of recolliding periodic orbits (RPOs) \mathcal{O}_A (see Fig. 5.2a) and a saddle periodic orbit \mathcal{O}_S (see Fig. 5.2b). We show how to determine these periodic orbits from the integrable systems (i.e., the SFA and the field-free atom). We show how, from \mathcal{O}_A , the periodic orbits \mathcal{O}_A^{\pm} , also depicted in Fig. 5.2a, are created.

Under the Poincaré map (5.2), the periodic orbits \mathcal{O}_A , \mathcal{O}_A^{\pm} and \mathcal{O}_S are fixed points. We define the stable and unstable manifolds of fixed points. We show how the stable and unstable manifolds of \mathcal{O}_A , \mathcal{O}_A^{\pm} and \mathcal{O}_S structure the phase space and drive the electron.

5.1.1 One-dimensional recolliding periodic orbits (RPOs): \mathcal{O}_A

Recolliding periodic orbits [81, 80] (RPOs) are a class of periodic orbits which exhibit the same shape as typical recollisions (see Fig. 1.8 and Fig. 5.2): One piece of the periodic orbit is close to the ionic core $(|\mathbf{r}| \sim 1 \text{ a.u.})$, another piece is far from the ionic core $(|\mathbf{r}| \sim E_0/\omega^2)$. The electron can mimic the shape of the weakly unstable RPOs (i.e., the largest eigenvalue of the monodromy matrix is not too large). In the theory of Ref. [81, 80], recolliding electrons follow RPOs in phase space to go far from the core and come back.

In Ref. [80], the simplest RPO is determined from the SFA. In the SFA, the equations of motion of the electron are given by Eq. (2.2). The position and momentum of the electron are

$$x(t) = x_0 + p_{x,0}t + \frac{E_0}{\omega^2} \left[\cos(\omega t) - 1 \right], \qquad (5.4a)$$

$$p_x(t) = p_{x,0} - \frac{E_0}{\omega} \sin(\omega t), \qquad (5.4b)$$

where $(x(0), p_x(0)) = (x_0, p_{x,0})$ is the initial condition of the electron. Under the Poincaré map (5.2) \mathcal{P} , the initial position and momentum are $\mathcal{P}(x_0, p_{x,0}) = (x(T), p_x(T))$. The periodicity of the electric field implies $\mathcal{P}(x_0, p_{x,0}) = (x_0 + p_{x,0}T, p_{x,0})$. The momentum of the electron is constant under the mapping \mathcal{P} . The position of the electron is constant under the mapping \mathcal{P} only if

$$p_{x,0} = 0.$$
 (5.5)

Therefore, $(x_0, 0)$ is a fixed point of the mapping \mathcal{P} in the SFA. There exists an infinity of fixed points of \mathcal{P} labeled by x_0 . By substituting Eq. (5.5) into Eq. (5.4), we observe that these fixed points are associated with periodic orbits which are ellipses in phase space. Periodic orbits in the SFA for $x_0 = E_0/\omega^2$ are shown in Fig. 1.8b.

However, when the ion-electron interaction is fully taken into account, Hamiltonian (5.3) is not integrable. In this case, periodic orbits must be computed numerically. The trajectory with initial condition (x^*, p_x^*) is a T-periodic orbit of Hamiltonian (5.3) if and only if

$$\mathcal{P}(x^{\star}, p_x^{\star}) = (x^{\star}, p_x^{\star}).$$

The fixed point (x^*, p_x^*) is computed using Newton's method (see Appendix D.2.2.2 for details on the numerical computation). We use the fixed point of the mapping \mathcal{P} in the SFA as initial guess for the Newton's method, i.e., $(x_0, p_{x,0}) = (E_0/\omega, 0)$. In Fig. 5.2a, we show \mathcal{O}_A computed numerically. We observe that, in contrast to its associated periodic orbit in the SFA, \mathcal{O}_A is not exactly an ellipse in phase space. In particular, we observe that the absolute value of its momentum increases drastically when it crosses x = 0 due to the ion-electron interaction.

5.1.1.1 Creation of RPOs through a bifurcation

The linear stability of the periodic orbit \mathcal{O}_A is given by the eigenvalues λ_j and the eigenvectors \mathbf{v}_j of the monodromy matrix \mathbf{M}_T . The fixed point of \mathcal{O}_A under \mathcal{P} is (x^*, p_x^*) , and the monodromy matrix is

$$\mathbf{M}_T(x^\star, p_x^\star) = \mathcal{J}_0^T(x^\star, p_x^\star), \tag{5.6}$$

with j = 1, 2, where $\mathcal{J}_0^T(\mathbf{z}) = \partial \varphi_0^T(\mathbf{z}) / \partial \mathbf{z}$ is the tangent flow (see Appendix C.1 for details) and $\mathbf{z} = [x, p_x]^\top$. For Hamiltonian systems, the eigenvalues of the monodromy matrix are conjugated, such that $\lambda_1 = 1/\lambda_2$ and $\lambda_1 = \lambda_2^*$ (see Appendix C.1). In the SFA, the evolution of the tangent flow is given by $\dot{\mathcal{J}}_0^t(x_0, p_{x,0}) = \mathbf{A}\mathcal{J}_0^t$ with $\mathbf{A} = [0, 1; 0, 0]$, and the monodromy matrix is given by $\mathbf{M}_T(x_0, p_{x,0}) = \exp(\mathbf{A}T)$. The eigenvalues of the monodromy matrix are unity. Hence, in the SFA, all periodic orbits are *parabolic*. For decreasing intensity, \mathcal{O}_A changes shapes, and also stability. In the lower panel of Fig. 5.3, the orange curve is the largest absolute eigenvalue of the monodromy matrix of \mathcal{O}_A as a function of the laser intensity (for relatively low intensities). The upper panels of Fig. 5.3 show Poincaré sections of Hamiltonian (5.3) under \mathcal{P} for (left panel) $I = 10^{13} \text{ W} \cdot \text{cm}^{-2}$ and (right panel) $I = 2 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$. For $I = 10^{13} \text{ W} \cdot \text{cm}^{-2}$, in the upper left panel of Fig. 5.3, the orange dot is the fixed point of \mathcal{O}_A under under \mathcal{P} . We observe that the orange



Figure 5.3: Upper panels: Stroboscopic plot for LP of Hamiltonian (5.3) under the Poincaré map \mathcal{P} defined by Eq. (5.2) for a = 1 and Z = 1. In the left panel at $I = 10^{13}$ W.cm⁻², right panel at $I = 2 \times 10^{13}$ W.cm⁻². The crosses shows the position of \mathcal{O}_A on the plots. Lower panel: Largest absolute eigenvalue of \mathcal{O}_A as a function of the laser intensity I.

dot is surrounded by invariant curves (black curves). The invariant curves create a stable region in phase space. Outside this region, we observe dots, where the dynamics is rather chaotic. Hence, periodic orbits can structure regions in phase space. Their linear stability encodes the structure of the phase space in their neighborhood.

In order to understand how the linear stability of the periodic orbits encodes the structure of the phase space in their neighborhood, we consider a trajectory $(x(t), p_x(t))$ initiated close to the periodic orbit \mathcal{O}_A , at $(x(0), p_x(0)) = (x^* + \delta x, p_x^* + \delta p_x)$, and such that $|\delta x| \ll 1$ and $|\delta p_x| \ll 1$. At k iterations of the Poincaré map, the trajectory is $\mathcal{P}^k(x^* + \delta x, p_x^* + \delta p_x) \approx \mathcal{P}^k(x^*, p_x^*) + \mathbf{M}_T(x^*, p_x^*)^k[\delta x, \delta p_x]^\top$. Hence, after k laser cycles, the deviation between the trajectory and the periodic orbit is given by $[x(T), p_x(T)]^\top - [x^*, p_x^*] \approx$ $\mathbf{M}_T(x^*, p_x^*)^k[\delta x, \delta p_x]^\top$. For simplicity, we assume the monodromy matrix \mathbf{M}_T to be diagonal. The deviation of the electron and the periodic orbit after k laser cycles is given by $x(T) - x^* \approx \lambda_1^k \delta x$ along the position component, and $p_x(T) - p_x^* \approx \lambda_2^k \delta p_x$ along the momentum component. We observe that, depending on the eigenvalues, the behavior of the trajectory $(x(t), p_x(t))$ is different.

- If $\lambda_1 = \lambda_2 = \pm 1$, the electron stays at the same distance from the periodic orbit, and in the same direction as it was initially. The stability of the periodic orbit is referred to as *parabolic*. Such periodic orbit corresponds to the RPO in the SFA with initial condition $(x_0, 0)$. In order to determine if the orbit is stable or unstable, one needs to compute the nonlinear stability of the periodic orbit.
- If $|\lambda_1| = |\lambda_2| = 1$ and $\lambda_1 \neq \pm 1$ (i.e., the eigenvalues are complex and $\lambda_1 = \lambda_2^*$), the electron stays at the same distance from the periodic orbit, and turns around it. The stability of the periodic orbit is referred to as *center*. The dynamics of the electron close to the periodic orbit is *stable*. For instance, \mathcal{O}_A is center for $I = 10^{13} \text{ W} \cdot \text{cm}^{-2}$. It corresponds to the orange marker in the upper left panel of Fig. 5.3. The invariant curves surrounding the orange marker are trajectories which turn around the periodic orbit between two iterations of the Poincaré map \mathcal{P} . On the invariant curves, the motion of the electron is quasi-periodic.
- If $|\lambda_j| \neq 1$ (i.e., the eigenvalues are reals and $\lambda_1 = 1/\lambda_2$), the electron goes away from the periodic orbit along one direction, and gets closer to the periodic orbit along the other direction. The stability of the periodic orbit is referred to as *saddle*. The dynamics of the electron close to the periodic orbit is *unstable*. In this case, there exist a stable and an unstable manifold associated with the periodic orbit.

In the lower panel of Fig. 5.3, we observe that \mathcal{O}_A is saddle at $I \sim 2 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$. For $I \sim 1.8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, there is a bifurcation. In the upper right panel of Fig. 5.3, we observe that the stable region still exists. However, here, the invariant curves form a "8" shape. At the node of the eight, there is the fixed point of \mathcal{O}_A indicated by an orange cross. \mathcal{O}_A is saddle, and trajectories go away from \mathcal{O}_A along branches of the eight-shaped invariant curves, and come closer to \mathcal{O}_A in other branches of the eight-shaped invariant curves, and come closer to \mathcal{O}_A in other branches of the eight-shaped invariant curves. Around the center of the loops of the eight-shaped invariant curves, there are two periodic orbits created when the stability of \mathcal{O}_A bifurcates. These periodic orbits are indicated by blue dots in the upper right panel of Fig. 5.3, and are referred to as \mathcal{O}_A^- (left blue dot) and \mathcal{O}_A^+ (right blue dot). The blue markers are surrounded by elliptical invariant curves, so we expect their stability to be center. Indeed, in the lower panel of Fig. 5.3, we observe that the largest eigenvalue of these orbits is unity, and therefore these orbits are center at this intensity. The periodic orbits \mathcal{O}_A^{\pm} are shown in Fig. 5.2a for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. We observe they are asymmetric with respect to x = 0, and symmetric with one another with respect to the symmetry $(x, p_x, t) \mapsto (-x, -p_x, t + T/2)$. At t = 0, \mathcal{O}_A^- is closer to the core than \mathcal{O}_A^+ . At t = T/2, \mathcal{O}_A^- is further away from the core than \mathcal{O}_A^+ .

In sum, we have identified a family of RPOs for d = 1, referred to as \mathcal{O}_A . The mother periodic orbit \mathcal{O}_A originates from the SFA. When the intensity of the laser changes, the linear stability of \mathcal{O}_A changes, and affects the structure of the phase space in its neighborhood. In particular, when the stability of \mathcal{O}_A goes by saddle to center, for $I \sim 1.8 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$, two auxiliary RPOs are created, namely \mathcal{O}_A^+ and \mathcal{O}_A^- .

5.1.2 Saddle periodic orbits (SPOs): \mathcal{O}_S



Figure 5.4: Poincaré section x = 0 and $\dot{x} > 0$ of Hamiltonian (5.3) (case d = 1) for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, a = 1 and Z = 1. Right panel: Zoom of the left panel. Crosses (resp. dots) are the intersection of the saddle (resp. center) SPOs with the Poincaré surface x = 0 and $\dot{x} > 0$. The red and blue markers are for periodic orbits of resonance 1:7 and 1:9, respectively.

Another important family of periodic orbits of Hamiltonian (5.3) are the saddle periodic orbits (SPOs). The location of the SPOs in phase space is revealed by the Poincaré section x = 0. In Fig. 5.4, we show the Poincaré section of Hamiltonian (5.3) for x = 0 and $\dot{x} > 0$. For $p_x \leq 1$, we observe invariant curves where the motion of the electron is quasi-periodic. The invariant curves in this representation correspond to the invariant curves observed near the origin of phase space in the upper panels of Fig. 5.3. In the right panel of Fig. 5.4, for $p_x \leq 1$, we observe regions where invariant curves are closed, such as the invariant curves surrounding the blue dots. The blue dots corresponds to the fixed points of one center SPO of resonance 1 : 9. There are nine dots, i.e., during one laser cycle, the periodic orbit crosses the Poincaré section x = 0 and $\dot{x} > 0$ nine times. Between two adjacent blue dots, we observe eight-shaped invariant curves. At the node of the eight-shaped invariant curves, there are blue crosses corresponding to the fixed points of one saddle SPO of resonance 1 : 9. For $p_x \gtrsim 1$, electrons are unbounded and ionize. For $p_x \sim 1$, we observe regular islands.

In these islands, the red circles are the fixed points of one center periodic orbit of resonance 1 : 7. Outside the last invariant curve of each regular island, there are dense chaotic regions. In these regions, the electrons spend several laser cycles before ionizing. In NSDI, these regions have been identified to be responsible for the recollision excitation with subsequent ionization [113, 112] (RESI). Near the chaotic regions, there are the fixed points of one saddle SPO indicated by the red crosses. This periodic orbit is referred to as \mathcal{O}_S , and is depicted in Fig. 5.2b for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. The chaotic regions are induced by the intersection of the stable and unstable manifolds of \mathcal{O}_S .

Here, we follow the procedure described in Refs. [113, 112] to approximate the location of the SPOs of resonance 1 : m in Fig 5.4. Close to the core, the laser interaction is small compared to the ion-electron interaction. We neglect the laser interaction. In action angle variables, in absence of electric field, Hamiltonian (5.3) reads [113, 112] $H_{\text{free}}(A) = \mathcal{E}_0 + \omega_0 [\exp(\gamma A) - 1]/\gamma$, where $\omega_0 = \sqrt{Z/a^3}$, $\gamma = -9/(8\sqrt{Za})$ and $\mathcal{E}_0 = -Z/a$. The action is defined by $A = (1/2\pi) \oint p_x dx$ and its canonically conjugate angle is θ . The frequency of the angle is given by $\nu_f = \dot{\theta} = \partial H_{\text{free}}/\partial A$. Hence, $\nu_f = \gamma(\mathcal{E} - \mathcal{E}_0) + \omega_0$. The resonance condition 1 : m reads $m\nu_f = \omega$, with $m \in \mathbb{Z}$. In Fig. 5.4, we also observe minor resonances k : m with k > 1. Here, we focus on the dominant resonances for which k = 1. Using Cartesian coordinates for the energy of the electron \mathcal{E} in the field-free atom, one obtains an approximation for the resonance condition of the SPOs

$$\omega \approx m\gamma \left[\frac{p_x^2}{2} - \frac{Z}{\sqrt{x^2 + a^2}} - \mathcal{E}_0\right] + \omega_0.$$
(5.7)

This provides a family of periodic orbits in the neighborhood of the ionic core, labeled by the integer m. The cases m = 7 and m = 9 correspond to the red and blue markers in the right panel of Fig. 5.4. We notice that the resonance 1 : 1 corresponds to the periodic orbit \mathcal{O}_A . From Eq. (5.7), for x = 0, we obtain

$$p_x \approx \pm \sqrt{2 \frac{\omega - \omega_0}{m\gamma}}$$

5.1.3 Invariant manifolds structuring the phase space of the recolliding electron

For d = 1, the phase space of Hamiltonian (5.3) is three-dimensional and periodic orbits are one-dimensional. Periodic orbits are codimension two with the phase space, and as a consequence, they do not partition the phase space. However, as we have shown in the previous sections, there are invariant structures associated with the periodic orbits, such as for instance invariant curves. In particular, stable and unstable manifolds of unstable periodic orbits are two-dimensional and partition the phase space of the electron for d = 1. We denote (x^*, p_x^*) the fixed points of the periodic orbits \mathcal{O}_A , \mathcal{O}_A^{\pm} and \mathcal{O}_S under \mathcal{P} , indicated by crosses in the lower panel of Fig. 5.2. The *invariant manifolds* of (x^*, p_x^*) correspond to a set of coordinates such that

• The points in the stable manifold converge to the fixed point (x^*, p_x^*) if one iterated \mathcal{P} infinitely forward in time, i.e.,

$$\mathcal{W}^{s}(x^{\star}, p_{x}^{\star}) = \left\{ (x, p_{x}) \in \mathbb{R}^{N} \mid \lim_{k \to \infty} \mathcal{P}^{k}(x, p_{x}) = (x^{\star}, p_{x}^{\star}) \right\}.$$
(5.8)

• The points in the *unstable manifold* converge to the fixed point (x^*, p_x^*) if one integrates infinitely backward in time, i.e.,

$$\mathcal{W}^{u}(x^{\star}, p_{x}^{\star}) = \left\{ (x, p_{x}) \in \mathbb{R}^{N} \mid \lim_{k \to \infty} \mathcal{P}^{-k}(x, p_{x}) = (x^{\star}, p_{x}^{\star}) \right\}.$$
(5.9)

The existence of invariant manifolds of fixed points is demonstrated in Ref. [32]. In the lower panel of Fig. 5.2, the black and light grey curves are the stable and unstable manifolds of \mathcal{O}_A and \mathcal{O}_A^{\pm} . The stable and unstable manifolds are computed numerically with Hobson's method [74] (see Appendix D.2.3 for details on the numerical computations). The invariant manifolds of \mathcal{O}_A and \mathcal{O}_A^{\pm} are connected since \mathcal{O}_A^{\pm} originate from \mathcal{O}_A . The black curve is of codimension one with the reduced phase space. As a consequence, it creates boundaries for the trajectories of the electron. The electrons are guided by the black curve, and cross the axis x = 0. It drives the electrons away from the core, and brings them back to recollide. In the lower panel of Fig. 5.2, we observe that the black curve reproduces the pattern of recollisions. Therefore, the stable manifold of \mathcal{O}_A and \mathcal{O}_A^{\pm} structures the phase space, and drives the recollisions.

The stable manifold of \mathcal{O}_S also structures the phase space, and drives the recollisions. In Fig. 5.2c, the red and dark grey curves are the stable and the unstable manifolds of the fixed point of \mathcal{O}_S under \mathcal{P} . We



Figure 5.5: Final distance of the electron of Hamiltonian (5.14) (case d = 2) as a function of the initial conditions $(x_0, p_{y,0}, y_0 = p_{x,0} = 0)$ for $I = 3 \times 10^{14}$ W · cm⁻², a = 1 and Z = 1. The orange marker is the fixed point of \mathcal{O}_S under the Poincaré map \mathcal{P} . The black crosses are the fixed points of \mathcal{O}_A and \mathcal{O}_A^{\pm} under \mathcal{P} . The green crosses are the fixed points of \mathcal{O}_F , \mathcal{O}_F^{\pm} clockwise (lower markers) and \mathcal{O}_F , \mathcal{O}_F^{\pm} anticlockwise (upper markers) under \mathcal{P} . The red dots show pieces of the intersection between the plane of initial conditions and the stable manifolds of the family of invariant tori of \mathcal{O}_S .

observe that it also reproduces the pattern of recollisions corresponding to the blue levels. In addition, we observe that the red and dark grey curves surround the dark blue region where electrons are bounded. The invariant manifolds of \mathcal{O}_S separate electrons which are bounded from electrons which ionize.

Therefore, for d = 1, the stable and unstable manifolds of the RPOs \mathcal{O}_A and the SPO \mathcal{O}_S partition the phase space and drive the electron dynamics, and in particular, the recollisions. Their stable manifolds are very similar. The periodic orbits \mathcal{O}_A and \mathcal{O}_S are, as a consequence, relevant candidates for driving the electron dynamics in higher dimensions, for d > 1.

5.2 Linearly polarized fields in higher dimensions

In this section, we study the phase space of an electron in an atom driven by a LP laser field for d > 1. In the SFA, for LP fields, the dynamics of the electron transverse to the polarization axis is

$$y(t) = y_0 + p_{y,0}t. (5.10)$$

If the electron ionizes with a small momentum $p_{y,0}$, it drifts away from the core without recolliding. A first difficulty when the dimension of the phase space is increased is that Poincaré sections can no longer be represented in two dimensions for d > 1. As a consequence, it is more delicate to draw a global picture of the electron dynamics. One way to roughly draw the global dynamics of the electron is to perform analyses of observables with respect to the initial conditions in a slice of phase space. Figure 5.5 shows the final distance of the electron from the core for Hamiltonian (1.14) as a function of the initial conditions $(x_0, p_{y,0})$ for $y_0 = p_{x,0} = 0$. Like for d = 1, we observe paths with a sensitivity with respect to the initial conditions which lead to recollisions. In particular, we observe paths for relatively large $p_{y,0}$, in contrast with the prediction of the SFA given by Eq. (5.10). The electron can come back to the core, even if $p_{y,0}$ is relatively large, due to the contribution of the Coulomb potential. We examine its role through the invariant structures associated with the periodic orbits.

The second difficulty arising, when the dimension of the phase space is increased, is the increase of the dimension of the relevant and important invariant structures. In particular, for d = 2, in order to participate

the phase space, the invariant structures must be at least four-dimensional. Under the Poincaré map, they must be at least three-dimensional. Computing and representing three-dimensional objects is numerically challenging. In this section, we determine another family of RPOs which exist exclusively for d > 1, referred to as \mathcal{O}_F . We show that, for d = 2 (five-dimensional phase space), the invariant structures associated with \mathcal{O}_A and \mathcal{O}_F are three-dimensional, and as a consequence, they cannot partition the phase space. In contrast, we show that the four-dimensional invariant structure associated with \mathcal{O}_S partitions the phase space and drives the recollisions.

5.2.1 Two-dimensional recolliding periodic orbits (RPOs): \mathcal{O}_F

In order to determine the family of RPOs \mathcal{O}_F , first, we consider the field-free atom where $E_0 = 0$. We consider d = 2 for which the electron evolves on the polarization plane $(\mathbf{e}_x, \mathbf{e}_y)$. In polar coordinates, where $\mathbf{r} = \mathbf{e}_x r \cos \theta + \mathbf{e}_y r \sin \theta$ and $\mathbf{p} = \mathbf{e}_x [p_r \cos \theta - (p_\theta/r) \sin \theta] + \mathbf{e}_y [p_r \sin \theta + (p_\theta/r) \cos \theta]$, Hamiltonian (1.14) reads

$$H_{\text{free}}(r,\theta,p_r,p_\theta) = \frac{p_r^2}{2} + \frac{p_\theta^2}{2r^2} - \frac{Z}{\sqrt{r^2 + a^2}}.$$
(5.11)

Hamiltonian (5.11) is invariant under time translation and rotation $\theta \mapsto \theta + \vartheta$, and as a consequence, the energy $\mathcal{E} = H_{\text{free}}(r, \theta, p_r, p_\theta)$ and the angular momentum $\ell = p_\theta$ are conserved. The goal is to determine a purely two-dimensional periodic orbit which persists when the laser field is turned on. The simplest periodic orbits of Hamiltonian (5.11) are circular periodic orbits, such that $\dot{r} = \dot{p}_r = 0$. The radius of the circular periodic orbits, denoted r_0 , is related to the angular momentum by

$$\ell^2 = \frac{Zr_0^4}{(r_0^2 + a^2)^{3/2}}.$$
(5.12)

In addition, we assume that the periodic orbits of Hamiltonian (5.11), which can persist once the laser field is turned on, have the same period than the laser field. The circular periodic orbits of angular frequency ω are such that $\dot{\theta} = \ell/r_0^2 = \pm \omega$. This implies that $\ell = \pm \omega r_0^2$. By substituting this condition in Eq. (5.12), we obtain that the radius of the circular periodic orbits with angular frequency $\dot{\theta} = \pm \omega$ is

$$r_0^2 = \left(\frac{Z}{\omega^2}\right)^{2/3} - a^2.$$
(5.13)

The condition $(Z/\omega^2)^{1/3} \ge a$ is always fulfilled for the parameters we use in this manuscript. The initial conditions $\mathbf{r} = r_0(\mathbf{e}_x \cos \theta_0 + \mathbf{e}_y \sin \theta_0)$ and $\mathbf{p} = (p_\theta/r)(-\mathbf{e}_x \sin \theta_0 + \mathbf{e}_y \cos \theta_0)$ correspond to a fixed point under \mathcal{P} such that $\mathcal{P}(\mathbf{r}, \mathbf{p}) = (\mathbf{r}, \mathbf{p})$. There exists an infinity of fixed points of \mathcal{P} labeled by θ_0 . This family of orbits is referred to as \mathcal{O}_F .

For d = 2, like for d = 1 (see Sec. 5.1.1), periodic orbits are created when periodic orbits bifurcate for varying parameters. For d = 1, this phenomenon is illustrated for varying intensity in Fig. 5.3. The upper panel of Fig. 5.6 shows the bifurcation diagram of the RPOs of the families \mathcal{O}_A and \mathcal{O}_F . The periodic orbit \mathcal{O}_F originates from the field-free atom approximation, for which $I \to 0$. In this approximation, \mathcal{O}_F is circular in configuration space and is *parabolic*. For increasing intensity, \mathcal{O}_F changes shape, it shrinks along \mathbf{e}_y , as observed in the lower right panel of Fig. 5.6. For all intensities, \mathcal{O}_F stays outside the invariant subspace.

For $I \leq 10^{11} \text{ W} \cdot \text{cm}^{-2}$, \mathcal{O}_F is center-center. At $I \sim 10^{11} \text{ W} \cdot \text{cm}^{-2}$, there is a first bifurcation of \mathcal{O}_F and the periodic orbits $\mathcal{O}_F^{\uparrow\downarrow}$ are created. Right after, there is a second bifurcation of \mathcal{O}_F and the periodic orbits \mathcal{O}_F^{\pm} are created. The periodic orbits $\{\mathcal{O}_F, \mathcal{O}_F^{\pm}, \mathcal{O}_F^{\uparrow\downarrow}\}$ are degenerate twice due to the symmetry $(x, y, p_x, p_y, t) \mapsto (x, -y, p_x, -p_y, t)$, specific to the LP case. Hence, for each of these periodic orbits, there is a clockwise and an anticlockwise periodic orbit, as observed in Fig. 5.5, for instance. At $I \sim 10^{16} \text{ W} \cdot \text{cm}^{-2}$, \mathcal{O}_F^{\pm} join the RPOs \mathcal{O}_A^{\pm} due to the strong laser interaction which shrinks the orbits along \mathbf{e}_y . At $I \sim 10^{17} \text{ W} \cdot \text{cm}^{-2}$, $\mathcal{O}_F^{\uparrow\downarrow}$ and \mathcal{O}_F join the RPO \mathcal{O}_A .

For $I \sim [10^{13}, 10^{16}] \text{W} \cdot \text{cm}^{-2}$, we observe that the family of RPOs \mathcal{O}_A and the family of RPOs \mathcal{O}_F are real saddle. Therefore, all eigenvalues of the RPOs are reals, and in particular $\lambda_1, \lambda_2 \in \mathbb{R}^2$. Therefore, the invariant structures associated with the RPOs of the families \mathcal{O}_A and \mathcal{O}_F are two-dimensional stable and unstable manifolds under the Poincaré map \mathcal{P} . In the extended five-dimensional phase space, these invariant structures correspond to three-dimensional stable and unstable manifolds. They are of codimension two with the phase space. Therefore, the invariant structures associated with these RPOs cannot partition the phase space and cannot drive the electrons for d > 1.



Figure 5.6: The laser ellipticity is $\xi = 0$ (LP case), a = 3, Z = 1 and d = 2. Upper panel: Bifurcation diagram as a function of the laser intensity (two largest absolute eigenvalues $|\lambda_1|$ and $|\lambda_2|$, such that $|\lambda_1| > |\lambda_2|$) of the RPOs of Hamiltonian (5.14) depicted in the lower panels. Lower panels: RPOs for $I = 10^{15}$ W · cm⁻². Left and right panels are the RPOs of the family \mathcal{O}_A (RPOs belonging to the invariant subspace for $\xi = 0$) and \mathcal{O}_F , respectively. All quantities are in a.u.

5.2.2 Recollisions in higher dimensions: A family of invariant tori drives the electron dynamics

We consider d = 2 and $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. At $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, the eigenvalues of \mathcal{O}_A are the largest $(\lambda_1 \sim 1000, \text{ see Fig. 5.6})$. The phase-space variables of the electron in Cartesian coordinates are (x, y, p_x, p_y) . The Hamiltonian reads

$$H(x, y, p_x, p_y) = \frac{p_x^2}{2} + \frac{p_y^2}{2} - \frac{Z}{\sqrt{x^2 + y^2 + a^2}} + xE_0\cos(\omega t).$$
(5.14)

For d = 2 and $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, as mentioned in Sec. 5.2.1, the invariant structures associated with the RPOs \mathcal{O}_A and \mathcal{O}_F (two-dimensional stable and unstable manifolds) is two-dimensional under \mathcal{P} . These invariant structures are of codimension two with the four-dimensional reduced phase space (phase space under \mathcal{P}), and as a consequence, they cannot partition the phase space and cannot drive the electrons. In contrast, for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, the stability of \mathcal{O}_S is *center-saddle*: The eigenvalues of the monodromy matrix of \mathcal{O}_S are $\lambda_1 \sim 7$ (largest absolute eigenvalue) and $|\lambda_2| = 1$ (second largest absolute eigenvalue). The invariant structure associated with \mathcal{O}_S is three-dimensional under \mathcal{P} . The invariant structure associated with \mathcal{O}_S is a set of stable and unstable manifolds of a family of invariant curves, denoted \mathcal{W}_F^s and \mathcal{W}_F^u , respectively.

In Fig. 5.7, the orange dot is the fixed point of \mathcal{O}_S under \mathcal{P} , denoted (x^*, p_x^*) . Due to the center component of \mathcal{O}_S , there is a family of invariant curves associated with \mathcal{O}_S . In the neighborhood of (x^*, p_x^*) , these invariant curves are close to the plane defined by the eigenvectors associated with the complex eigenvalues of the monodromy matrix of \mathcal{O}_S . In Fig. 5.7, the black curves are invariant curves of this family. The invariant curves of \mathcal{O}_S are denoted $\mathbf{x}(\theta)$ and parametrized by $\theta \in \mathbb{T}$ (\mathbb{T} corresponds to $\mathbb{R}/2\pi\mathbb{Z}$). If the curve is invariant under \mathcal{P} , there exists $\nu \in \mathbb{R} \setminus 2\pi\mathbb{Q}$ such that

$$\mathcal{P}(\mathbf{x}(\theta)) = \mathbf{x}(\theta + \nu). \tag{5.15}$$

The numerical computation of the invariant curves $\mathbf{x}(\theta)$ is performed using the Fourier representation method [79], and is detailed in Appendix D.2.4. Under \mathcal{P} , the invariant curves associated with \mathcal{O}_S are one-dimensional. The dimension of the family of invariant curves is *fractal*. However, numerically, the family of invariant curves is two-dimensional. Due to the saddle component of \mathcal{O}_S , there are stable and unstable manifolds associated with each invariant curve of \mathcal{O}_S . In Fig. 5.7, the red and grey surfaces are the stable and unstable manifolds of the black invariant curves, respectively. For each invariant curve, the stable and unstable manifolds are two-dimensional. As a consequence, the set of stable and unstable manifolds of the family of invariant curves associated with \mathcal{O}_S (\mathcal{W}_F^s and \mathcal{W}_F^u , respectively) is three-dimensional. Hence, \mathcal{W}_F^s and \mathcal{W}_F^u are codimension one with the phase space, and are relevant candidates to drive the electrons. In order to determine if \mathcal{W}_F^s and \mathcal{W}_F^u drive the dynamics of the electron, we must show that \mathcal{W}_F^s and \mathcal{W}_F^u reproduce the recollision patterns in Fig. 5.5, highlighted by the blue levels.



Figure 5.7: The orange marker is the fixed point of the \mathcal{O}_S of Hamiltonian (5.14) (case d = 2) under \mathcal{P} for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, $\xi = 0$ (LP case), a = 1 and Z = 1. The black curves are four invariant tori. The grey and red surfaces are the unstable \mathcal{W}_F^u and stable \mathcal{W}_F^s manifolds of the invariant tori, respectively. The dark grey and dark red curves are trajectories along the unstable and stable manifolds, respectively.

In order to clearly show that \mathcal{W}_F^s and \mathcal{W}_F^u reproduce the recollision patterns in Fig. 5.5, we compute the intersection between these invariant structures and the plane of initial conditions $(x_0, p_{y,0}, y_0 = p_{x,0} = 0)$. We label each invariant curve of the family by σ such that one invariant curve is denoted $\mathbf{x}_{\sigma}(\theta)$. We focus on the computation of \mathcal{W}_F^s . First, we consider the stable manifold of one invariant curve $\mathbf{x}_{\sigma}(\theta)$. The real eigenvalue of the normal behavior of the invariant curve smaller than unity is denoted λ , and its associated eigenvector is denoted $\Psi_{\sigma}^s(\theta)$ (see Appendix D.2.4 for detail on the computation of the normal behavior of invariant curves). The eigenvector $\Psi_{\sigma}^s(\theta)$ corresponds to the linear approximation of the stable manifold close to the invariant curve. The initial condition of a trajectory initiated in the fundamental domain of the stable manifold associated with the invariant curve $\mathbf{x}_{\sigma}(\theta)$ reads

$$\mathbf{z}_{\sigma}(s,\theta) = \mathbf{x}_{\sigma}(\theta) + sh\boldsymbol{\Psi}_{\sigma}^{s}(\theta), \qquad (5.16)$$

where $s \in [1, 1/\lambda]$ and $\mathbf{z} = (x, y, p_x, p_y)$. The parameter h is small enough such that the invariance equation $|\mathcal{P}^{-1}(\mathbf{z}_{\sigma}(1,\theta)) - \mathbf{z}_{\sigma}(1/\lambda, \theta - \nu)| < \epsilon$ for all $\theta \in \mathbb{T}$, where ϵ is infinitesimal. The stable manifold associated with the invariant curve $\mathbf{x}_{\sigma}(\theta)$ intersects the plane $(x, p_y, y = p_x = 0)$ in one point for $s = s^*$ and $\theta = \theta^*$. This condition reads $\prod_{\mathbf{r}} (\mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s^*, \theta^*))) \cdot \mathbf{e}_y = 0$ and $\prod_{\mathbf{p}} (\mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s^*, \theta^*))) \cdot \mathbf{e}_x = 0$, where $\prod_{\mathbf{r}}$ and $\prod_{\mathbf{p}}$ are the projection from phase space onto positions and momentum component, respectively. The parameter m corresponds to the number of iterations required for the trajectory to reach the plane $y = p_x = 0$. In order to compute s^* and θ^* , we use Newton's method. However, this event is rare, and it is difficult to have a good initial guess. We use the following technique to compute s^* and θ^* :

1. We compute a finite set of invariant curves $\mathbf{x}_{\sigma}(\theta)$ associated with \mathcal{O}_S such that $\sigma = 1, \ldots, N$. Then, we compute the eigenvectors $\Psi_{\sigma}^s(\theta)$ associated with the real eigenvalue smaller than unity λ .

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- 2. We initiate a mesh of points $\mathbf{z}_{\sigma}(s_l, \theta_j)$ given by Eq. (5.16) for all σ , and for $l = 0, \ldots, N_s$ and $j = 0, \ldots, N_{\theta}$. We obtain an ensemble of initial conditions $\mathbf{z}_{\sigma}(s_l, \theta_j)$ labeled by σ , l and j. Notice that $N_s \sim 100$ and $N_{\theta} \sim 100$, and as a consequence, there are approximately 10^4 initial conditions for the invariant curve $\mathbf{x}_{\sigma}(\theta)$.
- 3. We compute $\mathcal{P}^m(\mathbf{z}_{\sigma}(s_l,\theta_j))$ for $m = 1, \ldots, N_m$ ($N_m = 100$ in our numerical computations). We store the points which are close to the plane $y = p_x = 0$ and their labels: m, s, θ and σ . Close to the plane $y = p_x = 0$ means that $|\Pi_{\mathbf{r}}(\mathcal{P}^m(\mathbf{z}_{\sigma}(s^*, \theta^*))) \cdot \mathbf{e}_y| < \epsilon$ and $|\Pi_{\mathbf{p}}(\mathcal{P}^m(\mathbf{z}_{\sigma}(s^*, \theta^*))) \cdot \mathbf{e}_x| < \epsilon$, where $\epsilon \sim 10^{-2}$ (the value of ϵ depends on the discretization of the parameters s and θ and on the eigenvalue λ).
- 4. The points which are stored are refined using Newton's method. We compute an approximation of s^* and θ^* for given m and σ such that

$$\Pi_{\mathbf{r}}(\boldsymbol{\mathcal{P}}^{-m}(\mathbf{z}_{\sigma}(s^*,\theta^*))) \cdot \mathbf{e}_y = 0, \qquad (5.17a)$$

$$\Pi_{\mathbf{p}}(\boldsymbol{\mathcal{P}}^{-m}(\mathbf{z}_{\sigma}(s^*,\theta^*))) \cdot \mathbf{e}_x = 0.$$
(5.17b)

The two parameters to adjust in order to fulfill this condition are s and θ . The initial guess of the Newton's method are the points which have been stored in the third step, i.e., s_l and θ_j . At each iteration of the Newton's method, the derivatives of Eqs. (5.17) with respect to s and θ must be computed (see Sec. D.2.1). They read

$$\frac{\partial}{\partial s} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s,\theta)) = h \mathcal{J}_{0}^{-mT}(\mathbf{z}_{\sigma}(s,\theta)) \Psi(\theta),$$

$$\frac{\partial}{\partial \theta} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s,\theta)) = \mathcal{J}_{0}^{-mT}(\mathbf{z}_{\sigma}(s,\theta)) \sum_{k=1}^{N} k \left[(\mathbf{b}_{k} + hs\mathbf{B}_{k}) \cos(k\theta) - (\mathbf{a}_{k} + hs\mathbf{A}_{k}) \sin(k\theta) \right].$$

where $\mathcal{J}_0^{-mT}(\mathbf{z}_{\sigma}(s,\theta))$ is the tangent flow with initial condition $\mathbf{z}_{\sigma}(s,\theta)$ integrated backward over mlaser cycles. The Fourier coefficients of the invariant curve and its eigenvector are denoted $\{\mathbf{a}_0, \dots, \mathbf{a}_N, \mathbf{b}_1, \dots, \mathbf{b}_N\}$ and $\{\mathbf{A}_0, \dots, \mathbf{A}_N, \mathbf{B}_1, \dots, \mathbf{B}_N\}$, respectively. The maximum harmonic of the Fourier representation is N. At the kth iteration of the Newton's method, the parameters are $[s_k; \theta_k]$ and we compute $[s_{k+1}; \theta_{k+1}] = [s_k; \theta_k] + [\Delta s; \Delta \theta]$, where $[\Delta s; \Delta \theta]$ is solution of

$$\begin{bmatrix} \Pi_{\mathbf{r}} \left(\frac{\partial}{\partial s} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s,\theta)) \right) \cdot \mathbf{e}_{y} & \Pi_{\mathbf{r}} \left(\frac{\partial}{\partial \theta} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s_{k},\theta_{k})) \right) \cdot \mathbf{e}_{y} \\ \Pi_{\mathbf{p}} \left(\frac{\partial}{\partial s} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s,\theta)) \right) \cdot \mathbf{e}_{x} & \Pi_{\mathbf{p}} \left(\frac{\partial}{\partial \theta} \mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s,\theta)) \right) \cdot \mathbf{e}_{x} \end{bmatrix}_{(s=s_{k},\theta=\theta_{k})} \begin{bmatrix} \Delta s \\ \Delta \theta \end{bmatrix} \\ = - \begin{bmatrix} \Pi_{\mathbf{r}} (\mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s_{k},\theta_{k}))) \cdot \mathbf{e}_{y} \\ \Pi_{\mathbf{p}} (\mathcal{P}^{-m}(\mathbf{z}_{\sigma}(s_{k},\theta_{k}))) \cdot \mathbf{e}_{x} \end{bmatrix}.$$

In this way, we obtain an approximation of s^* and θ^* . The images of the points initiated on the stable manifold, which belong to the stable manifold, also belong to the plane $y = p_x = 0$. The points such that $s > 1/\lambda$ are not kept. The points which are kept are the red markers in Fig. 5.5. Hence, we observe that the red markers are in good agreement with the location of the dark blue paths. The invariant manifolds \mathcal{W}_F^s drive the recollisions.

5. (This part is a work in progress) In order to show that \mathcal{W}_F^s partition the phase space, we need to compute the one-dimensional curves corresponding to the intersection between \mathcal{W}_F^s and the plane $y = p_x = 0$. We use a continuation method to track the initial conditions in the fundamental domain of the invariant curves given by Eq. (5.16) which lead to a point in the plane $y = p_x = 0$. For a given σ and for known parameters s^* and θ^* , we compute a new torus $\sigma + 1$. We control the distance between two adjacent invariant curves with a parameter δ . Using s^* and θ^* as initial guess, we use the Newton's method of the fourth point to compute s^* and θ^* associated with σ . If the Newton's method converge, we reiterate the same procedure for $\sigma + 2$. Otherwise, we decrease δ , and we recompute $\sigma + 1$. With the parameter δ , we can also control the distance between two successive points in the plane $y = p_x = 0$. In this way, we compute curves in the plane $y = p_x = 0$ which belong to the stable manifold \mathcal{W}_F^s .

The same scheme can be used to compute the unstable manifold \mathcal{W}_F^u by changing $\lambda \to 1/\lambda$, $\nu \to -\nu$, $\mathcal{P}^{-1} \to \mathcal{P}$ and $\mathcal{J}_0^{-T} \to \mathcal{J}_0^T$.

For d = 3, in cylindrical coordinates along \mathbf{e}_x , Hamiltonian (1.14) reads

$$H(x,\rho,\theta,p_x,p_\rho,p_\theta) = \frac{p_x^2}{2} + \frac{p_\rho^2}{2} + \frac{p_\theta^2}{2r^2} - \frac{Z}{\sqrt{x^2 + \rho^2 + a^2}} + xE_0\cos(\omega t).$$
(5.18)

We observe that Hamiltonian (5.18) does not depend on θ , and therefore p_{θ} is conserved. In general, p_{θ} is conserved if the ion-electron potential is rotationnally invariant, which is the case for atoms. Since p_{θ} is a conserved quantity, the phase space is foliated by surfaces of constant p_{θ} . In particular, $p_{\theta} = 0$ corresponds to the case d = 2 studied above. Along the transverse direction to the surface of constant p_{θ} , the eigenvalues of the linear stability of invariant objects are unitary due to the foliation of constant p_{θ} . Therefore, for d = 3, one component of the linear stability of the invariant objects is *parabolic*. In particular, for d = 3, \mathcal{O}_S is *center-saddle-parabolic* for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$. For d = 3, the stable manifold \mathcal{W}_F^s also partitions the phase space and drive the electrons.

Result 8: \mathcal{O}_S driving recollisions for LP

Using a dimension analysis and numerical evidence depicted in Figs. 5.2 and 5.5, we have shown that the invariant manifolds of the family of invariant tori associated with \mathcal{O}_S , denoted \mathcal{W}_F^s and \mathcal{W}_F^u and represented in Fig. 5.7, drive the electrons for d = 1, 2, 3. The codimension of this invariant structure with the phase space is one, and as a consequence, it can partition the phase space between recolliding and non-recolliding trajectories. A clear partition of the phase space is observed in Fig. 5.2 for d = 1. The electron cannot cross \mathcal{W}_F^s , and as a consequence, the invariant manifolds \mathcal{W}_F^s act as fences in the phase space of the electron. The electron is guided by \mathcal{W}_F^s .

Near the intersection of the stable \mathcal{W}_F^s and unstable \mathcal{W}_F^u manifolds, there are an infinity of periodic orbits with a large period. In particular, in Fig. 5.2, we observe that RPOs are in these regions. The invariant objects associated with the RPOs are two-dimensional, and as a consequence, they cannot partition the phase space and cannot drive the electrons for d > 1. However, the RPOs can be used to probe the highly dimensional invariant structures \mathcal{W}_F^s and \mathcal{W}_F^u , and as a consequence, to probe the location of recollisions in phase space.

5.3 From linearly to circularly polarized fields: Consequences of the symmetry breaking on the periodic orbits

In this section, we study the symmetries of the system for varying ellipticity ξ . We track the RPOs as a function of the intensity and the ellipticity of the laser field using continuation methods. Some RPOs are observed only in a specific range of ellipticities. We determine the mechanism behind the disappearance of these periodic orbits. In contrast, the RPO \mathcal{O}_F exists for all ellipticities. We study the connection between \mathcal{O}_F and the fixed points in the RF for CP fields ($\xi = 1$).

5.3.1 Symmetries in phase space

We denote $\mathcal{V}(r) = V(\mathbf{r})$, with $r = |\mathbf{r}|$, the rotationnally invariant potential of the ion-electron interaction. The equations of motion are

$$\dot{\mathbf{r}} = \mathbf{p}, \tag{5.19a}$$

$$\dot{\mathbf{p}} = -(\mathbf{r}/r)\mathcal{V}'(r) - \mathbf{E}(t), \qquad (5.19b)$$

with $\mathcal{V}'(r) = \partial \mathcal{V}(r)/\partial r$. Two discrete symmetries of the monochromatic laser field given by Eq. (5.1) are $\mathbf{E}(t+T) = \mathbf{E}(t)$ and $\mathbf{E}(t+T/2) = -\mathbf{E}(t)$ for all time t and all ellipticities. In the equations of motion, symmetries with forward propagation time are:

• For all ellipticities, the equations of motion (5.19) are invariant under the transformation

$$(\mathbf{r}, \mathbf{p}, t) \mapsto (-\mathbf{r}, -\mathbf{p}, T/2 + t).$$
 (5.20)

The orbits in phase space are symmetric with respect to the origin. For instance, the central symmetry observed in the PMDs for rather long laser pulses (see the left panel of Fig. 3.5 and Fig. 8) is a consequence of the invariance of the equations of motion and the distribution of the initial conditions (see Sec. 1.2.2) under the transformation given by Eq. (5.20).

• For all ellipticities, the equations of motion (5.19) are invariant under the transformation

$$(x, y, z, p_x, p_y, p_z, t) \mapsto (x, y, -z, p_x, p_y, -p_z, t).$$
 (5.21)

The orbits in phase space are symmetric with respect to the polarization plane $(\mathbf{e}_x, \mathbf{e}_y)$. For instance, the axial symmetry observed in the PMDs along the perpendicular momentum of the electron (see Fig. 3.4) is a consequence of the invariance of the equations of motion and the distribution of the initial conditions (see Sec. 1.2.2) under the transformation given by Eq. (5.21). Also, as a consequence, the subspace $z = p_z = 0$ is invariant and the analysis of the dynamics can be reduced from d = 3 to d = 2.

• If $\xi = 0$ (LP fields), the equations of motion (5.19) are invariant under the transformations

$$(\mathbf{r}, \mathbf{p}, t) \mapsto (\mathbf{R}_x(\theta)\mathbf{r}, \mathbf{R}_x(\theta)\mathbf{p}, t),$$
 (5.22)

for all θ and all time t, where $\mathbf{R}_x(\theta)$ is the rotation matrix along \mathbf{e}_x (see Appendix A). The orbits in phase space are symmetric with respect to the rotation around \mathbf{e}_x . Also, as a consequence, the subspace $y = p_y = z = p_z = 0$ is invariant and the analysis of the dynamics can be reduced from d = 3 to d = 1.

• If $\xi = 1$ (CP fields), the equations of motion (5.19) are invariant under the transformation

$$(\mathbf{r}, \mathbf{p}, t) \mapsto (\mathbf{R}_z(\omega\tau)\mathbf{r}, \mathbf{R}_z(\omega\tau)\mathbf{p}, t+\tau),$$
(5.23)

where $\mathbf{R}_z(\omega \tau)$ is the rotation matrix along \mathbf{e}_z (see Appendix A). Therefore, the orbits in phase space are symmetric with respect to the rotation around \mathbf{e}_z and the translation in time.

We observe that the axial symmetries with respect to the axis \mathbf{e}_x and the axis \mathbf{e}_z are exclusive to the LP and CP cases, respectively. In LP, due to this symmetry, each periodic orbit exists clockwise and anticlockwise. For instance, this is the case for the RPOs of the family \mathcal{O}_F , as observed in Fig. 5.5. When the ellipticity of the laser is changed, this symmetry is broken and the periodic orbits are no longer symmetric.

5.3.2 Evolution of recolliding periodic orbits (RPOs) with respect to the laser ellipticity

The left panels of Fig. 5.8 show the linear stability of the RPOs \mathcal{O}_F clockwise and anticlockwise, and \mathcal{O}_A as a function of the intensity I and the ellipticity ξ of the laser. The linear stability is identified from the two largest absolute eigenvalues of the monodromy matrix λ_1 and λ_2 such that $|\lambda_1| > |\lambda_2|$ (see Fig. C.2 for detail). If $|\lambda_1| = |\lambda_2| = 1$, the periodic orbit is *center-center*. If $|\lambda_1| > 1$, $|\lambda_2| > 1$ and $|\lambda_1| \neq |\lambda_2|$, it is *real* saddle. If $|\lambda_1| > 1$ and $|\lambda_2| = 1$, it is *center-saddle*. If $|\lambda_1| = |\lambda_2| > 1$, it is *degenerate saddle*. For each case, in the left panels of Fig. 5.8, the color scale is for $\log |\lambda_1|$. For $\xi = 0$, \mathcal{O}_F clockwise and \mathcal{O}_F anticlockwise exists regardless of the ellipticities and the intensities. The laser field given by Eq. (5.1) is also anticlockwise, and preserves the shape of the periodic orbit. In contrast, \mathcal{O}_F clockwise and \mathcal{O}_A exist only for a specific range of ellipticities and intensities. Similar scenarii are observed for \mathcal{O}_A^{\pm} and \mathcal{O}_F^{\pm} .

In order to determine the mechanism of the disappearance of the periodic orbit \mathcal{O}_A and \mathcal{O}_F clockwise, we fix the ellipticity at $\xi = 0.1$ (close to LP). The middle panel of Fig. 5.8 shows the RPOs of the family \mathcal{O}_A and \mathcal{O}_F for $I = 2 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$ in the plane (x, p_x) . We use a continuation method to track these periodic orbits as a function of the intensity. The continuation method is described in Appendix D.2.2.3. The right panel of Fig. 5.8 shows the component p_y of the RPOs \mathcal{O}_A , \mathcal{O}_A^{\pm} , \mathcal{O}_F and \mathcal{O}_F^{\pm} clockwise as a function of the laser intensity. The upper piece of the black curve is associated with \mathcal{O}_A , and the lower piece of the black curve is associated with \mathcal{O}_F clockwise. Similarly, the upper piece of the red (resp. blue) curve is associated with \mathcal{O}_A^+ (resp. \mathcal{O}_A^-), and the lower piece of the blue (resp. red) curve is associated \mathcal{O}_F^+ clockwise (resp. \mathcal{O}_F^-). Clearly, we observe that \mathcal{O}_A is connected to \mathcal{O}_F clockwise, \mathcal{O}_A^+ is connected to \mathcal{O}_F^+ clockwise, and \mathcal{O}_A^-

5.3. FROM LINEARLY TO CIRCULARLY POLARIZED FIELDS



Figure 5.8: Left panels: Stability map (maximum absolute eigenvalue in logarithmic scale) of the RPOs \mathcal{O}_A , \mathcal{O}_F clockwise and anticlockwise of Hamiltonian (5.14) (case d = 2) as a function of the intensity and the ellipticity, for a = 3 and Z = 1. Middle panels: Projection of the RPOs (upper panel) \mathcal{O}_A and \mathcal{O}_A^{\pm} , (lower panel) \mathcal{O}_F and \mathcal{O}_F^{\pm} clockwise, on the plane (x, p_x) for $I = 2 \times 10^{13}$ W \cdot cm⁻² and $\xi = 0.1$. Right panel: Component p_y of the RPOs depicted in the middle panel as a function of the laser intensity, tracked using the continuation method described in Appendix D.2.2.3. The lower piece of each curve is associated with the periodic orbits of the family \mathcal{O}_F clockwise and the upper piece of each curve is associated with the periodic orbits of the family \mathcal{O}_A . Therefore, the red curve is associated with the periodic orbits $\mathcal{O}_A^+/\mathcal{O}_F^+$ clockwise, the black curve is associated with the periodic orbits $\mathcal{O}_A/\mathcal{O}_F$ clockwise, and the blue curve is associated with the periodic orbits $\mathcal{O}_A^-/\mathcal{O}_F^-$ clockwise.

is connected to \mathcal{O}_F^- clockwise. For $\xi = 0$ (LP case), due to the symmetry of the orbits with respect to the polarization axis, both \mathcal{O}_F clockwise and \mathcal{O}_F anticlockwise join \mathcal{O}_A at high intensities (see the bifurcation occurring for $I \sim 10^{16} \text{ W} \cdot \text{cm}^{-2}$ in Fig. 5.6). In contrast, for $\xi \neq 0$, only \mathcal{O}_F clockwise joins \mathcal{O}_A , while \mathcal{O}_F anticlockwise persists regardless of the intensities.

In Fig. 5.9, we show the final distance of the electron as a function of the initial conditions $(x_0, p_{y,0}, y_0 = p_{x,0} = 0)$ at time t = 0, for an integration time 100T and for varying ellipticities. Notice that this is the same plane of initial conditions as in Fig. 5.5. The final distance of the electrons from the core highlights the nonlinearities due to the strong ion-electron and laser-electron interactions. In particular, the light colored regions and the paths which exhibit a sensitivity with respect to the initial conditions correspond to the initial conditions leading to recollisions. The white squares indicate the location of the fixed points of the RPOs under \mathcal{P} . The panel $\xi = 0$ is discussed in Sec. 5.2.2. We observe that the region of initial conditions leading to recollisions is followed by the fixed point of the RPO \mathcal{O}_F anticlockwise. In the SFA, the initial conditions of the periodic orbit centered around the origin are given by

$$\mathbf{r}_0 = \mathbf{e}_x \frac{E_0}{\omega^2 \sqrt{\xi^2 + 1}}, \quad \mathbf{p}_0 = \mathbf{e}_y \frac{\xi E_0}{\omega \sqrt{\xi^2 + 1}},$$
 (5.24)

and correspond approximately to the location of the fixed point of \mathcal{O}_F anticlockwise under \mathcal{P} . Hence, the region of initial conditions leading to recollisions is approximately around $(\mathbf{r}_0, \mathbf{p}_0)$. We observe the distance in phase space between the fixed points of the family \mathcal{O}_A and \mathcal{O}_F clockwise decreases for increasing ellipticities. These orbits join each other for $\xi \in [0.11, 0.22]$. In the same way, The orbits \mathcal{O}_A^{\pm} and \mathcal{O}_F^{\pm} join each other for $\xi \in [0.22, 0.33]$. For $\xi > 0.55$, only \mathcal{O}_F anticlockwise persists.

5.3.3 Connection between \mathcal{O}_F and a fixed point in the rotating frame (RF): Case of circularly polarized laser fields

We consider the CP case ($\xi = 1$), and the dynamics of the electron in the RF. The RF is the frame in which the laser field does not depend on time for CP and f = 1 (see Sec. 4.1.2.1). We use the canonical transformation $\tilde{\mathbf{r}} = \mathbf{R}_z(\omega t)\mathbf{r}$ and $\tilde{\mathbf{p}} = \mathbf{R}_z(\omega t)\mathbf{p}$, with \mathbf{R}_z the rotation matrix around \mathbf{e}_z . The Hamiltonian is given by Eq. (4.9). In the RF and for rotationnally invariant potential, as it is the case for atoms, the



Figure 5.9: Final distance of the electron in logarithmic scale of Hamiltonian (5.14) (case d = 2) as a function of the initial conditions $(x_0, 0, 0, p_{y,0})$ for $I = 3 \times 10^{14} \text{ W} \cdot \text{cm}^{-2}$, a = 1 and Z = 1. White squares indicate the location of the fixed point of the RPOs under the Poincaré map \mathcal{P} . The upper squares are the location of the fixed points of (from left to right) \mathcal{O}_F^- , \mathcal{O}_F and \mathcal{O}_F^+ anticlockwise under \mathcal{P} . The middle squares are the location of the fixed points of (from left to right) \mathcal{O}_A^- , \mathcal{O}_A and \mathcal{O}_A^+ anticlockwise under \mathcal{P} . The lower squares are the location of the fixed points of (from left to right) \mathcal{O}_A^- , \mathcal{O}_F and \mathcal{O}_F^+ clockwise under \mathcal{P} . The lower squares are the location of the fixed points of (from left to right) \mathcal{O}_F^- , \mathcal{O}_F and \mathcal{O}_F^+ clockwise under \mathcal{P} . The insets are zooms around the periodic orbits \mathcal{O}_A^+ (middle squares), \mathcal{O}_F^+ clockwise (lower squares) and anticlockwise (upper squares). Positions and momenta are scaled by E_0/ω^2 and E_0/ω , respectively.

Hamiltonian does no longer depend on time. The energy is conserved $\tilde{H}(\tilde{\mathbf{r}}, \tilde{\mathbf{p}}) = \mathcal{K}$ and known as the *Jacobi* constant [77]. We use the notation $V(\tilde{\mathbf{r}}) = \mathcal{V}(\tilde{r})$ with $\tilde{r} = |\tilde{\mathbf{r}}|$. In the RF, the equations of motion are given by

$$\dot{\tilde{\mathbf{r}}} = \tilde{\mathbf{p}} - \omega \tilde{\mathbf{e}}_z \times \tilde{\mathbf{r}},$$
(5.25a)

$$\dot{\tilde{\mathbf{p}}} = -(\tilde{\mathbf{r}}/\tilde{r})\mathcal{V}'(\tilde{r}) + \omega \tilde{\mathbf{p}} \times \tilde{\mathbf{e}}_z - \tilde{\mathbf{e}}_x E_0/\sqrt{2}.$$
(5.25b)

The fixed points $(\mathbf{r}^*, \mathbf{p}^*)$ are such that $\dot{\mathbf{r}} = \dot{\mathbf{p}} = \mathbf{0}$. Using Eq. (5.25a), $\mathbf{\tilde{p}}^* = \omega \mathbf{\tilde{e}}_z \times \mathbf{\tilde{r}}^*$. Substituting this solution in Eq. (5.25b), we obtain

$$-(\tilde{\mathbf{r}}^{\star}/\tilde{r}^{\star})\partial\mathcal{V}'(\tilde{r}^{\star}) + \omega^{2}\left[\tilde{\mathbf{r}}^{\star} - (\mathbf{r}\cdot\tilde{\mathbf{e}}_{z})\tilde{\mathbf{e}}_{z}\right] - \tilde{\mathbf{e}}_{x}E_{0}/\sqrt{2} = \mathbf{0}.$$
(5.26)

Projecting Eq. (5.26) along $\tilde{\mathbf{e}}_z$, we get $\tilde{\mathbf{r}}^* \cdot \tilde{\mathbf{e}}_z = 0$. Projecting Eq. (5.26) along $\tilde{\mathbf{e}}_y$, we get $\tilde{\mathbf{r}}^* \cdot \tilde{\mathbf{e}}_y = 0$ or $\mathcal{V}'(\tilde{r}) = \omega^2 \tilde{r}$. Projecting Eq. (5.26) along $\tilde{\mathbf{e}}_y$, we get $\tilde{\mathbf{r}}^* \cdot \tilde{\mathbf{e}}_x [\omega^2 - \mathcal{V}'(\tilde{r})/\tilde{r}] = E_0$. If $\mathcal{V}'(\tilde{r}) = \omega^2 \tilde{r}$, the laser amplitude is $E_0 = 0$. For $E_0 > 0$, the fixed points are given by $\tilde{\mathbf{r}}^* = \tilde{x}^* \mathbf{e}_x$, where \tilde{x}^* is solution of the equation

$$\tilde{x}^{\star} \left[\omega^2 - \frac{\mathcal{V}'(|\tilde{x}^{\star}|)}{|\tilde{x}^{\star}|} \right] = \frac{E_0}{\sqrt{2}}.$$
(5.27)

For soft Coulomb potential $\mathcal{V}(|\tilde{x}^*|) = -Z(|\tilde{x}^*|^2 + a^2)^{-1/2}$, there are three fixed points. In particular, one fixed point is located far from the core, where $|\mathcal{V}'(|\tilde{x}^*|)/\tilde{x}^*| \ll \omega^2$. By substituting this condition in Eq. (5.27), we obtain that the fixed point is located at $\tilde{x}^* \approx E_0/\sqrt{2}\omega^2$. It corresponds to the top of the zero-velocity surface depicted in Fig. 4.3. In the LF and under the Poincaré map \mathcal{P} , the coordinates of this fixed point are approximately $\mathbf{r}_0^* = \mathbf{e}_x E_0/\omega^2\sqrt{2}$ and $\mathbf{p}_0^* = \mathbf{e}_y E_0/\omega\sqrt{2}$, which corresponds to the initial conditions given by Eq. 5.24. These initial conditions are close to the fixed point of \mathcal{O}_F . Numerically, we have seen that the top of the zero-velocity surface corresponds to \mathcal{O}_F anticlockwise in the RF.

In Ref. [81], the scenario is that RPOs in the RF drive the recollisions (see Fig. 5.2.2). In contrast, in Refs. [14, 129], the recollisions are driven by the invariant manifolds of the saddle point of the zero-velocity surface (fixed point indicated by a blue dot in Fig. 5.2.2). This latter scenario is in accordance with our Result 5.2.2, which shows that in LP, the periodic orbit driving the electron dynamics is close to the ionic core. Here, we have shown that \mathcal{O}_F anticlockwise is a fixed point in the RF for the CP case. In addition, this periodic orbit is center-saddle for most of the intensities and ellipticities, as observed in the left panel of Fig. 5.8. Therefore, it is a good candidate for driving the recollisions and structuring the dynamics. In Fig. 5.9, we have shown that this periodic orbit is in the region of initial conditions leading to recollisions for all ellipticities. However, one piece of the periodic orbit \mathcal{O}_F anticlockwise is close to the core only for small ellipticities. For large ellipticities, the minimum distance of this periodic orbit from the core is at least $\xi E_0/\omega^2 \sqrt{\xi^2 + 1}$. Therefore, \mathcal{O}_F anticlockwise is not a RPO in the sense it is defined in Refs. [81, 80, 109, 3].

5.4 Conclusions

In sum, we have shown that the electron dynamics for atoms subjected to intense laser fields is driven by invariant structures in phase space. For LP fields, the relevant invariant structure is associated with the SPO \mathcal{O}_S . For d = 2, this invariant structure is four-dimensional, and correspond to a set of stable \mathcal{W}_F^s and unstable \mathcal{W}_F^u manifolds of the family of invariant curves associated with \mathcal{O}_S . \mathcal{W}_F^s and \mathcal{W}_F^u are shown in Fig. 5.7. In Sec. 5.2.2, we have described a procedure to obtain a planar representation of \mathcal{W}_F^s and \mathcal{W}_F^u . This planar representation allowed us to show that \mathcal{W}_F^s and \mathcal{W}_F^u structure the phase space of the electrons, and as a consequence, drive their motion.

In general, we have seen that the relevant and important invariant structures for driving the electrons are of codimension one with the manifolds on which the electrons evolve. For instance, for LP fields and d = 2, invariant structures of codimension two (or larger) with the phase space, such as the invariant structures associated with the RPOs \mathcal{O}_A and \mathcal{O}_F , do not play a role for driving the electrons. However, we have observed that the RPOs are located near the intersections of \mathcal{W}_F^s and \mathcal{W}_F^u . In Sec. 5.3, we have used these RPOs to probe the high-dimensional invariant structure driving the electrons for varying ellipticity and intensity. In particular, the RPO \mathcal{O}_F anticlockwise persists for all ellipticities. For CP fields, \mathcal{O}_F corresponds to the fixed point at the top of the zero-velocity surface in the RF.

Conclusions

Attosecond science makes use of the known dynamics of the ionized electrons, teared off their parent atom or molecule by an intense laser pulse, to image structural dynamics during ultrafast nonequilibrium processes, such as for instance, charge migrations and electronic dynamics during chemical reactions. In this thesis, we have shown that, despite their fundamental quantal nature in atoms and molecules, electrons display some classical behaviors when subjected to intense laser pulses. Remarkably, classical mechanics, with the help of trajectories and nonlinear dynamics, turned out to be very insightful to identify and interpret mechanisms behind the nonlinear phenomena observed in experiments. However, there have always been fundamental obstacles to accurately interpret the electron trajectories underlying the experimental measurements.

The first difficulty was to account for the quantal nature of the electrons in atoms and molecules. Indeed, in nature, electrons in atoms and molecules are accurately described by quantum mechanics and their wave representation. It is only after ionizing, for sufficiently large laser intensities, that the quantum dynamics of the electrons clearly exhibits classical features. In our framework, the state of the electrons before ionization was treated quantum mechanically. According to the parameters, the electrons were able to ionize through or over the potential barrier induced by the intense laser field. While the ionization through the potential barrier is a purely quantum mechanical process, over-the-barrier ionization was also accurately described by classical mechanics. After ionization, the motion of the electron was treated purely classically in terms of trajectories. In both cases, tunnel or over-the-barrier ionization, quantum characteristics of the electrons were contained in the distribution of the initial conditions of their trajectory.

The second difficulty was to understand and analyze the trajectories of the ionized electrons by taking into account the interaction with their parent ion, which is commonly ignored but can make its presence known even after ionization. We have shown that the interplay between the interactions of the ionized electrons with the laser and their parent ion gives rise to multiple temporal and spatial scales, yielding their dynamics highly nonlinear, and giving rise to rich and diverse ionization channels. By changing the ellipticity of the driving laser, which acts as a simple control knob in experiments, we changed the prioritized ionization channel taken by the electrons. In order to understand and analyze the variety of ionization channels the ionized electrons can take, we have used two main methods. The first method was nonperturbative and consisted in understanding the electron dynamics through the analysis of invariant structures in phase space. One advantage of this technique was to provide a framework to analyze the global behavior of the trajectories, rather than their individual behavior. The second method was perturbative and consisted in deriving reduced models, and using them interpret the electron trajectories. These methods allowed us to identify the impact of the ion-electron interaction in the nonlinear phenomena observed in experiments, and to include its role in the description of the underlying mechanisms.

Considering the quantum nature of the electrons before ionization and their classical motion after ionization, in the light of nonlinear dynamics, allowed us to unravel mechanisms behind nonlinear phenomena observed in experiments, that linear dynamics was unable to predict. For instance, we have unraveled the mechanism behind the bifurcation observed in the PMDs measured in experiments [93]. We have determined, and been able to describe accurately, how electrons can return to the core after multiple laser cycles and how they can be trapped in Rydberg states. We have determined the conditions under which experimental manifestations of recollisions in CP and near-CP fields can be observed. This thesis work demonstrates the complementarity of quantum mechanics and nonlinear dynamics for understanding and illustrating the mechanisms involved when atoms are subjected to intense and elliptically polarized laser pulses.

Summary

In Chap. 1, we have derived the quantum and classical models for electrons in atoms driven by intense and elliptically polarized laser pulses. We have shown that the quantum dynamics of the electron clearly exhibits
classical features after ionization. We took advantage of the classical features of the electron to describe and analyze its dynamics, after ionization, in terms of trajectories. In particular, we have studied qualitatively the most probable trajectory according to the tunneling ionization theory, referred to as the T-trajectory. The qualitative analysis of the T-trajectory allowed us to introduce the tools from nonlinear dynamics we have used throughout the manuscript.

In Chap. 2, we have derived reduced models for approximating the electron dynamics. On the one hand, there are the strong field approximation (SFA) [39] and the Coulomb-corrected strong field approximation (CCSFA) [62]. These models are particularly efficient for the description of short time scale processes, i.e., which involve significantly the Coulomb potential on a short time scale. For instance, these models can qualitatively reproduce recolliding trajectories for a few laser cycles after ionization, which are particularly important for understanding the characteristics of the HHG intensity spectra. On the other hand, we have derived a hierarchy of reduced models for the guiding-center (GC) motion of electrons in intense laser fields [49, 50]. The GC motion corresponds to the averaged trajectory of the ionized electrons. In Fig. 5.10, the dark red curve represents the GC trajectory of an electron (light red curve) recolliding after multiple laser cycles. The light red electron undergoes a *Coulomb-driven recollision*. When it returns to the core, its energy is exchanged with a bound electron (light blue curve) and both ionize. In order to return to the core after multiple laser cycles, the contribution of the Coulomb potential on the motion of the electron during its excursion far from the core is significant for long time scales. The GC models are able to describe and reproduce accurately the motion of the ionized electrons on these long time scales. The second order model of this hierarchy of models (see Sec. 2.3.2.1) provides an intuitive view of the motion of the electron in the combined laser and Coulomb field, where the motion of the electron has two components: a slow component corresponding to the motion of the GC and a fast component due to the interaction with the laser. After ionization, the electron oscillates around its guiding center at the frequency of the laser field. The energy of the GC is conserved, which allowed us to define an energy for the ionized electrons.



Figure 5.10: Typical non-sequential double ionization (NSDI) induced by a Coulomb-driven recollision in Mg atoms subjected to an intense laser field of ellipticity $\xi = 0.5$ in 3D. The light red and light blue curves are the recolliding and bound electrons, respectively. The dark red curve is the guiding-center (GC) trajectory of the recolliding electron at the fifth order model $G_5 = (H_5, \Phi_5)$. We observe that the motion of the GC is bounded, its energy is negative. The GC drives the red electron back to the core.

In Chap. 3, we have studied the impact of the Coulomb potential in ATI using the reference Hamiltonian and the reduced models derived in Chap. 2. In particular, we have analyzed the Coulomb effects in the PMDs measured experimentally, namely Coulomb focusing and Coulomb asymmetry. We have shown that Coulomb focusing is a consequence of the decrease of the energy of the electron by the Coulomb potential compared to what it would be at the detector if the ion-electron interaction was ignored. Coulomb asymmetry is a consequence of the deviation of the drift velocity of the electrons by the Coulomb potential compared to what it would be at the detector if the ion-electron interaction was ignored. We have shown that including the Coulomb potential gives rise to a variety of trajectories such as the subcycle recollisions, direct ionizations, Coulomb-driven recollisions and Rydberg state creations, while only the subcycle recollisions and the direct ionizations are well described by the SFA which ignores the ion-electron interaction. The GC energy of the electron allowed us to clearly distinguish and describe this variety of ionization channels. The ellipticity of the laser highlights the contribution of the laser field in the PMDs. In particular, we have unraveled the mechanism behind the bifurcation observed in the PMDs for increasing ellipticity: At the critical ellipticity, the Coulomb asymmetry appears at the same time as Coulomb focusing begins to recede. Our numerical prediction of the critical ellipticity is in agreement with experiments [93, 104]. We have shown that, for large ellipticities, the rates of Rydberg state creations and Coulomb-driven recollisions decreases drastically. For CP fields, if the electron ionizes when the laser field reaches its peak amplitude, the probability the electron recollides is almost zero. This chapter shows particularly well the complementarity of quantum mechanics and nonlinear dynamics in the description and interpretation of nonlinear phenomena in attosecond science.

In Chap. 4, we have investigated the impact of the Coulomb potential and the laser envelope in the existence of recollisions for CP and near-CP fields [61, 111]. We have identified a highly probable recollision channel with large return energy by accounting for the effects of the pulse envelope f(t) and the Coulomb potential. The recollisions taking this channel are referred to as envelope-driven recollisions. This recollision channel is particularly effective for nearly-CP fields, since the conventional recollision channel disappears. The competition between the Coulomb force and the laser field makes this recollision channel highly probable by creating a channel of ionization early after the laser field is turned on. Just as the electron is outside the core region, the amplitude of the vector potential is small, and therefore the sideways drift of the electron can be compensated by its momentum. We have shown that this recollision channel and the return of the electron can be understood using the SFA. Then, we have shown that this recollision channel can be used to produce HHG with atoms driven by highly elliptically polarized laser fields, and is also responsible for the enhanced double ionization from specific target species subjected to CP fields observed in experiments and numerical simulations [61, 111]. The existence condition of this recollision channel has been derived using the GC model, and agrees well with the conditions for which the enhanced double ionization for CP fields is observed experimentally [55, 65, 61]. In addition, we have noticed that, under reasonable conditions, recollisions can take place in the Attoclock setup, where recollisions are always assumed to be nonexistent [53, 152].

In Chap. 5, we have used nonperturbative methods to understand and analyze the global behavior of the electron trajectories. In particular, we have studied relevant and important invariant structures to assess the motion of the electrons. We have identified a high-dimensional invariant structure which partitions the phase space and drives the electrons. This four-dimensional invariant structure corresponds to a set of stable and unstable manifolds of a family of invariant tori associated with a *center-saddle* periodic orbit. We have described a procedure to represent this high-dimensional structure on a plane, and we have compared it with the global behavior of the electrons. This structure originates near the core due to the competition between the laser and Coulomb interactions, and as a consequence, demonstrates the importance of the interplay between these two interactions in the dynamics of the ionized electrons return to the core with a relatively small momentum. In this case, they can be trapped for several laser cycles near the core due to chaotic regions in phase space. The analysis of the invariant structures in phase space allowed us to understand the origin of these chaotic regions.

Perspectives

In this manuscript, we have shown that the combination of the quantum description of the electron before ionization and the purely classical treatment of its dynamics after ionization, in the light of nonlinear dynamics, provides an interesting and promising framework for studying the mechanisms involved in the nonlinear phenomena observed in experiments. In particular, using perturbative and nonperturbative techniques, we have shown that the Coulomb potential and the variations of the laser envelope, which are commonly ignored, can manifest in different ways in the experimental measurements.

- The hierarchy of models for the GC dynamics has been derived purely classically. An analogue quantum mechanical derivation, by using unitary transformations at appropriate orders in analogy of the canonical transformations, could lead to additional terms in the quantum Hamiltonians for the GC compared to the classical Hamiltonians for the GC. This could reveal features specific to the quantum characteristics of the electrons [128].
- The study of the role of the laser envelope in the GC dynamics has been very limited, and derived only for slowly varying laser envelopes (see Sec. 2.2.2.5). The effects of the laser envelope on the dynamics of the GC could highlight particular effects of the laser envelope on the outcome of the electrons. In addition, the study of the influence of the laser envelope on the invariant structures could also highlight its effects on the dynamics of the electrons.
- In LP, we have shown that invariant structures partitioning the phase space and driving the recollisions for d = 2 and d = 3 are associated with an SPO. In contrast, it was shown in Ref. [81] that RPOs drive the recollisions in CP. In Ref. [14], it was shown that SPOs drive the recollisions in CP. Which invariant structures drive the recollisions in phase space regardless the dimension and the ellipticity is still under debate.
- We have found that the Coulomb potential plays a significant role on the motion of the electrons after ionization, and can even make their dynamics chaotic. Certainly, the Coulomb potential affects the dynamics of the electrons during the ionization process, when the electron is very close to the core, in particular during quantum tunneling. A nonperturbative analysis of these processes could reveal interesting features on tunneling processes in atoms.
- The motion of an electron in an isolated laser field and its motion in an Hydrogen-like atom are two fundamental and very well-known systems in physics. However, this work shows that the motion of the electron becomes very complex and sophisticated when these two fundamental forces are combined. It makes wonder if nonlinear dynamics could push the limits of the knowledge and understanding of ultrafast nonequilibrium phenomena in attosecond science.

Appendix A

Glossary

A.1 Acronyms

- ADK: Ammosov-Delone-Krainov.
- ATI: above threshold ionization.
- CCSFA: Coulomb-corrected strong field approximation.
- CEP: carrier envelope phase.
- CTMC: Classical trajectory Monte Carlo.
- FWHM: full width at half maximum.
- GC: guiding center.
- HHG: high horder harmonic generation.
- KH: Kramers-Henneberger.
- LF: laboratory frame
- NSDI: nonsequential double ionization.
- NSMI: nonsequential multiple ionization.

A.2 Notations

- $[\mathbf{x}]_j = x_j$ The *j*th element of a vector \mathbf{x} .
- $[\mathbf{A}]_{jk} = A_{jk}$ The coefficient of the *j*th row and *k*th column of the matrix \mathbf{A} .
- $[\mathbf{AB}]_{jk} = A_{jl}B_{lk}$ Multiplication between two matrices.
- \mathbf{I}_N Identity matrix of size $N \times N$.
- $\mathbf{0}_N$ Zero matrix of size $N \times N$.
- e. Unitary vector in the LF.
- $\tilde{\mathbf{e}}$. Unitary vector in the RF.
- \cdot^{\top} Transpose matrix.

- PAD: photoelectron angular distribution.
- PMD: photoelectron momentum distribution.
- PPT: Perelomov-Popov-Terent'ev.
- RF: rotating frame.
- RPO: recolliding periodic orbit.
- SAE: single-active electron.
- SFA: strong field approximation.
- SPO: saddle periodic orbit.
- TDSE: Time-dependent Schrödinger equation.
- TISE: Time-independent Schrödinger equation.
- T-trajectory: Most probable trajectory according to the ADK or PPT ionization rate.
- XUV: Extreme ultraviolet.
- $\hat{\cdot}$ Quantum operator.
- $i = \sqrt{-1}$ Imaginary number.
- $\nabla = \partial/\partial \mathbf{r} = \mathbf{e}_x \partial/\partial x + \mathbf{e}_y \partial/\partial y + \mathbf{e}_z \partial/\partial z$ The gradient operator.
- $\partial \mathbf{y}(\mathbf{x}_0) / \partial \mathbf{x}$ Derivative of $\mathbf{y}(\mathbf{x})$ with respect to \mathbf{x} evaluated at $\mathbf{x} = \mathbf{x}_0$.
- $\Delta = \boldsymbol{\nabla} \cdot \boldsymbol{\nabla}$ Laplacian operator.
- · Scalar product between two vectors.
- $\langle \cdot \rangle$ Average or mean value.
- $\cdot \times \cdot$ For scalars: Multiplication. For vectors: Cross product.

- $|\cdot|$ Euclidean norm.
- .* Coordinates of a fixed point of a dynamical system or a mapping.
- ℜ· Real part.
- \Im · Imaginary part.
- R. Rotation matrix around the axis e.:

- $\Pi_{\mathbf{r}}$ · Projection into the position components in phase space.
- $\Pi_{\mathbf{p}}$ · Projection into the momentum components in phase space.
- Sp Spectrum of a matrix.
- det Determinant of a matrix.

$$\mathbf{R}_x(\theta) = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\theta & -\sin\theta \\ 0 & \sin\theta & \cos\theta \end{bmatrix}, \quad \mathbf{R}_y(\theta) = \begin{bmatrix} \cos\theta & 0 & \sin\theta \\ 0 & 1 & 0 \\ -\sin\theta & 0 & \cos\theta \end{bmatrix}, \quad \mathbf{R}_z(\theta) = \begin{bmatrix} \cos\theta & -\sin\theta & 0 \\ \sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

• $\mathcal{F}[f(\mathbf{x})](\mathbf{y})$ Fourier transform of $f(\mathbf{x})$ as a function of \mathbf{y} :

$$\mathcal{F}[f(\mathbf{x})](\mathbf{y}) = \frac{1}{(2\pi)^{d/2}} \int_{-\infty}^{\infty} f(\mathbf{x}) \exp(-i\mathbf{x} \cdot \mathbf{y}) \, \mathrm{d}^d \mathbf{x},$$

$$\mathcal{F}^{-1}[F(\mathbf{y})](\mathbf{x}) = \frac{1}{(2\pi)^{d/2}} \int_{-\infty}^{\infty} F(\mathbf{y}) \exp(i\mathbf{x} \cdot \mathbf{y}) \, \mathrm{d}^d \mathbf{y}.$$

A.2.1 Numerator layout convention

For vectors and matrices, we use the numerator layout convention, also known as the *Jacobian formulation*. Let **x** and **y** be vectors of size *n* and *m*, with elements such that $\mathbf{x} = [x_1, \dots, x_n]^\top$ and $\mathbf{y} = [y_1, \dots, y_m]^\top$, respectively.

• The derivatives $\partial \mathbf{y} / \partial \mathbf{x}$ layout according to \mathbf{y} and \mathbf{x}^{\top} , and as a consequence

$$\left[\frac{\partial \mathbf{y}}{\partial \mathbf{x}}\right]_{ij} = \frac{\partial y_i}{\partial x_j}.$$

The differentiation of a vector $\mathbf{y}(\mathbf{x})$ with respect to a scalar \mathbf{x} is $d\mathbf{h}(\mathbf{z}) = (\partial \mathbf{h}/\partial \mathbf{z}) d\mathbf{z}$ using the chain rule. The transpose of the derivatives is the derivative of the transposes $(\partial \mathbf{y}(\mathbf{x})/\partial \mathbf{x})^{\top} = \partial \mathbf{y}^{\top}/\partial \mathbf{x}^{\top}$.

• The gradient of a scalar $y(\mathbf{x})$ with respect to the variables \mathbf{x} is a row vector of size n (size $n \times 1$) such that

$$\frac{\partial y(\mathbf{x})}{\partial \mathbf{x}} = \begin{bmatrix} \frac{\partial y}{\partial x_1} & \cdots & \frac{\partial y}{\partial x_n} \end{bmatrix}.$$

The Hessian matrix of $y(\mathbf{x})$ with respect to \mathbf{x} is

A.2.2 Laser

 $H_{ij} = \partial^2 y / \partial x_i \partial x_j$, and in compact notation

$$\mathbf{H} = \frac{\partial^2 y(\mathbf{x})}{\partial \mathbf{x}^\top \partial \mathbf{x}} = \begin{bmatrix} \frac{\partial^2 y(\mathbf{x})}{\partial x_1 \partial x_1} & \cdots & \frac{\partial^2 y(\mathbf{x})}{\partial x_1 \partial x_n} \\ \vdots & \ddots & \vdots \\ \frac{\partial^2 y(\mathbf{x})}{\partial x_n \partial x_1} & \cdots & \frac{\partial^2 y(\mathbf{x})}{\partial x_n \partial x_n} \end{bmatrix}.$$

• The derivative of a vector $\mathbf{y}(x)$ with respect to a scalar x is a column vector

$$\frac{\partial \mathbf{y}(x)}{\partial x} = \begin{bmatrix} \frac{\partial y_1}{\partial x} \\ \vdots \\ \frac{\partial y_m}{\partial x} \end{bmatrix}.$$

- The identity is $\partial \mathbf{x} / \partial \mathbf{x} = \mathbf{I}_n$, with \mathbf{I}_n the *identity* matrix of size $n \times n$.
- If $\mathbf{u} = \mathbf{u}(\mathbf{x})$, then

$$\frac{\partial \mathbf{g}(\mathbf{u}(\mathbf{x}))}{\partial \mathbf{x}} = \frac{\partial \mathbf{g}(\mathbf{u})}{\partial \mathbf{u}} \frac{\partial \mathbf{u}(\mathbf{x})}{\mathbf{x}}$$

- *I* Intensity of the laser field.
- λ Wavelength of the laser field.
- ω Frequency of the laser field.
- T Period of the laser field.
- E_0 Amplitude of the laser field.
- f Envelope of the laser field.
- T_f Time when the laser field is turned off.
- ξ Ellipticity of the laser field.
- Ω Frequency of the radiated photon.
- **E**(*t*) Electric field as a function of time in the LF.

A.2.3 Atomic model

- Ψ Wavefunction in the position representation.
- W Wigner quasi-probability distribution.
- I_p Ionization potential of the electron in the atom.
- \mathcal{E}_q Ground state energy of the atom.
- *a* Softening parameter of the soft Coulomb potential.
- (**r**, **p**) Position and canonical conjugate momentum of the electron in the laboratory frame (LF).
- $(\mathbf{r}_0, \mathbf{p}_0)$ Initial position and momentum of the electron.
- ($\tilde{\mathbf{r}}, \tilde{\mathbf{p}}$) Position and canonical conjugate momentum of the electron in the rotating frame (RF).
- Φ_n The *n*th order transformation from the electron to the guiding-center coordinates.
- *H* Total Hamiltonian of the system.
- $H_{\rm free}$ Hamiltonian of the field-free atom.
- H_n Hamiltonian of the guiding center.
- V Ion-electron interaction potential.

A.2.4 Dynamical systems and Hamiltonian formalism

- *d* Dimension of the configuration space.
- $\dot{x} = dx/dt$ Derivative of x with respect to time.
- $\ddot{x} = d^2 x/dt^2$ Double derivative of x with respect to time.

- $\mathbf{A}(t)$ Vector potential as a function of time in the LF.
- $\tilde{\mathbf{E}}(t)$ Electric field as a function of time in the RF.
- $\tilde{\mathbf{A}}(t)$ Vector potential as a function of time in the RF.
- $\mathbf{e}_{\parallel}(t_0) = \mathbf{E}(t_0)/|\mathbf{E}(t_0)|$ Unitary vector along the laser field direction at ionization time t_0 .
- $\mathbf{e}_{\perp}(t_0) = -[\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_y]\mathbf{e}_x + [\mathbf{e}_{\parallel}(t_0) \cdot \mathbf{e}_x]\mathbf{e}_y$ Unitary vector transverse to the laser field direction at ionization time t_0 .
- V Rotationally invariant ion-electron interaction potential.
- $G_n = (H_m, \Phi_n)$ (with $m \le n$) The *n*th order guiding-center model.
- ($\bar{\mathbf{r}}, \bar{\mathbf{p}}$) Position and canonical conjugate momentum of the guiding center in the LF.
- (*˜***r**, *˜***p**) Position and canonical conjugate momentum of the guiding center in the RF.
- t_0 Ionization time of the electron.
- W_{ADK}, W_{PPT} Ionization rate in the ADK and PPT theory, respectively.
- $(\mathbf{r}_{ADK}, \mathbf{p}_{ADK})$, $(\mathbf{r}_{PPT}, \mathbf{p}_{PPT})$ Initial position and momentum of the electron after tunneling in the ADK and PPT theory, respectively.
- $(\mathbf{r}_{ADK}^{T}, \mathbf{p}_{ADK}^{T})$, $(\mathbf{r}_{PPT}^{T}, \mathbf{p}_{PPT}^{T})$ Initial position and momentum after tunneling of the Ttrajectory in the ADK and PPT theory, respectively.
- $\mathbf{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y + P_z \mathbf{e}_z$ Final momentum of the T-trajectory.
- R_{\min} , R_{\max} Distance threshold for detecting the recolliding trajectories.
- $\varphi_{t_1}^{t_2}$ Flow from time t_1 to time t_2 .
- $\mathcal{J}_{t_1}^{t_2}$ Tangent flow from time t_1 to t_2 .
- \mathcal{P} Poincaré map.
- $\{\cdot, \cdot\}$ Poisson bracket.

- ${\cal K}$ Jacobi constant in the RF.
- $\bullet~ \mathcal{O}_{\cdot}$ periodic orbits.
- \mathcal{W}_{F}^{\cdot} Invariant manifold of the family of invariant tori of the SPO \mathcal{O}_{S} .
- \mathcal{W}^{\cdot} Invariant manifold of a fixed point.
- \mathcal{F}^{\cdot} Fundamental domain of the invariant manifold of a fixed point.
- ν Frequency of the invariant curve.
- $\mathbf{x}(\theta)$ Representation of the invariant curve.

Appendix B

Atomic scales

The general results on the Bohr semi-classical approach of the Hydrogen atom [26] are presented. These results are used to define the atomic units and the system of conversion used in this manuscript, and the atomic scales involved in these systems.

B.1 Hydrogen atom: The Bohr semi-classical approach

We consider a proton of mass m_p and an electron of mass m_e . The position and the momentum of the proton is denoted $(\mathbf{r}_p, \mathbf{p}_p)$ and that of the electron is denoted $(\mathbf{r}_e, \mathbf{p}_e)$. In ISU, the Hamiltonian of the system reads

$$H(\mathbf{r}_e, \mathbf{r}_p, \mathbf{p}_e, \mathbf{p}_p) = \frac{|\mathbf{p}_e|^2}{2m_e} + \frac{|\mathbf{p}_p|^2}{2m_p} - \frac{\mathrm{e}^2}{4\pi\varepsilon_0|\mathbf{r}_e - \mathbf{r}_p|}$$

with ε_0 the permittivity of the vacuum and e the charge of one electron. We perform the canonical change of coordinates $\mathbf{r} = \mathbf{r}_e - \mathbf{r}_p$, $\mathbf{p} = \mu(\mathbf{p}_e/m_e - \mathbf{p}_p/m_p)$, the coordinates of the *center of mass* $\mathbf{R} = \mu(\mathbf{r}_e/m_p + \mathbf{r}_p/m_e)$, $\mathbf{P} = \mathbf{p}_e + \mathbf{p}_p$, where the reduced mass is $\mu = m_e m_p/(m_e + m_p)$. Therefore, the Hamiltonian in the new coordinates is

$$H(\mathbf{r}, \mathbf{R}, \mathbf{p}, \mathbf{P}) = \frac{|\mathbf{P}|^2}{2(m_e + m_p)} + \frac{|\mathbf{p}|^2}{2\mu} - \frac{e^2}{4\pi\varepsilon_0|\mathbf{r}|}$$

The motion of the center of mass is uniform. We choose the center of mass to be static at the origin, i.e., $\mathbf{R} = \mathbf{P} = \mathbf{0}$. Then, we use the polar-nodal coordinates (see Sec. 2.3.2.1). In polar-nodal coordinates and for static center of mass, the latter Hamiltonian becomes

$$H(r,\theta,\nu,p_r,p_\theta,p_\nu) = \frac{p_r^2}{2\mu} + \frac{p_\theta^2}{2\mu r^2} - \frac{e^2}{4\pi\varepsilon_0 r}.$$

In these variables, the angle ν and its conjugate momentum p_{ν} are conserved, which is related to the degeneracy of the magnetic number m. The angular momentum $p_{\theta} = \ell$ is also conserved. The reduced dynamics of the electron is therefore

$$H(r, p_r) = \frac{p_r^2}{2\mu} + \frac{\ell}{2\,\mu\,r^2} - \frac{e^2}{4\pi\varepsilon_0 r}.$$
(B.1)

Therefore, the proton and the electron are balanced between attraction, due to the proton-electron interaction, and repulsion, due to the inertial force $\ell^2/2\mu r$. Bohr's assumption for the quantification of the angular momentum is

$$\ell = n\hbar,\tag{B.2}$$

where $n \in \mathbb{N}^*$ corresponds to the principal quantum number and \hbar is the *reduced Plank constant* or *infinitesimal action*. In 1924, in his thesis on the theory of quanta [45], de Broglie provides an interpretation of the Bohr's assumption based on the wave-particle duality: "it can be seen as a resonance condition of the electronic wave written in the system related to the nucleus of the atom", where the wavelength of the wave function of the electron h/p_r is a multiple integer of the circumference $|\mathbf{r}|$ of the circular orbit described by the electron. The energy of the atom is $\mathcal{E} = H(r, p_r)$, and substituting Eq. (B.2) in Eq. (B.1), one obtains

$$\mathcal{E} = \frac{p_r^2}{2\mu} + \frac{n^2\hbar^2}{2\mu r^2} - \frac{e^2}{4\pi\varepsilon_0 r}.$$

For a given principal quantum number n, the minimum energy of the electron is reached for a radius r_n which minimizes the energy, for which the orbit is circular, i.e., $p_r = 0$. The energy is given by

$$\mathcal{E}_n = -\frac{1}{2n^2} \frac{m_e}{\hbar^2} \left(\frac{e^2}{4\pi\varepsilon_0}\right)^2,\tag{B.3}$$

where $\mu \approx m_e$. The energy \mathcal{E}_1 is the ground state energy of the Hydrogen atom, and therefore n = 1 labels the ground state of the Hydrogen atom. The Bohr radius a_0 is defined as the smallest radius of the ground state of the Hydrogen atom r_1 , hence,

$$a_0 \equiv \frac{\hbar^2 4\pi \,\varepsilon_0}{m_e \,e^2}.\tag{B.4}$$

B.2 Atomic units

The Bohr radius a_0 in Eq. (B.4) and the first ionization potential of the Hydrogen atom \mathcal{E}_1 in Eq. (B.3) correspond to the characteristic length and the characteristic energy of the electron in atom. First, the dimensions are written in terms of the physical quantities of the Hydrogen atom

$$\mathbf{L} = \begin{bmatrix} \frac{\hbar^2 \, 4\pi \, \varepsilon_0}{m_e \, e^2} \end{bmatrix}, \quad \mathbf{M} = [m_e], \quad \mathbf{T} = \begin{bmatrix} \frac{\hbar^3}{m_e} \, \left(\frac{4\pi \, \varepsilon_0}{e^2}\right)^2 \end{bmatrix}.$$

Second, we use the *Gaussian units*, where $4\pi \varepsilon_0 = 1$. Then, we define a non-dimensional system of units by setting

$$\hbar = m_e = -e = 4\pi \,\varepsilon_0 = 1.$$

By performing this transformation, the Bohr radius becomes unity. Therefore, 1 a.u. corresponds to the characteristic length of the electron in the atom. In order to determine $A = A(\hbar, m_e, e)$ in atomic units from the international system of units (I.S.U.), the following relation

$$A(1,1,-1)$$
 (a.u.) = $A(1.055 \times 10^{-34}, 9.109 \times 10^{-31}, -1.602 \times 10^{-19})$ (I.S.U.).

B.2.1 Laser parameters

We consider a laser electric field \mathbf{E} and magnetic field \mathbf{B} . The *intensity* of the laser field I is defined as the time-averaged magnitude of the *Poynting vector*, i.e.,

$$I = \frac{1}{T} \int_0^T \frac{\mathbf{E} \times \mathbf{B}}{\mu_0} \,\mathrm{d}t,\tag{B.5}$$

where μ_0 is the vacuum permeability and T is the laser period. We consider an elliptically polarized laser field of the form $\mathbf{E}(\mathbf{r},t) = E_0/\sqrt{\xi^2 + 1}[\mathbf{e}_x \cos(\omega t - kz) + \mathbf{e}_y \xi \sin(\omega t - kz)]$, where k is the absolute value of the vector propagation of the electromagnetic wave. Substituting the expression of the laser field and the associated magnetic field in Eq. (B.5), we obtain the relation between the intensity of the laser and its amplitude

$$I (\mathbf{W} \cdot \mathbf{m}^{-2}) = \frac{\varepsilon_0 c}{2} \left(E_0 \left[\mathbf{V} \cdot \mathbf{m}^{-1} \right] \right)^2,$$

where $\varepsilon_0 \approx 8.85 \times 10^{-12} \text{ F} \cdot \text{m}^{-1}$ is the permittivity of vacuum. The scaling $1/\sqrt{\xi^2 + 1}$ in the laser field [see for instance Eq. (1.10)] is used to conserve the intensity of the laser with respect to the polarization of the field. In the literature, the properties of the laser can be given by its energy $\mathcal{E}_{\text{laser}} = P_{\text{max}}T_{\text{FWHM}}$, where $P_{\text{max}} = \pi R^2 I/2$ is the peak amplitude of the power of the laser beam, R is the radius of the laser beam and T_{FWHM} is the time of the full width at half maximum of the laser beam. The wavelength λ is related to the frequency of the laser with

$$\lambda \,[\mathrm{m}] = \frac{2\pi c}{\omega \,[\mathrm{rad} \cdot \mathrm{s}^{-1}]}.$$

Physical quantities and conversions	I.S.U.	a.u.
Electron charge e	$-1.602 \times 10^{-19} \text{ C}$	-1
Reduced Planck constant \hbar	$6.625 \times 10^{-34} \text{ J.s}$	1
Electron mass m_e	$9.109 \times 10^{-31} \text{ kg}$	1
Bohr radius a_0	$5.292 \times 10^{-11} \text{ m}$	1
Electric potential in H at the Bohr radius	27.219 V	1
Ground state energy of H	-13.6 eV	-1/2
Proton mass m_p	$1.673 \times 10^{-27} \text{ kg}$	1836.15
Neutron mass m_n	$1.673 \times 10^{-27} \text{ kg}$	1836.15
Orbit period at the Bohr radius T_0	$15.198 \times 10^{-17} \text{ s}$	2π
Electron velocity at the Bohr radius	$2.187 \times 10^{6} \text{ m.s}^{-1}$	1
Celerity	$3.000 \times 10^8 \text{ m.s}^{-1}$	137.037
Temperature $k_B T$	$T\times 1.3806\times 10^{-23}~{\rm J}$	$T \times 3.16 \times 10^{-6}$
Laser intensity I	$3.510 \times 10^{16} \mathrm{W} \cdot \mathrm{cm}^{-2}$	1
Laser wavelength λ	0.05292 nm	1
Laser frequency ω	$4.134 \times 10^{16} \text{ rad} \cdot \text{s}^{-1}$	1
Distance r	$5.292 \times 10^{-11} \text{ m}$	1
Velocity v	$2.187 \times 10^{6} \text{ m} \cdot \text{s}^{-1}$	1
Time t	$2.419 \times 10^{-17} \text{ s}$	1

Table B.1: Value of different physical quantities in the International System of Units (I.S.U) and in the atomic units (a.u.) The quantity r denotes the distance between the proton and the electron.

Appendix C

Dynamical systems

C.1 Continuous flow

A *dynamical system* is associated with an evolution in time. We consider a dynamical system described by the first order ordinary differential equation

$$\dot{\mathbf{z}} = \mathbf{f}(\mathbf{z}, t),\tag{C.1}$$

where $\dot{\mathbf{z}} = d\mathbf{z}/dt$, $\mathbf{z} \in \mathbb{R}^n$ is the set of dynamical variables with $n \in \mathbb{N}^*$, and t is the evolution parameter. For a given initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$, the solution is unique, and as a consequence the system is deterministic. The function

$$\boldsymbol{\varphi}_{t_0}^t: \boldsymbol{\zeta} \mapsto \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{z}(t), \tag{C.2}$$

is the flow function of the vector field **f** such that $\varphi_{t_0}^{t_0}(\boldsymbol{\zeta}) = \boldsymbol{\zeta}$ (i.e., φ_t^t is the identity function), and $\varphi_{t_1}^t(\varphi_{t_0}^{t_1}(\boldsymbol{\zeta})) = \varphi_{t_0}^t(\boldsymbol{\zeta})$ (i.e., $\varphi_{t_1}^t \circ \varphi_{t_0}^{t_1}$ is the group law). The flow of the vector field **f** maps an initial condition $(\boldsymbol{\zeta}, t_0)$ to the corresponding solution of Eq. (C.1). The derivatives of the flow function with respect to the initial conditions $\boldsymbol{\zeta}$, denoted $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \partial \varphi_{t_0}^t(\boldsymbol{\zeta})/\partial \boldsymbol{\zeta}$ and called the *tangent flow*, is solution of the equation

$$\dot{\boldsymbol{\mathcal{J}}}_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{A} \left(\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}), t \right) \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}), \tag{C.3}$$

with initial condition $\mathcal{J}_{t_0}^{t_0}(\boldsymbol{\zeta}) = \mathbf{I}_n$. The matrix $\mathbf{A}(\mathbf{z}(t), t) = \partial \mathbf{f}(\mathbf{z}, t) / \partial \mathbf{z}|_{\mathbf{z}=\mathbf{z}(t)}$ is the Jacobian matrix of the mapping \mathbf{f} or the matrix of variations. Using the group law of the flow and the chain rule, the tangent flow matrix is such that $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \mathcal{J}_{t_1}^t(\varphi_{t_0}^{t_1}(\boldsymbol{\zeta})) \mathcal{J}_{t_0}^{t_1}(\boldsymbol{\zeta})$.

C.1.1 Invariant objects

The linear stability assesses the stability of a trajectory under a small perturbation with respect to the initial conditions $\delta \boldsymbol{\zeta}$. Under a small perturbation, the flow becomes $\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta} + \delta \boldsymbol{\zeta}) = \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}) + \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta})\delta \boldsymbol{\zeta} + O(|\delta \boldsymbol{\zeta}|^2)$. The normal behavior of a trajectory $\mathbf{z}(t)$ corresponds to the evolution of the perturbation $\delta \mathbf{z}(t) = \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta} + \delta \boldsymbol{\zeta}) - \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})$ at the first order in $\delta \mathbf{z}(t_0) = \delta \boldsymbol{\zeta}$. The normal behavior of $\mathbf{z}(t)$ is

$$\delta \mathbf{z}(t) = \mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) \delta \mathbf{z}(t_0). \tag{C.4}$$

We observe that the perturbation evolves with respect to time as $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})\delta\mathbf{z}(t_0)$. If $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})$ is diagonalizable, i.e., if there exists **S** an invertible matrix such that $\mathcal{D}_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{S}^{-1}\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})\mathbf{S}$ is diagonal, we use the change of coordinates $\delta\mathbf{y}(t) = \mathbf{S}^{-1}\delta\mathbf{z}(t)$ for all time t. The small perturbation expressed in the basis of the eigenvectors of the tangent flows $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})$ [see Eq. (C.4)] becomes

$$\delta \mathbf{y}(t) = \mathcal{D}_{t_0}^t(\boldsymbol{\zeta}) \delta \mathbf{y}(t_0), \tag{C.5}$$

Therefore, the spectrum of the tangent flow and their associated eigenvectors provide information on the local stability of a trajectory. We denote $\{\lambda_i(\boldsymbol{\zeta})\}_{i=0}^n = \operatorname{Sp}(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}))$ the eigenvalues of the tangent flow matrix $\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta})$. If the real part of λ_i , $\Re(\lambda_i) < 1$, the small perturbation shrinks to zero along the associated eigenvector. In contrast, if $\Re(\lambda_i) > 1$, the small perturbation enlarges along the associated eigenvector.

C.1.1.1 Lyapunov characteristic exponents

According to the Lyapunov theorem, if the matrix $\mathbf{A}(\mathbf{z},t)$ is bounded, then for each nontrivial solution of Eq. (C.1) $\varphi_{t_0}^t(\boldsymbol{\zeta})$, the *finite time Lyapunov exponent* is the real number defined by

$$\Lambda(t) = \max_{i} \frac{\log |\lambda_i(t)|}{t},$$
(C.6)

with $\{\lambda_i\}_{i=0}^n = \operatorname{Sp}(\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}))$ are the eigenvalues of the tangent flow matrix $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})$. The Lyapunov exponent is given by the limit when time goes to infinity $\Lambda = \lim_{t\to\infty} \Lambda(t)$. The Lyapunov exponents estimates the strength of the sensitivity of a trajectory with respect to the initial conditions. Along one trajectory, the stability can change, going from stable to unstable.

C.1.1.2 Fixed point

We consider that $\boldsymbol{\zeta}$ is a fixed point of the flow if and only if $\mathbf{f}(\boldsymbol{\zeta},t) = \boldsymbol{\zeta}$ for all t, or equivalently

$$\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}) = \boldsymbol{\zeta},\tag{C.7}$$

for all time t. As a consequence, the Jacobian matrix of the fixed point $\boldsymbol{\zeta}$ is constant and $\mathbf{A}(\boldsymbol{\zeta},t) = \mathbf{A}(\boldsymbol{\zeta})$. The solution of the tangent flow [see Eq. (C.3)] is

$$\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \exp\left[\mathbf{A}(\boldsymbol{\zeta})(t-t_0)\right]. \tag{C.8}$$



Figure C.1: (a) Poincaré diagram of a two-dimensional linear system $\dot{\mathbf{z}} = \mathbf{A}\mathbf{z}$ with $\mathbf{z} = [x, y]^{\top}$ in the space (trA, detA) of solution $\mathbf{z}(t) = \exp[\mathbf{A}(t-t_0)]\mathbf{z}(t_0)$. The fixed point of the mapping is $\boldsymbol{\zeta} = \mathbf{0}$. It is depicted by a black dot in (b-m). If det $\mathbf{A} = 0$, the fixed point is a line passing by $\boldsymbol{\zeta} = 0$, see (g) and (l). The constant matrix \mathbf{A} is the Jacobian of the mapping with eigenvalues $\lambda_{\pm} = \operatorname{tr} \mathbf{A}/2 \pm [(\operatorname{tr} \mathbf{A})^2 - 4 \det \mathbf{A}]^{1/2}/2$. The system is Hamiltonian if $\operatorname{tr} \mathbf{A} = 0$ (implying $|\det \mathcal{J}_{t_0}^t(\boldsymbol{\zeta})| = 1$, see Sec. C.2). The fixed point is: (b) center (here $\operatorname{tr} \mathbf{A} = 0$ and det $\mathbf{A} = 1$), (c) saddle (here $\operatorname{tr} \mathbf{A} = 0$ and det $\mathbf{A} = -1$), (d) spiral sink, (e) degenerate sink, (f) sink, (g) line (thick black line) of stable fixed points, (h) saddle with $|\lambda_{-}| > |\lambda_{+}|$, (i) spiral source, (j) degenerate source, (k) source, (l) line (thick black line) of unstable fixed point is parabolic (see legend). (b-m) The thin lines with arrows are the trajectories around in (x, y), the blue and red curves are the stable manifold \mathcal{W}^s and the unstable manifold \mathcal{W}^u if any, respectively.

Let $\mathcal{D}_{\mathbf{A}}(\zeta) = \mathbf{S}^{-1}\mathbf{A}(\zeta)\mathbf{S}$ be the diagonal matrix of $\mathbf{A}(\zeta)$, then $\mathcal{D}_{t_0}^t(\zeta) = \mathbf{S}^{-1}\mathcal{J}_{t_0}^t(\zeta)\mathbf{S}$ since $\mathbf{A}^k(\zeta) = \mathbf{S}\mathcal{D}_{\mathbf{A}}^k(\zeta)\mathbf{S}^{-1}$ and $\mathcal{D}_{\mathbf{A}}^k(\zeta)$ is a diagonal matrix for all $k \in \mathbb{N}$, where we recall that $\mathcal{D}_{t_0}^t(\zeta)$ is the diagonal matrix similar to the tangent flow. Therefore, on the one hand, the spectrum of the tangent flow $\{\lambda_k(\zeta)\}_{k=0}^n = \operatorname{Sp}(\mathcal{J}_{t_0}^t(\zeta))$ is related to the spectrum of the Jacobian matrix $\{\mu_k(\zeta)\}_{k=0}^n = \operatorname{Sp}(\mathbf{A}(\zeta))$ through

$$\lambda_k(\boldsymbol{\zeta}) = \exp[\mu_k(\boldsymbol{\zeta})(t-t_0)].$$

In particular, $\lambda_k(\boldsymbol{\zeta}) = \exp[\Re(\mu_k(\boldsymbol{\zeta}))(t-t_0)] \exp[i\Im(\mu_k(\boldsymbol{\zeta}))(t-t_0)]$. As a consequence, the real part of μ_k increases or decreases the perturbation with respect to time, while the imaginary part is the frequency of the rotation of the perturbation [see Eq. (C.5) and Fig. C.1 for an example with a two-dimensional map linearized in the neighborhood of the fixed point]. On the other hand, since det $\mathcal{D}_{t_0}^t(\boldsymbol{\zeta}) = \det \mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \prod_{k=1}^n \lambda_k(\boldsymbol{\zeta})$ and $\operatorname{tr} \mathcal{D}_{\mathbf{A}}(\boldsymbol{\zeta}) = \operatorname{tr} \mathbf{A}(\boldsymbol{\zeta}) = \sum_{k=1}^n \mu_k(\boldsymbol{\zeta})$, one has

$$\det \mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \exp\left[(t - t_0) \operatorname{tr} \mathbf{A}(\boldsymbol{\zeta})\right].$$
(C.9)

C.1.1.3 Periodic trajectory

The trajectory $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$ is a *periodic orbit* with period T if and only if

$$\boldsymbol{\varphi}_t^{t+T}(\mathbf{z}(t)) = \mathbf{z}(t), \tag{C.10}$$

for all time t. The stability of the periodic orbit is given by $\operatorname{Sp}(\mathbf{M}_T(\boldsymbol{\zeta}))$, where $\mathbf{M}_T(\boldsymbol{\zeta}) = \mathcal{J}_{t_0}^{t_0+T}(\boldsymbol{\zeta})$ is the monodromy matrix. In addition, if the mapping is periodic $\mathbf{f}(\mathbf{z}, t+T) = \mathbf{f}(\mathbf{z}, t)$ for all time t, then by uniqueness of the solution of Eq. (C.1), the trajectory $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$ is a periodic orbit with period T if and only if

$$\varphi_{t_0}^{t_0+T}(\boldsymbol{\zeta}) = \boldsymbol{\zeta}. \tag{C.11}$$

C.1.2 *T*-periodic change of coordinates

We consider a change of coordinates $\tilde{\mathbf{z}} = \mathbf{h}(\mathbf{z},t)$, *T*-periodic $\mathbf{h}(\mathbf{z},t+T) = \mathbf{h}(\mathbf{z},t)$, of class \mathcal{C}^1 and bijective, such that $\tilde{\mathbf{z}} = \mathbf{h}(\mathbf{h}^{-1}(\tilde{\mathbf{z}},t),t)$ for all $\tilde{\mathbf{z}}$. We notice that the inverse function \mathbf{h}^{-1} is also *T*-periodic because of the bijectivity. We denote $\tilde{\varphi}_{t_0}^t(\tilde{\mathbf{z}})$ and $\varphi_{t_0}^t(\mathbf{z})$ the flow functions in the new and old set of coordinates, respectively. The flow function in the new variables is also *T*-periodic such that $\tilde{\varphi}_{t_0+mT}^{t_0+mT}(\tilde{\mathbf{z}}) = \tilde{\varphi}_{t_0}^{t_0+T}(\tilde{\mathbf{z}})$ for all $m \in \mathbb{N}^*$, while the flow function in the old set of coordinates is invariant under time translation such that $\varphi_{t_0}^t(\mathbf{z}) = \varphi_0^{t-t_0}(\mathbf{z})$ for all *t*. Typically, the old system of coordinates corresponds to the position and momentum of the electron in the rotating frame (resp. the guiding-center model), while the new system of coordinates corresponds to the order for which the guiding-center model is time-independent). We consider the trajectories $\tilde{\mathbf{z}}(t) = \tilde{\varphi}_{t_0}^t(\tilde{\boldsymbol{\zeta}})$ with initial condition $\tilde{\mathbf{z}}(t_0) = \boldsymbol{\zeta}$, and $\mathbf{z}(t) = \varphi_{t_0}^t(\boldsymbol{\zeta})$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$, such that $\tilde{\boldsymbol{\zeta}} = \mathbf{h}(\boldsymbol{\zeta}, t_0)$. For all time *t*, the flows are related through $\varphi_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{h}^{-1}(\tilde{\varphi}_{t_0}^t(\mathbf{h}(\boldsymbol{\zeta}, t_0)), t)$, and respectively, $\tilde{\varphi}_{t_0}^t(\tilde{\boldsymbol{\zeta}}) = \mathbf{h}(\varphi_{t_0}^t(\mathbf{h}^{-1}(\tilde{\boldsymbol{\zeta}}, t_0)), t)$. Using the periodicity of the change of coordinates $\mathbf{h}(\mathbf{z}, t+mT) = \mathbf{h}(\mathbf{z}, t)$ and $\mathbf{h}^{-1}(\mathbf{z}, t+mT) = \mathbf{h}^{-1}(\mathbf{z}, t)$ with $m \in \mathbb{N}^*$, one gets

$$\tilde{\boldsymbol{\varphi}}_{t_0}^{t_0+mT}(\tilde{\boldsymbol{\zeta}}) = \mathbf{h}\left(\boldsymbol{\varphi}_{t_0}^{t_0+mT}\left(\mathbf{h}^{-1}(\tilde{\boldsymbol{\zeta}},t_0)\right), t_0\right), \tag{C.12}$$

and respectively, $\varphi_{t_0}^{t_0+mT}(\boldsymbol{\zeta}) = \mathbf{h}^{-1}(\tilde{\varphi}_{t_0}^{t_0+mT}(\mathbf{h}(\boldsymbol{\zeta},t_0)),t_0)$. Therefore, $\tilde{\varphi}_{t_0}^{t_0+mT} = \mathbf{h} \circ \varphi_{t_0}^{t_0+mT} \circ \mathbf{h}^{-1}$, i.e., $\tilde{\varphi}_{t_0}^{t_0+mT}$ and $\varphi_{t_0}^{t_0+mT}$ are topologically conjugate [148, 1]. We denote $\tilde{\mathcal{J}}_{t_0}^t(\tilde{\boldsymbol{\zeta}}) = \partial \tilde{\varphi}_{t_0}^t(\tilde{\boldsymbol{\zeta}})/\partial \tilde{\boldsymbol{\zeta}}$ and $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \partial \varphi_{t_0}^t(\boldsymbol{\zeta})/\partial \boldsymbol{\zeta}$ the tangent flow functions in the new and the old set of coordinates, respectively. We differentiate the leftand right-hand side of Eq. (C.12) with respect to $\boldsymbol{\zeta}$. Using the chain rule, one gets

$$\tilde{\boldsymbol{\mathcal{J}}}_{t_0}^{t_0+mT}(\mathbf{h}(\boldsymbol{\zeta},t_0)) = \left. \frac{\partial \mathbf{h}(\mathbf{z},t_0)}{\partial \mathbf{z}} \right|_{\mathbf{z}=\boldsymbol{\varphi}_{t_0}^{t_0+mT}(\boldsymbol{\zeta})} \boldsymbol{\mathcal{J}}_{t_0}^{t_0+mT}(\boldsymbol{\zeta}) \left(\left. \frac{\partial \mathbf{h}(\mathbf{z},t_0)}{\partial \mathbf{z}} \right|_{\mathbf{z}=\boldsymbol{\zeta}} \right)^{-1}, \quad (C.13)$$

where we have used $\partial \mathbf{h}^{-1}(\tilde{\mathbf{z}})/\partial \tilde{\mathbf{z}} = (\partial \mathbf{h}(\mathbf{z})/\partial \mathbf{z}|_{\tilde{\mathbf{z}}=\mathbf{h}^{-1}(\tilde{\mathbf{z}})})^{-1}$ which comes from $\tilde{\mathbf{z}} = \mathbf{h}(\mathbf{h}^{-1}(\tilde{\mathbf{z}}))$, and where we have substituted $\tilde{\boldsymbol{\zeta}} = \mathbf{h}(\boldsymbol{\zeta}, t_0)$.

Remark 1 (Invariance of the structural stability with a regular change of coordinates) We consider a T-periodic change of coordinates \mathbf{h} of class \mathcal{C}^1 and bijective, such that $\mathbf{h} : \mathbf{z} \mapsto \tilde{\mathbf{z}} = \mathbf{h}(\mathbf{z})$ with \mathbf{z} and $\tilde{\mathbf{z}}$ the old and the new set of coordinates, respectively. The flow in the old set of coordinates is $\varphi_{t_0}^t(\mathbf{z})$ and such that $\varphi_{t_0}^t(\mathbf{z}) = \varphi_0^{t-t_0}(\mathbf{z})$. The flow in the new set of coordinates is $\tilde{\varphi}_{t_0}^t(\tilde{\mathbf{z}})$ and T-periodic such that $\tilde{\varphi}_{t_0}^{t_0+T}(\tilde{\mathbf{z}}) = \tilde{\varphi}_{t_0+T}^{t+2T}(\tilde{\mathbf{z}})$.

- If {z(t)}_t with initial condition z(t₀) = ζ is a fixed point, i.e., φ^t_{t₀}(ζ) = ζ for all t [see Eq. (C.7)], then, using Eq. (C.12) with m = 1, it implies that φ^{t₀+T}_{t₀}(ζ) = ζ, i.e., ζ is a periodic orbit of period T [see Eq. (C.11)]. Therefore, the fixed points in the old set of coordinates ž are transformed in periodic orbits of period T in the new set of coordinates ž. Moreover, using Eq. (C.13), the tangent flows are related through the J^{t₀+T}_{t₀}(**k**) = (∂**h**/∂**z**) ∘ J^{t₀+T}_{t₀}(ζ) ∘ (∂**h**/∂**z**)⁻¹. Therefore, they have the same spectrum.
- If $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$ is a periodic orbit of period $T_{\mathcal{O}}$, i.e., $\varphi_{t_0}^{t_0+T_{\mathcal{O}}}(\boldsymbol{\zeta}) = \boldsymbol{\zeta}$ [see Eq. (C.11)]:
 - If there exists $m, q \in \mathbb{N}^*$ such that $mT = nT_{\mathcal{O}}$, i.e., if $T/T_{\mathcal{O}} \in \mathbb{Q}$, it implies that $\tilde{\varphi}_{t_0}^{t_0+mT}(\tilde{\zeta}) = \tilde{\zeta}$, i.e., $\tilde{\zeta}$ is a periodic orbit of period $mT = nT_{\mathcal{O}}$ [see Eq. (C.11)]. Moreover, using Eq. (C.13), the tangent flow are related through the $\tilde{\mathcal{J}}_{t_0}^{t_0+mT}(\mathbf{h}(\zeta)) = (\partial \mathbf{h}/\partial \mathbf{z}) \circ \mathcal{J}_{t_0}^{t_0+mT}(\zeta) \circ (\partial \mathbf{h}/\partial \mathbf{z})^1$. Therefore, they have the same spectrum.
 - If $T/T_{\mathcal{O}} \in \mathbb{R} \setminus \mathbb{Q}$, the periodic orbit in the old set of coordinates $\tilde{\mathbf{z}}$ is transformed in an invariant curve in the new set of coordinates \mathbf{z} .
- A periodic orbit of period T in the new set of coordinates z becomes either a fixed point, or a periodic orbit of period T, in the old set of coordinates \tilde{z} .

C.2 Hamiltonian systems

A Hamiltonian system is a dynamical system whose dynamics is given by a scalar function $H(\mathbf{z})$, with $\mathbf{z} \in \mathbb{R}^n$ the set of *phase-space variables*, the Hamiltonian, and a Poisson bracket $\{\cdot, \cdot\}$ such that the evolution of an observable $F(\mathbf{z})$ is determined by

$$\frac{\mathrm{d}F(\mathbf{z})}{\mathrm{d}t} = \{F(\mathbf{z}), H(\mathbf{z})\},\tag{C.14}$$

where t is the evolution parameter. Here, all the properties hold for a time dependent or time independent Hamiltonian system, since in any case, the phase-space can be extended and time can be considered as a dynamical variable. The Poisson bracket is a bilinear operator, satisfying for any observable F, G and K:

- The anti-symmetry property $\{F, G\} = -\{G, F\}$,
- The Leibniz rule $\{F, GK\} = \{F, G\}K + G\{F, K\}$,
- The Jacobi identity $\{\{F, G\}, K\} + \{\{K, F\}, G\} + \{\{G, K\}, F\} = 0$.

Due to the anti-symmetry of the Poisson bracket $dH(\mathbf{z})/dt = \{H(\mathbf{z}), H(\mathbf{z})\} = 0$. Hence, the value of the Hamiltonian is a constant of motion. The Hamiltonian system, with the Poisson bracket, defines a Poisson algebra. In a set of dynamical variables \mathbf{z} , the Poisson bracket is defined as

$$\{F(\mathbf{z}), G(\mathbf{z})\} = \frac{\partial F(\mathbf{z})}{\partial \mathbf{z}} \mathbf{J}(\mathbf{z}) \frac{\partial G(\mathbf{z})}{\partial \mathbf{z}^{\top}}, \qquad (C.15)$$

where $\mathbf{J}(\mathbf{z})$ is the *Poisson matrix*. The elements of the Poisson matrix are given by $J_{ij}(\mathbf{z}) = \{z_i, z_j\}$. According to the properties of the Poisson bracket, namely the anti-symmetry and the Jacobi identity, the Poisson matrix is anti-symmetric $\mathbf{J}(\mathbf{z}) + \mathbf{J}^{\top}(\mathbf{z}) = \mathbf{0}$ and satisfies the *Jacobiator*

$$S_{ijk}(\mathbf{z}) = J_{il}(\mathbf{z})\frac{\partial J_{jk}(\mathbf{z})}{\partial z_l} + J_{kl}(\mathbf{z})\frac{\partial J_{ij}(\mathbf{z})}{\partial z_l} + J_{jl}(\mathbf{z})\frac{\partial J_{ki}(\mathbf{z})}{\partial z_l} = 0, \quad \text{or} \quad J_{il}(\mathbf{z})\frac{\partial J_{jk}(\mathbf{z})}{\partial z_l} + \bigcirc_{ijk} = 0, \quad (C.16)$$

for all *i*, *j* and *k*. Hence, if the Poisson matrix is independent of the dynamical variables \mathbf{z} , the Jacobi identity is satisfied. We assume that the Poisson matrix is invertible, which is possible only if det $\mathbf{J}(\mathbf{z}) = (-1)^n \det \mathbf{J}(\mathbf{z}) \neq 0$, and therefore only if n = 2N, where N is the number of degrees of freedom. The

inverse of the Poisson matrix is the Lagrange matrix $\boldsymbol{\omega}(\mathbf{z}) = \mathbf{J}^{-1}(\mathbf{z})$, and its derivative with respect to the dynamical variables \mathbf{z} is $\partial \omega_{ij}(\mathbf{z})/\partial z_k = \omega_{im}(\mathbf{z})(\partial J_{ml}(\mathbf{z})/\partial z_k)\omega_{lj}(\mathbf{z})$ and conversely $\partial J_{ij}(\mathbf{z})/\partial z_k = J_{il}(\mathbf{z})(\partial \omega_{lm}(\mathbf{z})/\partial z_k)J_{mj}(\mathbf{z})$ using $\omega_{ik}(\mathbf{z})J_{kj}(\mathbf{z}) = \delta_{ij}$ (with δ_{ij} the Kronecker symbol). Substituting the derivatives of the Poisson matrix in terms of the derivatives of the derivatives of the Lagrange matrix in Eq. (C.16), one gets the Jacobi identity in terms of the Lagrange matrix

$$\frac{\partial \omega_{ij}(\mathbf{z})}{\partial z_k} + \frac{\partial \omega_{ki}(\mathbf{z})}{\partial z_j} + \frac{\partial \omega_{jk}(\mathbf{z})}{\partial z_i} = 0, \quad \text{or} \quad \frac{\partial \omega_{ij}(\mathbf{z})}{\partial z_k} + \circlearrowright_{ijk} = 0, \quad (C.17)$$

for all i, j and k. The evolution of the set of phase-space variables \mathbf{z} is given by the Hamilton's equations. The Hamilton's equations are obtained after substituting $F(\mathbf{z}) = \mathbf{z}$ in Eq. (C.14), and read

$$\dot{\mathbf{z}} = \mathbf{J}(\mathbf{z}) \frac{\partial H(\mathbf{z})}{\partial \mathbf{z}^{\top}},$$
 (C.18)

with initial conditions $\mathbf{z}(t_0) = \boldsymbol{\zeta}$. We notice that Hamilton's equations have the same form of Eq. (C.1) with $\mathbf{f}(\mathbf{z},t) = \mathbf{J}(\mathbf{z})\partial H(\mathbf{z})/\partial \mathbf{z}^{\top}$. Hence, the Hamiltonian flow, or phase flow, is $\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{z}(t)$ solution of Eq. (C.18).

C.2.1 Change of coordinates

We consider the change of coordinates $\tilde{\mathbf{z}} = \mathbf{h}(\mathbf{z})$. According to the *scalar invariance*, any observable in the old and new system of coordinates remains the same $\tilde{F}(\tilde{\mathbf{z}}) = F(\mathbf{z})$, i.e., the value of the observable does not depends on the set of coordinates. In particular, the Hamiltonian is mapped into $\tilde{H}(\tilde{\mathbf{z}}) = H(\mathbf{z})$. In addition, the Poisson bracket which is an algebra structure is also invariant under change of coordinates, and therefore

$$\{F(\mathbf{z}), G(\mathbf{z})\} = \{\tilde{F}(\tilde{\mathbf{z}}), \tilde{G}(\tilde{\mathbf{z}})\}^{\sim}, \tag{C.19}$$

where $\{F, G\}$ is the Poisson bracket in the old set of coordinates, and $\{\tilde{F}(\tilde{\mathbf{z}}), \tilde{G}(\tilde{\mathbf{z}})\}^{\sim}$ the Poisson bracket in the new set of coordinates, such that

$$\{\tilde{F}, \tilde{G}\}^{\sim} = \frac{\partial F}{\partial \tilde{\mathbf{z}}} \tilde{\mathbf{J}}(\tilde{\mathbf{z}}) \frac{\partial G}{\partial \tilde{\mathbf{z}}^{\top}},\tag{C.20}$$

with $\hat{\mathbf{J}}(\tilde{\mathbf{z}})$ the Poisson matrix in the new system of coordinates. Due to the scalar invariance $F(\mathbf{z}) = \tilde{F}(\mathbf{h}(\mathbf{z}))$, and using the chain rule $\partial F(\mathbf{z})/\partial \mathbf{z} = (\partial \tilde{F}(\tilde{\mathbf{z}})/\partial \tilde{\mathbf{z}}|_{\tilde{\mathbf{z}}=\mathbf{h}(\mathbf{z})})(\partial \mathbf{h}(\mathbf{z})/\partial \mathbf{z})$. Substituting this expression in Eq. (C.19), the relation between the new and the old Poisson matrix is $\tilde{J}_{ij}(\tilde{\mathbf{z}}) = \{\tilde{z}_i, \tilde{z}_j\}$. In compact notation, it reads

$$\tilde{\mathbf{J}}(\mathbf{h}(\mathbf{z})) = \left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right) \mathbf{J}(\mathbf{z}) \left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right)^{\top}.$$
(C.21)

Substituting Eq. (C.21) in Eq. (C.16) and using a permutation an indexes permutation, one can show that the Jacobi identity is conserved under a change of coordinates. In terms of the Lagrange matrix $\boldsymbol{\omega}(\mathbf{z}) = \mathbf{J}^{-1}(\mathbf{z})$ and $\tilde{\boldsymbol{\omega}}(\mathbf{\tilde{z}}) = \mathbf{\tilde{J}}^{-1}(\mathbf{\tilde{z}})$, Eq. (C.21) becomes

$$\left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right)^{\top} \tilde{\boldsymbol{\omega}}(\mathbf{h}(\mathbf{z})) \left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right) = \boldsymbol{\omega}(\mathbf{z}).$$
(C.22)

Substituting Eq. (C.22) in Eq. (C.17) and using an indexes permutation, one can show that the Jacobi identity in terms of the Lagrange matrix is also conserved under a change of coordinates.

C.2.1.1 Canonical transformation

If the expression of the Poisson bracket is unchanged, i.e., if the expression of the coefficients of the Poisson matrix is unchanged in Eq. (C.20), which implies $\tilde{\mathbf{J}}(\tilde{\mathbf{z}}) = \mathbf{J}(\tilde{\mathbf{z}})$, the transformation is a *canonical transformation*. Otherwise, the transformation is a *noncanonical transformation*. If the expression of the Poisson matrix is unchanged and the Poisson matrix is invertible, then the expression of the Lagrange matrix is also unchanged $\tilde{\boldsymbol{\omega}}(\tilde{\mathbf{z}}) = \boldsymbol{\omega}(\tilde{\mathbf{z}})$. Equation (C.22) reads

$$\left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right)^{\top} \boldsymbol{\omega} \left(\mathbf{h}(\mathbf{z})\right) \left(\frac{\partial \mathbf{h}(\mathbf{z})}{\partial \mathbf{z}}\right) = \boldsymbol{\omega}(\mathbf{z}).$$
(C.23)

Therefore $\mathbf{h}(\mathbf{z})$ is a canonical transformation if and only if $\partial \mathbf{h}(\mathbf{z})/\partial \mathbf{z}$ is *symplectic*.

Remark 2 (Properties of the Hamiltonian flow) We consider a trajectory $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$ and the change of coordinates $\mathbf{h}(\boldsymbol{\zeta}) = \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})$, where $\mathbf{z}(t) = \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})$ is the Hamiltonian flow. Clearly, the Hamiltonian flow let the expression of the Poisson matrix and the Lagrange matrix unchanged, i.e., the phase flow is a canonical transformation. Using Eq. (C.22), the Hamiltonian flow and the tangent flow $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \partial \boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})/\partial \boldsymbol{\zeta}$ are such that

$$\left(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta})\right)^\top \boldsymbol{\omega} \left(\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})\right) \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) = \boldsymbol{\omega}(\boldsymbol{\zeta}),\tag{C.24}$$

for all time t. Therefore, the tangent flow is symplectic and the Hamiltonian flow is said to be a one-parameter group of ω -symplectic transformation.

Remark 3 (Spectrum of fixed points and periodic orbits in Hamiltonian systems) We consider a trajectory $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$, Hamiltonian flow $\varphi_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{z}(t)$ and tangent flow $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}) = \partial \varphi_{t_0}^t(\boldsymbol{\zeta})/\partial \boldsymbol{\zeta}$. The invertible Poisson matrix is $\mathbf{J}(\mathbf{z})$ and the Lagrange matrix is $\boldsymbol{\omega}(\mathbf{z}) = \mathbf{J}^{-1}(\mathbf{z})$ for all \mathbf{z} . Using Eq. (C.24):

• If $\boldsymbol{\zeta}$ is a fixed point, then $\varphi_{t_0}^t(\boldsymbol{\zeta}) = \boldsymbol{\zeta}$ for all time t [see Eq. (C.7)], and Eq. (C.24) is $(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}))^\top \boldsymbol{\omega}(\boldsymbol{\zeta}) \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) = \boldsymbol{\omega}(\boldsymbol{\zeta})$. Therefore, the tangent flow of a fixed point is such that $|\det \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta})| = 1$, and using Eq. (C.9), the Jacobian matrix is $\operatorname{tr} \mathbf{A}(\boldsymbol{\zeta}) = 0$, as depicted in Fig. C.1 for a one degree of freedom system. In addition, it implies that

$$\det \left(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) - \lambda_k \mathbf{I}_n \right) = \lambda_k^{2N} \det \left(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) \right)^{-1} \det \left(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) - \lambda_k^{-1} \mathbf{I}_n \right),$$

and its characteristic polynomial is reflexive. As a consequence, since $\mathcal{J}_{t_0}^t$ is real, if λ_k is an eigenvalue of $\mathcal{J}_{t_0}^t(\boldsymbol{\zeta})$, then λ_k^* , $1/\lambda_k$ and $1/\lambda_k^*$ are also eigenvalues.

• If $\boldsymbol{\zeta}$ is a periodic orbit of period T, then $\boldsymbol{\varphi}_{t_0}^{t_0+T}(\boldsymbol{\zeta}) = \boldsymbol{\zeta}$ [see Eq. (C.11)]. Using Eq. (C.24), the monodromy matrix $\mathbf{M}_T(\boldsymbol{\zeta}) = \mathcal{J}_{t_0}^{t_0+T}(\boldsymbol{\zeta})$ is such that $(\mathbf{M}_T(\boldsymbol{\zeta}))^\top \boldsymbol{\omega}(\boldsymbol{\zeta}) \mathbf{M}_T(\boldsymbol{\zeta}) = \boldsymbol{\omega}(\boldsymbol{\zeta})$. Hence, $|\det \mathbf{M}_T(\boldsymbol{\zeta})| = 1$ and the characteristic polynomial of the monodromy matrix is

$$\det \left(\mathbf{M}_T(\boldsymbol{\zeta}) - \lambda_k \mathbf{I}_{2N} \right) = \lambda_k^{2N} \det \left(\mathbf{M}_T(\boldsymbol{\zeta}) \right)^{-1} \det \left(\mathbf{M}_T(\boldsymbol{\zeta}) - \lambda_k^{-1} \mathbf{I}_{2N} \right).$$

Therefore, since \mathbf{M}_T is real, if λ_k is an eigenvalue of $\mathbf{M}_T(\boldsymbol{\zeta})$, then λ_k^* , $1/\lambda_k$ and $1/\lambda_k^*$ are also eigenvalues.

Therefore, the spectrum of the tangent flow of a fixed point, or the spectrum of the monodromy matrix of a periodic orbit, in Hamiltonian systems, is composed of eigenvalues coupled with each other with λ , $1/\lambda$, λ^* and $1/\lambda^*$ (see Fig. C.2). Therefore, the stable (resp. unstable) manifold of a fixed point always comes with its unstable (resp. stable) manifold. In addition, the dynamics contracts along the unstable manifold while it enlarges along the stable manifold, such that the volume in phase space is preserved.

C.2.2 Canonical Poisson bracket

The canonical coordinates are a class of phase-space variables in which the phase-space variables are coupled by pairs. The number of phase space variables is n = 2N with: N positions r_i and N canonically conjugate momentum p_i , for i = 1, ..., N. The phase space variables are such that $\{r_i, p_j\} = \delta_{ij}, \{r_i, r_j\} = 0$ and $\{p_i, p_j\} = 0, r_i$ and p_i are said to be canonically conjugate variables. We denote $\mathbf{z} = [r_1, ..., r_N, p_1, ..., p_N]^{\top}$. The Poisson bracket is a canonical Poisson bracket if and only if the Poisson matrix is the symplectic form

$$\mathbf{J}_{c} = \begin{bmatrix} \mathbf{0} & \mathbf{I}_{N} \\ -\mathbf{I}_{N} & \mathbf{0} \end{bmatrix}.$$
(C.25)

Some properties of the symplectic Poisson matrix are $\mathbf{J}_c^{-1} = \mathbf{J}_c^{\top} = -\mathbf{J}_c$, which implies in particular that $\mathbf{J}_c^2 = -\mathbf{I}_{2N}$ and $\mathbf{J}_c = -\boldsymbol{\omega}_c$.

C.2.2.1 Properties of the tangent flow

We consider a trajectory $\{\mathbf{z}(t)\}_t$ with initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$. The phase flow is $\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta}) = \mathbf{z}(t)$ and the tangent flow is $\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta})$. Substituting $\boldsymbol{\omega}(\boldsymbol{\varphi}_{t_0}^t(\boldsymbol{\zeta})) = \boldsymbol{\omega}(\boldsymbol{\zeta}) = \boldsymbol{\omega}_c$ in Eq. (C.24), for all t_0 , t and $\boldsymbol{\zeta}$, one has $(\boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}))^\top \boldsymbol{\omega}_c \boldsymbol{\mathcal{J}}_{t_0}^t(\boldsymbol{\zeta}) = \boldsymbol{\omega}_c$, and therefore

$$\mathcal{J}^{\top}\boldsymbol{\omega}_{c}\mathcal{J}=\boldsymbol{\omega}_{c}.$$
 (C.26)

Therefore, the tangent flow \mathcal{J} is symplectic. Considering the infinitesimal transformation of the phase flow $\varphi_{t_0}^{t_0+\delta t}(\mathbf{z})$ with $\delta t \ll 1$, the tangent flow matrix writes $\mathcal{J}_{t_0}^{t_0+\delta t}(\mathbf{z}) = \mathbf{I}_{2N} + \delta t \mathbf{A}(\mathbf{z}, t_0) + O(\delta t^2)$. In the first order in δt , Eq. (C.26) becomes $\boldsymbol{\omega}_c \mathbf{A} + \mathbf{A}^\top \boldsymbol{\omega}_c = \mathbf{0}$. Therefore, \mathbf{A} is a symplectic transformation, or an infinitesimal generator. In addition, applying the determinant on the left- and right-hand side of the previous equation, it immediately follows that

$$|\det \mathcal{J}| = 1$$

for all time t_0 and t and for all \mathbf{z} . Therefore, the Hamiltonian flow is phase space volume preserving, which is an elementary property for canonical Hamiltonian systems. Among other things, there are no dissipation in Hamiltonian systems, and in particular no attractors. The dynamics stretch out along some direction, and compress along other directions such that the phase-space volume is preserved. This has obviously consequences on the linear stability of a trajectory. We denote $\{\lambda_k(\boldsymbol{\zeta},t)\}_{k=1}^{2N} = \operatorname{Sp}(\mathcal{J}_{t_0}^t(\boldsymbol{\zeta}))$ the eigenvalues of the tangent flow at time t associated with the initial condition $\mathbf{z}(t_0) = \boldsymbol{\zeta}$. Since \mathcal{J} is symplectic, it implies that $\mathcal{J}^{-1} = -\boldsymbol{\omega}_c \mathcal{J}^\top \boldsymbol{\omega}_c$ so the characteristic polynomial is reflexive, i.e.,

$$\det\left(\boldsymbol{\mathcal{J}}-\lambda_{k}\mathbf{I}_{2N}\right)=\lambda_{k}^{2N}\det\left(\boldsymbol{\mathcal{J}}_{t_{0}}^{t}(\boldsymbol{\zeta})\right)^{-1}\det\left(\boldsymbol{\mathcal{J}}-\lambda_{k}^{-1}\mathbf{I}_{2N}\right),\tag{C.27}$$

for all k = 1, ..., 2N, for all t and for all ζ . Therefore, if λ_k is eigenvalue of \mathcal{J} , then $1/\lambda_k$, λ_k^* , $1/\lambda_k^*$ are also eigenvalues. Real eigenvalues always come paired as λ_k , $1/\lambda_k$, which is a direct consequence of the phase space volume preservation: The dynamics stretch along some directions and extends along others such that the volume preserves. The possibilities of the stability are illustrated in Fig. C.2.



Figure C.2: Eigenvalues of the monodromy matrix of periodic orbits (or eigenvalues of the tangent flow of a fixed point) of a two degrees of freedom Hamiltonian system (N = 2). Let $\rho, \mu, \theta, \phi \in \mathbb{R}^4$. The eigenvalues are: (a) Complex saddle { $\rho \exp(i\theta), \rho^{-1} \exp(i\theta), \rho \exp(-i\theta), \rho^{-1} \exp(-i\theta)$ }, (b) real saddle { $\rho, \rho^{-1}, \mu, \mu^{-1}$ }, (c) degenerate saddle: { $\rho, \rho, \rho^{-1}, \rho^{-1}$ }, (d) saddle-center { $\rho, \rho^{-1}, \exp(i\theta), \exp(-i\theta)$ }, (e) generic center { $\exp(i\theta), \exp(-i\theta), \exp(-i\theta)$ }, exp($-i\theta$), exp($-i\theta$), exp($-i\theta$)}, and (f) degenerate center { $\exp(i\theta), \exp(i\theta), \exp(-i\theta), \exp(-i\theta)$ }.

Appendix D Computational methods

In this appendix, we show the numerical methods and we detail the algorithms we have used throughout the manuscript for the numerical calculations. In Sec. D.1, we detail the methods used to generate the initial conditions of the electron. In particular, we show the algorithms for computing classical trajectory Monte Carlo (CTMC) simulations and the microcanonical generation of the initial conditions. In Sec. D.2, we detail the methods to compute invariant objects of a dynamical system. In particular, the methods to compute fixed points, periodic orbits, invariant tori, their linear stability and their stable and unstable manifolds.

D.1 Statistical analysis

In this section, we show the computational methods we use to generate the initial distribution of the electrons in phase space. In this manuscript, two types of methods are used to generate the initial conditions of the electrons for the statistical analysis: The CTMC simulations and the microcanonical generation of the initial conditions.

D.1.1 Classical trajectory Monte Carlo (CTMCs) simulations

The classical trajectory Monte Carlo simulations (CTMCs) consists in computing the distribution of an observable, such as for instance the distribution of the momentum of the ionized electrons, given an initial distribution of the electron in phase space W. In our simulations, the initial distribution of the electron in phase space is given by the quantum theory of ionization described in Sec. 1.2.2. In order to make a general description of the method, we consider the N-dimensional integral

$$I = \frac{\int_{\mathbb{D}} \mathrm{d}^{N} \mathbf{z} f(\mathbf{z}) W(\mathbf{z})}{\int_{\mathbb{D}} \mathrm{d}^{N} \mathbf{z} W(\mathbf{z})},\tag{D.1}$$

with $\mathbb{D} \subset \mathbb{R}^N$ a connected domain and $\mathbf{z} \in \mathbb{R}^N$. For instance, for computing the PMD of Fig. 3.4, we have used $f(\mathbf{z}) = \delta(p_z - \mathbf{e}_z \cdot \mathbf{p}(\mathbf{r}_{\text{PPT}}, \mathbf{p}_{\text{PPT}}, t_0; t \to \infty))$, where $W(\mathbf{z}) = W_{\text{PPT}}(t_0, \mathbf{p}_{\text{PPT}})$ is given in Eq. (1.27), $\mathbf{z} = (t_0, \mathbf{p}_{\text{PPT}})$ and δ is the Dirac distribution. The quantity $\mathbf{p}(\mathbf{r}_{\text{PPT}}, \mathbf{p}_{\text{PPT}}, t_0; t \to \infty)$ is the asymptotic momentum of the ionized trajectories (trajectories with positive energy) initiated at time t_0 with position \mathbf{r}_{PPT} and momentum \mathbf{p}_{PPT} given by Eqs. (1.25). In this case, the integral I corresponds to the normalized distribution of the asymptotic momentum of the ionized electron along \mathbf{e}_z . The Monte Carlo method for approximating multidimensional integrals consists in integrating fW by sampling f with non-uniform probability density $W(\mathbf{z})d^N\mathbf{z}$. We consider the set of points $\mathbf{z}_k \in \mathbb{D}$, with $k = 1, \ldots, M$, distributed with a probability density $W(\mathbf{z})$. If M is large, then

$$I \approx \frac{1}{M} \sum_{k=1}^{M} f(\mathbf{z}_k), \tag{D.2}$$

where terms of order $O(1/\sqrt{M})$ are neglected. If the density probability function $W(\mathbf{z})$ and \mathbb{D} have trivial forms, the generation of the set of sample points $\{\mathbf{z}_k\}_{k=1}^M$ can be performed using a *transformation method*. For instance, if $W(\mathbf{z}) = (2\pi)^{-N/2} \exp(-|\mathbf{z}|^2/2)$ (normal distribution) and $\mathbb{D} = \mathbb{R}^N$, the *Box-Muller algorithm* provides a normally distributed set of sample points (see Ref. [46]). However, if the density probability

function $W(\mathbf{z})$ does not allow the use of a transformation method, the set of sample points must be generated using an alternative algorithm. In this section, we show the *random walk Metropolis* (RMW) algorithm we use to generate a set of sample points distributed with an arbitrary probability density function $W(\mathbf{z})$ which allows us to find the approximation of I with Eq. (D.2).

The RWM algorithm [118], also referred to as the rejection method, consists in generating the sample points with nonuniform probability density $W(\mathbf{z})d^N\mathbf{z}$ using a biased random walk. The sample points we generate are a Markov chain [158, 159], for which the probability of being at \mathbf{z} is $W(\mathbf{z})$ and the transition kernel of the chain is $\mathbb{P}(\mathbf{z}, \cdot)$. Figure D.3 shows the organigram of the RWM algorithm. For a given point $\mathbf{z} \in \mathbb{D}$, a point \mathbf{z}^* is generated randomly, with $\mathbf{z}^* - \mathbf{z}$ uniformly and symmetrically distributed around the origin. For instance, in Ref. [118], $\mathbf{z}^* - \mathbf{z}$ is picked randomly in a square of length δ centered at the origin. The adjustable parameter δ is the *jump parameter*. We choose to pick $\mathbf{z}^* - \mathbf{z}$ in the ball of dimension N of radius δ and centered at the origin. In order to generate $\mathbf{z}^* - \mathbf{z}$ uniformly in the ball of dimension N centered at the origin, we notice that:

- The vector $\mathbf{z}^* \mathbf{z} = \delta \times u^{1/N} \mathbf{y}$ is uniformly distributed over the ball of dimension N of radius δ if and only if the vector \mathbf{y} is uniformly distributed over the sphere of dimension N of unitary radius and u is uniformly distributed, i.e., $u \sim \mathcal{U}(0, 1)$.
- The vector $\mathbf{y} = \mathbf{x}/|\mathbf{x}|$ is uniformly distributed over the sphere of dimension N of unitary radius if and only if the components of the vector $\mathbf{x} = (x_1, \ldots, x_N)$ are normally distributed, i.e., $x_k \sim \mathcal{N}(0, 1)$ for $k = 1, \ldots, N$. The generation of normally distributed random variables is achieved with the Box-Muller algorithm [46].

Then, the point \mathbf{z}^* is accepted with a probability $\min\{1, W(\mathbf{z}^*)/W(\mathbf{z})\}$. If $W(\mathbf{z}^*) > W(\mathbf{z}), \mathbf{z}^*$ is accepted and the sampling point \mathbf{z} takes the value of \mathbf{z}^* , i.e., $\mathbf{z} \leftarrow \mathbf{z}^*$. If $W(\mathbf{z}^*) < W(\mathbf{z}), \mathbf{z}^*$ is accepted with a probability $W(\mathbf{z}^*)/W(\mathbf{z})$, otherwise, it is rejected. If the point is rejected, \mathbf{z} keeps its value. The nonzero acceptance probability if $W(\mathbf{z}^*) < W(\mathbf{z})$ ensures we probe ergodically the domain \mathbb{D} . With zero acceptance probability,



Figure D.1: Random walk Metropolis (RWM) algorithm. Initially, δ is fixed. The variable $\mathbf{x} = (x_1, ..., x_N)$. The N_{th} iterations of thermalization are used for mixing the initial conditions, and the integral I is not computed during these steps.

D.1. STATISTICAL ANALYSIS



Figure D.2: Left panel: Schematic of the RWM for the ADK ionization rate given in Eq. (1.24) in the plane of initial conditions (t_0, p_{\perp}) . Middle panel: Histogram of the ADK ionization rate using the RWM for 5×10^4 iterations in logarithmic scale. Right panel: ADK ionization rate (1.24) in logarithmic scale.

there would exists δ such that \mathbf{z} would converge to a point in configuration space with a local minimum of probability $W(\mathbf{z})$. The probability of acceptance is chosen so that the equilibrium equation of the Markov chain is stationary

$$W(\mathbf{z}^{\star})\mathbb{P}(\mathbf{z}^{\star},\mathbf{z}) = W(\mathbf{z})\mathbb{P}(\mathbf{z},\mathbf{z}^{\star}).$$

In the algorithm, this equilibrium is reached after several iteration of the random walk. As a consequence, it is important to let a number of *thermalization steps* $N_{\rm th}$ in which there are no calculation of I. The thermlization steps are used to reach the stationary density probability. A schematic of the method and a resulting distribution for a low number of iterations is shown in Fig. D.2.

In the expressions of ionization rate of laser atom-interaction, for low ellipticities, the amplitude of the electric field (i.e., the ionization rate) is very low between two peaks. As a consequence, for low ellipticities, the probability that the the ionization time of the electron goes from one peak of the laser to another is very low using the RWM. In order to overcome this difficulty, we initiate multiple RWM around each peak of the electric field (i.e., ionization rate). In our simulations, the envelope of electric field is trapezoidal. We neglect the ionization during the ramp-up and the ramp-down. The initialization on each peak of the laser is done randomly and uniformly, i.e., there is the same probability for the RWM to be initiated on each of the peak amplitude of the electric field.

The parameter δ is the jump of the RWM algorithm. It is clear that δ plays a crucial role in the computation: If δ is small compared to the gradient of the probability function $W(\mathbf{z})$, the number of iterations required to probe the entire domain Ω is large, and as a consequence, the convergence of the approximation in Eq. (D.2) is slow with respect to M. If δ is large compared to the gradient of the probability function $W(\mathbf{z})$, the probability of acceptance is small, the point stays in the same region and the domain of integration is poorly probed, and as a consequence, the convergence of the integral is also slow. We denote α the acceptance probability of the method, i.e., the number of points in the Markov chain that transits over the number of points that have been generated. The acceptance probability is given by $\alpha_k = \sum_{j=1}^k c_j/k$, with $c_j = 1$ if the point \mathbf{z}^* is accepted at the *j*th iteration and $c_j = 0$ otherwise. The recurrence between the acceptance probability at the *k*th and the (k + 1)th iteration is $\alpha_{k+1} = (k\alpha_k + c_{k+1})/(k+1)$. The jump parameter δ is optimal if and only if

$$\lim_{k \to \infty} \alpha_k \approx 1/4.$$

Notice that for $k \to \infty$, $\alpha_{k+1} = \alpha_k$. There are no rigorous demonstration for this optimal value 1/4, but many problems show that this value optimizes the convergence of the approximation of I [158]. Given $\alpha_0 = 0$, the acceptance probability is computed according to $\alpha_k = (1 - 1/k)\alpha_{k-1} + c_k/k$ and is displayed at each iteration k in order to check its convergence.

D.1.2 Microcanonical generation of the initial conditions

We consider an atom of M electrons, whose positions and momenta are denoted $\mathbf{r}_j \in \mathbb{R}^d$ and $\mathbf{p}_j \in \mathbb{R}^d$, respectively, with $j = 1, \ldots, M$ and d is the dimension of the configuration space. We consider a time-

APPENDIX D. COMPUTATIONAL METHODS

independent Hamiltonian of the form

$$H\left(\{\mathbf{r}_{j},\mathbf{p}_{j}\}_{j=1}^{M}\right) = \sum_{j=1}^{M} \frac{|\mathbf{p}_{j}|^{2}}{2} + V\left(\mathbf{r}_{1},\dots,\mathbf{r}_{M}\right),$$
(D.3)

where $\sum_{j=1}^{M} |\mathbf{p}_j|^2/2$ is the total kinetic energy and $V(\mathbf{r}_1, \ldots, \mathbf{r}_M)$ is the total potential energy. For computing statistics, such as for instance the ionization probability curves (see Fig. 4.9) or the ionization time of the recolliding electron (see Fig. 4.12), the initial conditions of the electrons are generated randomly such that

$$\mathcal{E}_g = H\left(\{\mathbf{r}_j, \mathbf{p}_j\}_{j=1}^M\right),\tag{D.4}$$

where \mathcal{E}_g is the energy of the ground state of the atom. The initial conditions of the atom are generated randomly in two steps:

- The positions are generated uniformly in \mathbb{R}^d while $\mathcal{E}_q V(\mathbf{r}_1, \dots, \mathbf{r}_M) < 0$.
- The momenta are generated uniformly on the sphere of dimension dM of radius $[2(\mathcal{E}_g V(\mathbf{r}_1, \dots, \mathbf{r}_M)]^{1/2}$ centered at the origin of the momentum space.

D.2 Dynamical systems

We consider a dynamical system

$$\dot{\mathbf{z}}(t) = \mathbf{f}(\mathbf{z}, t),\tag{D.5}$$

where $\mathbf{z} \in \mathbb{R}^N$ is the set of dynamical variables and $\dot{\mathbf{z}} = d\mathbf{z}/dt$, where t is the evolution parameter. The flow is given by $\varphi_{t_0}^t(\boldsymbol{\zeta})$, and we consider $\varphi_{t_0+T}^{t_0+2T}(\mathbf{z}) = \varphi_{t_0}^{t_0+T}(\mathbf{z})$ for all t_0 and \mathbf{z} . We define the stroboscopic map $\boldsymbol{\mathcal{P}}$ such that

$$\begin{aligned} \mathcal{P}: \ \mathbf{z} &\to \boldsymbol{\varphi}_0^T(\mathbf{z}), \\ & \mathbb{R}^N &\mapsto \mathbb{R}^N, \end{aligned}$$
 (D.6)

where we chose \mathcal{P} with initial time $t_0 = 0$ without loss of generality. Poincaré map (D.6) corresponds to the Poincaré map given by Eq. (5.2) in the manuscript. We notice that the derivative of \mathcal{P} with respect to the set of coordinates \mathbf{z} is the tangent flow over one period $\mathcal{J}_0^T(\mathbf{z}) = \partial \varphi_0^T(\mathbf{z})/\partial \mathbf{z}$. Such map is particularly well suited for *T*-periodic dynamical systems, since *T*-periodic orbits solution of Eq. (D.5) are fixed points under the Poincaré map \mathcal{P} .

D.2.1 Newton's method

We consider the function \mathbf{F} of class \mathcal{C}^1 such that

$$\begin{split} \mathbf{F} \colon \mathbb{C}^N &\to \mathbb{C}^N, \\ \mathbf{q} &\mapsto \mathbf{F}(\mathbf{q}) \end{split}$$

Newton's method is an iterative procedure to determine an approximation of the zero of the function \mathbf{F} , denoted $\mathbf{q}^* \in \mathbb{C}^N$, and such that $\mathbf{F}(\mathbf{q}^*) = \mathbf{0}$. The algorithm of the Newton's method is shown in Fig. D.3. We assume that an initial guess \mathbf{q}_0 is known. For a given point \mathbf{q}_k , a new point $\mathbf{q}_{k+1} = \mathbf{q}_k + \Delta \mathbf{q}$ is computed in order to get closer to the true solution \mathbf{q}^* . We assume that the increment is small (i.e., $|\Delta \mathbf{q}| \ll 1$) and that the new point is close to the true solution [i.e., $\mathbf{F}(\mathbf{q}_k + \Delta \mathbf{q}) \approx \mathbf{0}$]. One obtains

$$\mathbf{F}(\mathbf{q}_k + \Delta \mathbf{q}) = \mathbf{F}(\mathbf{q}_k) + \left. \frac{\partial \mathbf{F}(\mathbf{q})}{\partial \mathbf{q}} \right|_{\mathbf{q}=\mathbf{q}_k} \Delta \mathbf{q} + O(\Delta \mathbf{q}^2) \approx \mathbf{0}$$

By neglecting the terms of order $\Delta \mathbf{q}^2$ and higher orders, one obtains a recurrence between the point \mathbf{q}_k and the new point $\mathbf{q}_{k+1} = \mathbf{q}_k + \Delta \mathbf{q}$. The increment $\Delta \mathbf{q} = \mathbf{q}_{k+1} - \mathbf{q}_k$ corresponds to the solution of the equation

$$\left. \frac{\partial \mathbf{F}(\mathbf{q})}{\partial \mathbf{q}} \right|_{\mathbf{q}=\mathbf{q}_k} \Delta \mathbf{q} = -\mathbf{F}(\mathbf{q}_k), \tag{D.7}$$

This equation can be solved in multiple ways: For instance, by multiplying the left- and right-hand side of the equation by the inverse matrix of $\partial \mathbf{F}(\mathbf{q})/\partial \mathbf{q}|_{\mathbf{q}=\mathbf{q}_k}$, by using the *Cramer rule* or the *Gauss elimination*. The recurrence process of Newton's method is iterated until $|\mathbf{F}(\mathbf{q}_k)| < \epsilon$, where ϵ is infinitesimal.

D.2.2 Periodic orbits and fixed points

Initial conditions \mathbf{z}^* of trajectories leading to fixed points and periodic orbits can be written in the form $\mathbf{F}(\mathbf{z}^*) = \mathbf{0}$. Hence, Newton's method can be used to find \mathbf{z}^* . In this section, we identify the form of the function \mathbf{F} and the recurrence equation (D.7) of the Newton's method for computing fixed points and periodic orbits of the dynamical system (D.5).

D.2.2.1 Fixed points of the continuous system

Fixed points of the dynamical system (D.5), denoted \mathbf{z}^* , are such that $\boldsymbol{\varphi}_{t_0}^t(\mathbf{z}^*) = \mathbf{z}^*$. Hence, determining fixed points of the continuous dynamical system (D.5) consists in finding the zeros of the function $\mathbf{f}(\mathbf{z}, t)$. One can use the Newton's method to determine \mathbf{z}^* such that $\mathbf{f}(\mathbf{z}^*, t) = \mathbf{0}$ for all t (see Sec. D.2.1 with $\mathbf{F} \to \mathbf{f}$ and $\mathbf{q} \to \mathbf{z}$).

D.2.2.2 Fixed points of the Poincaré map (D.6)

We consider a *T*-periodic orbit of the dynamical system (D.5). Under the Poincaré map \mathcal{P} , *T*-periodic orbits are fixed points. As a consequence, the initial point \mathbf{z}^* leading to a periodic orbit of the dynamical system (D.5) is such that $\mathcal{P}(\mathbf{z}^*) = \mathbf{z}^*$. Computing a periodic orbit of period *T* of the dynamical system (D.5) consists in computing the zero of the function

$$\mathbf{F}(\mathbf{z}) = \boldsymbol{\mathcal{P}}(\mathbf{z}) - \mathbf{z}.$$
 (D.8)

To compute \mathbf{z}^* such that $\mathbf{F}(\mathbf{z}^*) = \mathbf{0}$, we use Newton's method (see Sec. D.2.1). First, Newton's method needs an initial guess \mathbf{z}_0 . For instance, the initial guess can be chosen as a fixed point of the map of an integrable system using perturbative theory. Then, one has $\partial \mathcal{P}(\mathbf{z})/\partial \mathbf{z} = \mathcal{J}_0^T(\mathbf{z})$, where $\mathcal{J}_0^T(\mathbf{z}) = \partial \varphi_0^T(\mathbf{z})/\partial \mathbf{z}$ is the tangent flow. The increment in the Newton's method $\Delta \mathbf{z}$ is solution of the recurrence equation [equivalent of Eq. (D.7) with \mathbf{F} given by Eq. (D.8)]

$$\left[\boldsymbol{\mathcal{J}}_{0}^{T}(\mathbf{z}_{k})-\mathbf{I}_{N}\right]\Delta\mathbf{z}=\mathbf{z}_{k}-\boldsymbol{\mathcal{P}}(\mathbf{z}_{k}).$$
(D.9)

We recall that the evolution of the tangent flow is given by $\mathcal{J}_0^t(\mathbf{z}) = \mathbf{A}(\boldsymbol{\varphi}_0^t(\mathbf{z}), t)\mathcal{J}_0^t(\mathbf{z})$, where $\mathbf{A}(\mathbf{z}(t), t) = \partial \mathbf{f}(\mathbf{z}, t)/\partial \mathbf{z}|_{\mathbf{z}=\mathbf{z}(t)}$ is the Jacobian matrix of the map. The initial conditions of the tangent flow are $\mathcal{J}_0^0(\mathbf{z}) = \mathbf{I}_N$. In total, an ordinary differential equation of N(N+1) variables (N quantities for the flow $\boldsymbol{\varphi}_0^T$ and N^2 for



Figure D.3: Newton's method algorithm for finding \mathbf{q}^* such that $\mathbf{F}(\mathbf{q}^*) = \mathbf{0}$. The solution of Eq. (D.7) is found using Gauss elimination with maximum pivoting. The tolerance ϵ is infinitesimal and adjustable. The maximum of iteration k_{max} is used to quit the algorithm in case that it does not converge.

the tangent flow \mathcal{J}_0^T is integrated at each iteration of the Newton's method. For sufficiently large k, the point \mathbf{z}_k converges to the fixed point \mathbf{z}^* , i.e., $\mathbf{z}_k \approx \mathbf{z}^*$.

D.2.2.3 Continuation methods: Evolution of the fixed point as a function of a parameter

We consider the dynamical system (D.5) depends explicitly in a parameter μ such that $\mathbf{f}(\mathbf{z}, \mu, t)$. As a consequence, the flow solution of the ordinary differential equation (D.5) also depends on μ . We denote $\varphi_{t_0}^t(\mathbf{z}, \mu)$. We assume that there exists a curve in the extended coordinates-parameter space for which \mathbf{z}^* is a fixed point of the dynamical system (D.5) for a given parameter μ . The set of fixed points \mathbf{z}^* as a function of the parameter μ is a curve in the extended coordinates-parameter space defined by

$$g(\mathbf{z}^{\star}, \mu) = 0, \tag{D.10}$$

as depicted in Figs. D.4a and D.4b. The objective is to track the fixed points \mathbf{z}^* as a function of the parameter μ along this curve. We consider a discrete set of points and parameters $\{\mathbf{z}^{(j)}\}_j$ and $\{\mu^{(j)}\}_j$ belonging to this curve, such that $g(\mathbf{z}^{(j)}, \mu^{(j)}) = 0$. The intuitive way for tracking \mathbf{z}^* as a function of μ consists in using the mesh of points $\mu = \mu^{(0)}, \ldots, \mu^{(m)}$ with $\mu^{(j)} < \mu^{(j+1)}$ and to assume that there exists a fixed point $\mathbf{z}^{(j)}$ such that $g(\mathbf{z}^{(j)}, \mu^{(j)}) = 0$ for all j. We assume that the fixed point $\mathbf{z}^{(0)}$ is known for $\mu^{(0)}$. For a given $\mu^{(j+1)}$, we compute the associated fixed point $\mathbf{z}^{(j+1)}$. In Sec. D.2.2.2, we show the method to compute a fixed point $\mathbf{z}^{(j+1)}$, the fixed point $\mathbf{z}^{(j+1)}$ is solution of the equation

$$\mathbf{F}^{\star}\left(\mathbf{z}^{(j+1)},\mu^{(j+1)}\right) = \mathbf{0},\tag{D.11}$$

where $\mathbf{F}^{\star}(\mathbf{z}, \mu) = \mathbf{f}(\mathbf{z}, \mu, t)$ if \mathbf{z}^{\star} is the fixed point of the flow (see Sec. D.2.2.1), and $\mathbf{F}^{\star}(\mathbf{z}, \mu) = \mathcal{P}(\mathbf{z}, \mu) - \mathbf{z}$ if \mathbf{z}^{\star} it the fixed of the Poincaré map (D.6) [see Eq. (D.8)]. In both cases, Newton's method is employed with the initial guess $\mathbf{z}_{0}^{(j+1)} = \mathbf{z}^{(j)}$, where we denote $\mathbf{z}_{k}^{(j)}$ the kth iteration of the Newton's method for the computation of the fixed point $\mathbf{z}^{(j)}$ associated with $\mu^{(j)}$. However most of the time, the fixed point does not vary monotonically with respect to the parameter, as it is the case, for instance, in the right panel of Fig. 5.8 and Fig. D.4a. In particular, the method fails at the bifurcation of the fixed point \mathbf{z}^{\star} with respect to μ , for instance, when the period of the periodic orbit double, and this method stops. Instead, we use a continuation method which let μ free.

Given a point $(\mathbf{z}^{(j)}, \mu^{(j)})$ on the red curve depicted in Fig. D.4, the objective is to move along this curve by an increment δ . If δ is too small, the method requires a large amount of points to reproduce the curve. If δ is too large, the curve is not necessary smooth, and the algorithm does not necessarily converges to fixed points associated with the previous fixed point. Hence, δ is controlled during the computation. The relation between two successive points on the curve is

$$F_{\delta}\left(\mathbf{z}^{(j)}, \mathbf{z}^{(j+1)}, \mu^{(j)}, \mu^{(j+1)}\right) = \left|\mathbf{z}^{(j+1)} - \mathbf{z}^{(j)}\right|^{2} + \left|\mu^{(j+1)} - \mu^{(j)}\right|^{2} - \delta^{2} = 0.$$
(D.12)

To conclude, given a point $(\mathbf{z}^{(j)}, \mu^{(j)})$, the point $(\mathbf{z}^{(j+1)}, \mu^{(j+1)})$ is solution of Eqs. (D.11) and (D.12). In other words, the method requires that for each sample points on the red curve of Fig. D.4, we use Newton's method to determine the zeros $(\mathbf{z}^{(j+1)}, \mu^{(j+1)})$ of the function

$$\mathbf{F}\left(\mathbf{z}^{(j+1)}, \mathbf{z}^{(j)}, \mu^{(j+1)}, \mu^{(j)}\right) = \begin{bmatrix} \mathbf{F}^{\star}\left(\mathbf{z}^{(j+1)}, \mu^{(j+1)}\right) \\ F_{\delta}\left(\mathbf{z}^{(j)}, \mathbf{z}^{(j+1)}, \mu^{(j)}, \mu^{(j+1)}\right) \end{bmatrix}.$$
 (D.13)

The method is two steps, as shown in Fig. D.4b. We assume that a fixed point $\mathbf{z}^{(j)}$ is known for a given parameter $\mu^{(j)}$, the point $(\mathbf{z}^{(j+1)}, \mu^{(j+1)})$ is determined with Newton's method as follow:

(i) The initial guess is determined with

$$\mathbf{z}_{0}^{(j+1)} = \mathbf{z}^{(j)} + \delta \left[\mathbf{z}^{(j)} - \mathbf{z}^{(j-1)} \right],$$

$$\mu_{0}^{(j+1)} = \mu^{(j)} + \delta \left[\mu^{(j)} - \mu^{(j-1)} \right].$$

If j = 0, $\mathbf{z}_0^{(1)} = \mathbf{z}^{(0)}$ and $\mu_0^{(1)} = \mu^{(0)} \pm \delta$. The sign of $\mu_0^{(1)} - \mu^{(0)}$ determines the direction in which the curve $\mathbf{g}(\mathbf{z}^*, \mu)$ is traveled. When j > 1, the curve is automatically traveled in the direction set at j = 1.



Figure D.4: (a) Position of the fixed point projected along \mathbf{e}_x of the periodic \mathcal{O}_A (see Chap. 5) for $\xi = 0.5$ as a function of the intensity $\mu = I$. (b) Illustration of the step (i) and (ii) of the continuation method for tracking a fixed point \mathbf{z}^* as a function of a parameter μ . The thick red curve $g(\mathbf{z}^*, \mu) = 0$ is the location of the fixed point as a function the parameter μ . (b) is a zoom of (a). (c) Continuous method algorithm. The objective of this algorithm is to track the fixed point \mathbf{z}^* as a function of the parameter μ . The value of the parameter δ is changed along the method to improve the convergence of the Newton's method. The threshold k_{\min} and k_{\max} are the criteria for the convergence of the Newton's method. The method stops when the parameter $\mu^{(j)}$ is out of the bounds $[\mu_{\min}, \mu_{\max}]$ or when the point $(\mathbf{z}^{(j)}, \mu^{(j)})$ is close to the first point $(\mathbf{z}^{(0)}, \mu^{(0)})$. In the latter case, $g(\mathbf{z}^*, \mu)$ describes a closed curve in the extended coordinates-parameter space.

APPENDIX D. COMPUTATIONAL METHODS

(ii) The initial guess $(\mathbf{z}_0^{(j+1)}, \mu_0^{(j+1)})$ is refined using Newton's method. Given the point at the *k*th iteration of the Newton's method $(\mathbf{z}_k^{(j+1)}, \mu_k^{(j+1)})$, we increment the guess with $(\mathbf{z}_{k+1}^{(j+1)}, \mu_{k+1}^{(j+1)}) = (\mathbf{z}_k^{(j+1)}, \mu_k^{(j+1)}) + (\Delta \mathbf{z}, \Delta \mu)$. The increment is solution of the equation

$$\begin{bmatrix} \frac{\partial \mathbf{F}^{\star}(\mathbf{z},\mu)}{\partial \mathbf{z}} \Big|_{\left(\mathbf{z}=\mathbf{z}_{k}^{(j+1)},\mu=\mu_{k}^{(j+1)}\right)} & \frac{\partial \mathbf{F}^{\star}(\mathbf{z},\mu)}{\partial \mu} \Big|_{\left(\mathbf{z}=\mathbf{z}_{k}^{(j+1)},\mu=\mu_{k}^{(j+1)}\right)} \\ 2\left(\mathbf{z}_{k}^{(j+1)}-\mathbf{z}^{(j)}\right)^{\top} & 2\left(\mu_{k}^{(j+1)}-\mu^{(j)}\right) \end{bmatrix} \begin{bmatrix} \Delta \mathbf{z} \\ \Delta \mu \end{bmatrix} \\ &= \begin{bmatrix} \mathbf{F}^{\star}\left(\mathbf{z}^{(j+1)},\mu^{(j+1)}\right) \\ \left|\mathbf{z}_{k}-\mathbf{z}^{(j)}\right|^{2}+\left|\mu_{k}-\mu^{(j)}\right|^{2}-\delta^{2} \end{bmatrix}, \quad (D.14)$$

If δ is sufficiently small and the curve g sufficiently smooth, there exists only two solutions for the system. The parameter δ is adjusted during the continuation method. The number of iteration in which the Newton's method converges for a given j is denoted k_c : If $k_c < k_{\min}$, δ increases, and if $k_c > k_{\max}$, δ decreases, where k_{\min} and k_{\max} are two adjustable parameters.

In step (ii) of the continuation method, we use Newton's method. The derivatives of the function \mathbf{F}^* has to be computed with respect to \mathbf{z} and μ . If $\mathbf{F}^*(\mathbf{z}, \mu) = \mathbf{f}(\mathbf{z}, \mu, t)$ (i.e., if \mathbf{z}^* is the fixed point of the flow) we compute $\partial \mathbf{f}(\mathbf{z}, \mu, t)/\partial \mathbf{z}$ and $\partial \mathbf{f}(\mathbf{z}, \mu, t)/\partial \mu$. However, if $\mathbf{F}^*(\mathbf{z}, \mu) = \mathcal{P}(\mathbf{z}, \mu) - \mathbf{z}$ [i.e., if \mathbf{z}^* is the fixed of the Poincaré map (D.6)] the computation of the derivatives is more complex. First, $\partial \mathbf{F}^*(\mathbf{z}, \mu)/\partial \mathbf{z} = \mathcal{J}_0^T(\mathbf{z}, \mu) - \mathbf{I}_N$, such as it is done in Sec. D.2.2.2 for \mathcal{P} which does not dependent on μ . Concerning the derivative of \mathcal{P} with respect to μ , we substitute $\mathbf{z}(t) = \varphi_0^t(\mathbf{z}, \mu)$ in Eq. (D.5), and we derive the left- and right-hand side of the equation with respect to μ . Using the regularity of the flow function and the chain rule, the evolution of the derivative of the flow with respect to μ is given by

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[\frac{\partial \boldsymbol{\varphi}_0^t(\mathbf{z}, \mu)}{\partial \mu} \right] = \mathbf{A} \left(\boldsymbol{\varphi}_0^T(\mathbf{z}, \mu), t \right) \left[\frac{\partial \boldsymbol{\varphi}_0^t(\mathbf{z}, \mu)}{\partial \mu} \right] + \left. \frac{\partial \mathbf{f} \left(\mathbf{z}, \mu, t \right)}{\partial \mu} \right|_{\mathbf{z} = \boldsymbol{\varphi}_0^t(\mathbf{z}, \mu)}. \tag{D.15}$$

The initial condition of the derivative of the flow with respect to μ is $\partial \varphi_0^0(\mathbf{z}, \mu)/\partial \mu = \mathbf{0}$. In Eq. (D.14), $\partial \mathbf{F}^*(\mathbf{z}, \mu)/\partial \mu = \partial \mathcal{P}(\mathbf{z}, \mu)/\partial \mu = \partial \varphi_0^T(\mathbf{z}, \mu)/\partial \mu$. At each step of the Newton's method, N(N+2) variables are integrated: N variables for the flow φ_0^T , N^2 variables for the tangent flow $\mathcal{J}_0^T = \partial \varphi_0^T$, and N variables for the derivative of the flow with respect to the parameter $\partial \varphi_0^T/\partial \mu$. The evolution of the latter is given by Eq. (D.15).

D.2.3 Computation of one-dimensional invariant manifolds of fixed points: Hobson's method

We consider a fixed point \mathbf{z}^* under the Poincaré map \mathcal{P} defined in Eq. (D.6). We consider \mathbf{M}_T to be the monodromy matrix of the fixed point. The eigenvalues and associated eigenvectors of the monodromy matrix are denoted λ_j and \mathbf{v}_j , respectively. We consider a small displacement h from the fixed point \mathbf{z}^* along one eigendirection \mathbf{v}_j , i.e., $\mathbf{z} = \mathbf{z}^* + h\mathbf{v}_j$. We consider h small enough, such that $h|\mathbf{v}_j| \ll 1$, $\mathcal{P}^k(\mathbf{z}^* + h\mathbf{v}_j) \approx \mathbf{z}^* + h\mathcal{J}_0^{hT}(\mathbf{z}^*)\mathbf{v}_j$ and $\mathcal{P}^{-k}(\mathbf{z}^* + h\mathbf{v}_j) \approx \mathbf{z}^* + h\mathcal{J}_{kT}^0(\mathbf{z}^*)\mathbf{v}_j$. Since \mathbf{z}^* is a fixed point of the map $\mathcal{P}: \mathbf{z} \mapsto \varphi_0^T(\mathbf{z}), \ \mathcal{J}_0^{kT}(\mathbf{z}^*) = [\mathcal{J}_0^T(\mathbf{z}^*)]^k$ and $\mathcal{J}_{kT}^0(\mathbf{z}^*) = [\mathcal{J}_0^T(\mathbf{z}^*)]^k$. Using the chain rule, one can show that $\mathbf{M}_T(\mathbf{z}^*) = \mathcal{J}_0^T(\mathbf{z}^*) = [\mathcal{J}_T^0(\mathbf{z}^*)]^{-1}$. Therefore,

$$\boldsymbol{\mathcal{P}}^{k}(\mathbf{z}^{\star} + h\mathbf{v}_{j}) \approx \mathbf{z}^{\star} + h\mathbf{M}_{T}(\mathbf{z}^{\star})^{k}\mathbf{v}_{j} = \mathbf{z}^{\star} + h\lambda_{j}^{k}\mathbf{v}_{j}, \qquad (D.16a)$$

$$\boldsymbol{\mathcal{P}}^{-k}(\mathbf{z}^{\star} + h\mathbf{v}_j) \approx \mathbf{z}^{\star} + h\mathbf{M}_T(\mathbf{z}^{\star})^{-k}\mathbf{v}_j = \mathbf{z}^{\star} + h\lambda_j^{-k}\mathbf{v}_j, \qquad (D.16b)$$

where we have used $\mathbf{M}_T(\mathbf{z}^*)\mathbf{v}_j = \lambda_j \mathbf{v}_j$. According to Eqs. (5.8) and (5.9), the set of points which converge to the fixed point for infinite forward (resp. backward) integration time is on the stable (resp. unstable) manifold. In Eq. (D.16a), for $k \to \infty$, $\mathcal{P}^k(\mathbf{z}^* + h\mathbf{v}_j) = \mathbf{z}^*$ if and only if $\lambda_j < 1$. We denote λ_s this eigenvalue and \mathbf{v}_s its associated eigenvector. Therefore, the eigenvector \mathbf{v}_s associated with the eigenvalue λ_s is the linear approximation of the stable manifold of \mathbf{z}^* . In Eq. (D.16b), for $k \to \infty$, $\mathcal{P}^{-k}(\mathbf{z}^* + h\mathbf{v}_j) = \mathbf{z}^*$ if and only if $\lambda_j > 1$. We denote λ_u this eigenvalue and \mathbf{v}_u its associated eigenvector. Therefore, the eigenvector \mathbf{v}_u is the linear approximation of the unstable manifold of \mathbf{z}^* .

The fundamental domain of the invariant manifolds is the interval parametrized by s in which the linear approximation of the manifolds is valid. The fundamental domain of the stable and unstable manifolds is



Figure D.5: Hobson's method [74] algorithm to compute the one-dimensional unstable manifold of the fixed point \mathbf{z}^* of the mapping \mathcal{P} . The parameter $ds \in [ds_{\min}, ds_{\max}]$ is the key parameter of the method and has to be initiated small. Threshold parameters are $ds_{\max}, ds_{\min}, \alpha_{\min}, \alpha_{\max}$ and $d\alpha_{\max}$. The quantity L is the total length of the computed manifold, while L_{\max} is the total length of the manifold to compute. The ensemble $\mathcal{U} = \{\mathbf{z}_1, \ldots, \mathbf{z}_{\#\mathcal{U}}\}$ is a mesh points on a piece of the unstable manifold, such that $\mathbf{z}_{\#\mathcal{U}} = \mathcal{P}(\mathbf{z}_1)$. Each point of \mathcal{U} is ordered along the manifold and such that $|\mathbf{z}_j - \mathbf{z}_{j-1}| < ds_{\min}$, or $\alpha = \cos^{-1}(\mathbf{z}_j \cdot \mathbf{z}_{j-1}/(|\mathbf{z}_j||\mathbf{z}_{j-1}|)) < \alpha_{\max}$ and $\alpha |\mathbf{z}_j - \mathbf{z}_{j-1}| < d\alpha_{\max}$.

the set of points such that

$$\mathcal{F}^{s}(\mathbf{z}^{\star};s) = \left\{ \mathbf{z} \in \mathbb{R}^{N}, s \in [0,1] \mid \mathbf{z} = (1-s)\left(\mathbf{z}^{\star} + h\mathbf{v}^{s}\right) + s\mathcal{P}^{-1}(\mathbf{z}^{\star} + h\mathbf{v}^{s}) \right\}$$

$$\mathcal{F}^{u}(\mathbf{z}^{\star};s) = \left\{ \mathbf{z} \in \mathbb{R}^{N}, s \in [0,1] \mid \mathbf{z} = (1-s)\left(\mathbf{z}^{\star} + h\mathbf{v}^{u}\right) + s\mathcal{P}(\mathbf{z}^{\star} + h\mathbf{v}^{u}) \right\},$$

where h is the largest possible value such that $|\mathcal{P}^{-1}(\mathbf{z}^* + h\mathbf{v}^s) - (\mathbf{z}^* + h\lambda_s^{-1}\mathbf{v}^s)| < \epsilon$ for the stable manifold, and $|\mathcal{P}(\mathbf{z}^* + h\mathbf{v}^u) - (\mathbf{z}^* + h\lambda_u\mathbf{v}^u)| < \epsilon$ for the unstable manifold, where ϵ is infinitesimal. Each point of the grown invariant manifold is an image of one point in the fundamental domain. Therefore, the fundamental domain contains the information on the grown invariant manifolds. In order to compute the grown manifolds, we use the Hobson's method [74] which is shown in Fig. D.5. Figure D.5 shows algorithm for computing the unstable manifold. The algorithm for computing the stable manifold is the same, except that \mathbf{v}_u is replaced by \mathbf{v}_s and \mathcal{P} is replaced by \mathcal{P}^{-1} . The idea of the algorithm of the Hobson's method is to consider a sample of points $\mathcal{U} = \{\mathbf{z}_1, \dots, \mathbf{z}_{\#\mathcal{U}}\}$ which approximate locally the invariant manifold, and such that $\mathbf{z}_{\#\mathcal{U}} = \mathcal{P}(\mathbf{z}_1)$. In other words, the points \mathbf{z}_j are the skeleton of the invariant manifold, for $j = 1, \dots, \#\mathcal{U}$. The points \mathbf{z}_j are used to compute new points of the invariant manifolds. By interpolating the set of points, we find two points at a distance ds and 2ds from \mathbf{z}_1 , denoted \mathbf{u}_1 and \mathbf{u}_2 , respectively. The point $\mathbf{u}_1' = \mathcal{P}(\mathbf{u}_1)$ is possibly included in the set of sample points \mathcal{U} , while $\mathbf{u}_2' = \mathcal{P}(\mathbf{u}_2)$ is a test point for anticipating the sudden variations of the invariant manifolds. We also denote $\mathbf{u}_0 = \mathbf{z}_{\#\mathcal{U}} = \mathcal{P}(\mathbf{z}_1)$.

The graphics representation of the stable and unstable manifolds are a set of points of the stable and unstable manifolds, respectively. In order to have a smooth two-dimensional representation of the invariant manifolds, the angle between three successive sampled points of the representation must be small enough. We use a threshold parameter α_{\max} to control the smoothness of the representation. Let α be the angle between $\mathbf{u}_1 - \mathbf{u}_0$ and $\mathbf{u}_2 - \mathbf{u}_1$. The sampled point must be such that $\alpha < \alpha_{\max}$. Otherwise, the point \mathbf{u}'_1 is rejected and ds is decreased. Also, the quantitative resolution of the invariant manifold is ensured by keeping $\alpha |\mathbf{u}'_0 - \mathbf{u}'_1|$ small enough. The sampled point must be such that $\alpha |\mathbf{u}'_0 - \mathbf{u}'_1| < d\alpha_{\max}$. Otherwise, the point \mathbf{u}'_1 is rejected and ds is decreased.

D.2.4 Computation of invariant curves and their stability

We denote $\mathbf{x}(\theta)$ an invariant curve under \mathcal{P} parametrized by $\theta \in \mathbb{T}$ (which corresponds to $\mathbb{R}/2\pi\mathbb{Z}$). If the curve is invariant, there exists $\nu \in \mathbb{R} \setminus 2\pi\mathbb{Q}$ such that

$$\mathcal{P}(\mathbf{x}(\theta)) = \mathbf{x}(\theta + \nu). \tag{D.17}$$

Figure D.6a illustrates the invariant curve under the map \mathcal{P} . The invariant curves $\mathbf{x}(\theta)$ are defined on a Cantor set. We describe the method for two cases are studied: The case for which \mathcal{P} depends explicitly on the parameter of the invariant curve θ is referred to as the *non-autonomous* case. The case for which \mathcal{P} does not depend explicitly on the parameter of the invariant curve θ is referred to as the *non-autonomous* case. The case for which \mathcal{P} does not depend explicitly on the parameter of the invariant curve θ is referred to as the *autonomous* case. The autonomous case is an extension of the non-autonomous case. In both cases, we use the Fourier representation method [79] for computing invariant curves of the map \mathcal{P} and their linear stability. The Fourier representation of the invariant curve is

$$\mathbf{x}(\theta; \boldsymbol{\alpha}) = \mathbf{a}_0 + \sum_{k=1}^{M} \left[\mathbf{a}_k \cos(k\theta) + \mathbf{b}_k \sin(k\theta) \right],$$
(D.18)

where 2M + 1 is the number of coefficients and $\boldsymbol{\alpha} = [\mathbf{a}_0, \dots, \mathbf{a}_M, \mathbf{b}_1, \dots, \mathbf{b}_M]^\top$. In theory, M goes to infinity, but numerically, M must be finite. The function $\mathbf{x}(\theta; \boldsymbol{\alpha})$ is analytic and therefore the Fourier coefficients decrease exponentially as k increases, as observed in Fig. D.6b. Numerically, M must be large enough for the series of coefficients to converge, i.e., $|\mathbf{a}_M| + |\mathbf{b}_M| < \epsilon$ with ϵ infinitesimal. The objective is, as well in the autonomous as in the non-autonomous case, to determine the set of coefficients $\boldsymbol{\alpha}$ such that $\mathbf{x}(\theta; \boldsymbol{\alpha})$ is solution of Eq. (D.17). Then, we compute the linear stability of the invariant curves surrounding the fixed point of \mathcal{O}_S . The linear stability of these curves is used to compute their associated one-dimensional invariant manifolds along the saddle direction. The invariant manifolds of the invariant curves of \mathcal{O}_S are compared with the dynamics of the electron.

D.2.4.1 Non-autonomous map

We consider a map \mathcal{P} which depends explicitly in θ , a skew-product

$$\overline{\mathbf{z}} = \boldsymbol{\mathcal{P}}(\mathbf{z}, \theta), \\ \overline{\theta} = \theta + \nu,$$



Figure D.6: Non-autonomous map $\mathcal{P}: (x, y) \mapsto (x(2+y) + \epsilon \cos(\theta), y(1/2-x) + \epsilon \sin(\theta))$. (a) Invariant curve $\mathbf{x}(\theta)$ of frequency $\nu = \sqrt{2}$, the mesh of points $\mathbf{x}_j(\boldsymbol{\alpha}) = \mathbf{x}(\theta_j; \boldsymbol{\alpha})$, the unstable and stable eigenvectors for each angle parameter $\Psi_j^u(\boldsymbol{\alpha}) = \Psi^u(\theta_j; \boldsymbol{\alpha})$ (red vectors) and $\Psi_j^s(\boldsymbol{\alpha}) = \Psi^s(\theta_j; \boldsymbol{\alpha})$ (blue vectors), respectively. Notice that the eigenvectors $-\Psi_j^u(\boldsymbol{\alpha})$ and $-\Psi_j^s(\boldsymbol{\alpha})$ are not shown but are also eigenvectors. (b) Norm of the coefficients of the Fourier representation of the invariant curve and the eigenvectors as a function of the harmonic order k. (c) Spectrum λ of the matrix of stability of the invariant curve in the complex plane.

with $\mathcal{P}: \mathbb{R}^N \to \mathbb{R}^N$ and $\mathbf{z} \in \mathbb{R}^N$. In Eq. (D.17), we use the Fourier representation of the invariant curve given by Eq. (D.18). There are 2M + 1 coefficients, and as a consequence, (2M + 1)N unknowns. We consider a mesh of parameters $\theta = \{\theta_j\}_{j=0}^{2M}$. We define

$$\theta_j = j \frac{2\pi}{2M+1},\tag{D.19}$$

with $0 \le j \le 2M$. Each point associated with each angle $\mathbf{x}(\theta_j; \boldsymbol{\alpha})$ is a solution of the Eq. (D.17). Hence, the zero function of the 2M + 1 points solution of the invariance equation (D.17) reads

$$\mathbf{F}(\boldsymbol{\alpha}) = \begin{bmatrix} \boldsymbol{\mathcal{P}}\left(\mathbf{x}(\theta_{0};\boldsymbol{\alpha}),\theta_{0}\right) - \left(\mathbf{x}(\theta_{0};\boldsymbol{\alpha})\right) \\ \vdots \\ \boldsymbol{\mathcal{P}}\left(\mathbf{x}(\theta_{2M};\boldsymbol{\alpha}),\theta_{2M}\right) - \left(\mathbf{x}(\theta_{2M};\boldsymbol{\alpha})\right) \end{bmatrix} = \mathbf{0}.$$
 (D.20)

We use Newton's method described in Sec. D.2.1 to compute $\boldsymbol{\alpha}$, the zero of $\mathbf{F}(\boldsymbol{\alpha})$. The initial guess of the Newton's method are given by $\mathbf{a}_0 = \mathbf{z}^*$ and $\mathbf{a}_j = \mathbf{b}_j = \mathbf{0}$ for $j = 2, \ldots, M$. Also, $\mathbf{a}_1 = h\Re(\mathbf{v}_c)$ and $\mathbf{b}_1 = h\Re(\mathbf{v}_c)$, or $\mathbf{a}_1 = h\Re(\mathbf{v}_c)$ and $\mathbf{b}_1 = h\Re(\mathbf{v}_c)$, depending on the running direction of the electrons along the invariant curve. The complex vector \mathbf{v}_c is the eigenvector associated with the center component of the fixed point (complex eigenvalue of the fixed point). In order to compute the derivatives of the function \mathbf{F} with respect to the coefficients $\boldsymbol{\alpha}$, it is rewritten in the form $\mathbf{F}(\boldsymbol{\alpha}) = \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\boldsymbol{\alpha})) - \mathbf{Y}(\boldsymbol{\alpha})$, where $\mathbf{X}(\boldsymbol{\alpha}) = [\mathbf{x}_0(\boldsymbol{\alpha}), \ldots, \mathbf{x}_{2M}(\boldsymbol{\alpha})]^\top$, $\mathbf{Y}(\boldsymbol{\alpha}) = [\mathbf{y}_0(\boldsymbol{\alpha}), \ldots, \mathbf{y}_{2M}(\boldsymbol{\alpha})]^\top$, $\mathbf{P}_{\mathbf{X}}(\mathbf{X}(\boldsymbol{\alpha})) = [\mathcal{P}(\mathbf{x}_0, \theta_0), \ldots, \mathcal{P}(\mathbf{x}_{2M}, \theta_{2M})]^\top$, $\mathbf{x}_j(\boldsymbol{\alpha}) = \mathbf{x}(\theta_j; \boldsymbol{\alpha})$ and $\mathbf{y}_j(\boldsymbol{\alpha}) = \mathbf{x}(\theta_j + \nu; \boldsymbol{\alpha})$. Hence,

$$\frac{\partial \mathbf{X}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} = \begin{bmatrix} \mathbf{I}_N & \cdots & \mathbf{I}_N \cos(M\theta_0) & \mathbf{I}_N \sin(\theta_0) & \cdots & \mathbf{I}_N \sin(M\theta_0) \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{I}_N & \cdots & \mathbf{I}_N \cos(N\theta_{2M}) & \mathbf{I}_N \sin(\theta_{2M}) & \cdots & \mathbf{I}_N \sin(M\theta_{2M}) \end{bmatrix}, \quad (D.21)$$

where \mathbf{I}_N the identity matrix of size N. Similarly,

$$\frac{\partial \mathbf{Y}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} = \begin{bmatrix} \mathbf{I}_N & \cdots & \mathbf{I}_N \cos(M(\theta_0 + \nu)) & \mathbf{I}_N \sin(\theta_0 + \nu) & \cdots & \mathbf{I}_N \sin(M(\theta_0 + \nu)) \\ \vdots & \ddots & \vdots & \ddots & \vdots \\ \mathbf{I}_N & \cdots & \mathbf{I}_N \cos(M(\theta_{2M} + \nu)) & \mathbf{I}_N \sin(\theta_{2M} + \nu) & \cdots & \mathbf{I}_N \sin(M(\theta_{2M} + \nu)) \end{bmatrix}.$$
(D.22)

The derivative of the function $\mathbf{P}_{\mathbf{X}}(\mathbf{X}(\alpha))$ with respect to the coefficients is computed using the chain rule $\partial \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\alpha))/\partial \alpha = (\partial \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\alpha))/\partial \mathbf{X}(\alpha))(\partial \mathbf{X}(\alpha))/\partial \alpha$, where

$$\frac{\partial \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\boldsymbol{\alpha}))}{\partial \mathbf{X}(\boldsymbol{\alpha})} = \begin{bmatrix} \frac{\partial \boldsymbol{\mathcal{P}}(\mathbf{z}, \theta)}{\partial \mathbf{z}} \Big|_{(\mathbf{z}=\mathbf{x}_{0}(\boldsymbol{\alpha}), \theta=\theta_{0})} & \cdots & \mathbf{0}_{N} \\ \vdots & \ddots & \vdots \\ \mathbf{0}_{N} & \cdots & \frac{\partial \boldsymbol{\mathcal{P}}(\mathbf{z}, \theta)}{\partial \mathbf{z}} \Big|_{(\mathbf{z}=\mathbf{x}_{2M}(\boldsymbol{\alpha}), \theta=\theta_{2M})} \end{bmatrix}, \quad (D.23)$$

which is a block diagonal matrix. If \mathcal{P} is given by Eq. (D.6), one has $\partial \mathcal{P}(\mathbf{z})/\partial \mathbf{z} = \mathcal{J}_0^T(\mathbf{z})$, where \mathcal{J} is the tangent flow. At the *k*th iteration of the Newton's method, α_k is known, and the next set of coefficients is given by $\alpha_{k+1} = \alpha_k + \Delta \alpha$, where $\Delta \alpha$ is solution of

$$\left[\frac{\partial \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\boldsymbol{\alpha}))}{\partial \mathbf{X}(\boldsymbol{\alpha})}\frac{\partial \mathbf{X}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} - \frac{\partial \mathbf{Y}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}}\right]_{\boldsymbol{\alpha}=\boldsymbol{\alpha}_{k}} \Delta \boldsymbol{\alpha} = \mathbf{Y}(\boldsymbol{\alpha}_{k}) - \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\boldsymbol{\alpha}_{k})).$$
(D.24)

Notice that the dimension of the matrix $[\partial \mathbf{P}_{\mathbf{X}}(\mathbf{X}(\alpha))/\partial \mathbf{X}(\alpha)][\partial \mathbf{X}(\alpha)/\partial \alpha] - [\partial \mathbf{Y}(\alpha)/\partial \alpha]$ is $(2M+1)N \times (2M+1)N$. At each iteration of the Newton's method, a (2M+1)N dimensional linear system must be solved. Notice that the matrices $\partial \mathbf{X}(\alpha)/\partial \alpha$ and $\partial \mathbf{Y}(\alpha)/\partial \alpha$ are constant matrices with respect to the coefficients α . In the algorithm of the Newton's method, they are constant matrices in this case.

D.2.4.2 Autonomous map

We consider the map \mathcal{P} given by Eq. (D.6) does not depend explicitly in the parameter θ of the invariant curve, which is the case for the map (5.2) we consider in Chap. 5. This case is an extension of the non-autonomous case, i.e., additional conditions are required to compute the Fourier coefficients of the invariant curves. We consider

$$\overline{\mathbf{z}} = \boldsymbol{\mathcal{P}}(\mathbf{z}), \tag{D.25}$$

where $\mathcal{P} : \mathbb{R}^N \to \mathbb{R}^N$ and $\mathbf{z} \in \mathbb{R}^N$. In this case, the Fourier representation of the invariant curve is not unique. Indeed, let $\mathbf{x}(\theta; \boldsymbol{\alpha}) = \mathbf{y}(\theta + \phi; \boldsymbol{\alpha})$ with $\phi \in \mathbb{T}$. the invariant curves parametrized by θ have different Fourier series but represent the same curve, only the origin $\theta = 0$ is shifted in the θ -space. As a consequence, an infinite number of Fourier series represents the same curve. This implies some numerical difficulty to solve Eq. (D.24) at each iteration of the Newton's method. In order to overcome this difficulty, we fix arbitrarily one component of the origin of the invariant curve $\mathbf{x}_0(\boldsymbol{\alpha}) = \mathbf{x}(0; \boldsymbol{\alpha}) = \sum_{k=0}^{M} \mathbf{a}_k$. For instance, if the invariant curve surrounds a fixed point \mathbf{z}^* , we consider

$$F_{\mathbf{e}}(\boldsymbol{\alpha}) = \mathbf{e}^{\top}(\mathbf{x}_0(\boldsymbol{\alpha}) - \mathbf{z}^{\star}) = 0, \qquad (D.26)$$

where **e** is a unitary vector which depends on the considered dynamical system. In the autonomous case, the coefficients of the Fourier series of the invariant curves are also zeros of the function $F_{\mathbf{e}}$. The derivatives of this function with respect to the coefficients of the invariant curve $\boldsymbol{\alpha}$ are

$$rac{\partial F_{\mathbf{e}}(oldsymbol{lpha})}{\partial oldsymbol{lpha}} = [\underbrace{\mathbf{e}^{ op},\ldots,\mathbf{e}^{ op}}_{M+1 ext{ times}}, \underbrace{\mathbf{0}^{ op},\mathbf{0}^{ op}}_{M ext{ times}}].$$

This condition is added as an extra equation in the linear system for the computation of the invariant curve in the non-autonomous case [see Eq. (D.24)]. Adding an extra condition in the linear system implies the system becomes non-square, i.e., there are more conditions than unknowns. In order to solve the non-square linear system, we use a Gaussian elimination with maximal pivoting. In this way, the redundant equation is sent to the last row, and reads "0 = 0".

In addition, in the autonomous case, invariant curves come in family of invariant curves, labeled by different irrational frequencies ν . We consider a family of invariant curves surrounding a fixed point \mathbf{z}^* . For instance, in Chap. 5 this is the case for the fixed point of \mathcal{O}_S under the Poincaré map (5.2) (see Fig. 5.7). We use a *continuation method* to compute the invariant curves of the family. The frequency of each invariant curve ν is let free in the continuation method. Hence, $\mathbf{Y}(\boldsymbol{\alpha})$ in the non-autonomous case becomes $\mathbf{Y}(\boldsymbol{\alpha},\nu)$ in the

autonomous case, and as a consequence, $\mathbf{F}(\boldsymbol{\alpha})$ becomes $\mathbf{F}(\boldsymbol{\alpha}, \nu)$, and one has $\partial \mathbf{F}(\boldsymbol{\alpha}, \nu)/\partial \nu = -\partial \mathbf{Y}(\boldsymbol{\alpha}, \nu)/\partial \nu$. The derivative of $\mathbf{F}(\boldsymbol{\alpha}, \nu)$ with respect to ν reads

$$\frac{\partial \mathbf{F}(\boldsymbol{\alpha}, \nu)}{\partial \nu} = \sum_{k=1}^{M} k \begin{bmatrix} \mathbf{a}_k \sin[k(\theta_0 + \nu)] - \mathbf{b}_k \cos[k(\theta_0 + \nu)] \\ \vdots \\ \mathbf{a}_k \sin[k(\theta_{2M} + \nu)] - \mathbf{b}_k \cos[k(\theta_{2M} + \nu)] \end{bmatrix}.$$
(D.27)

The continuation method consists in computing the invariant curves one after the others. A parameter δ controls the distance between two invariant curves. We distinguish two cases. One case for which no invariant curves are computed yet. Only the fixed point is known. The other case for which at least one invariant curve of the family is known. The Fourier representation of the last invariant curve which has been computed is denoted $\tilde{\mathbf{x}}(\theta; \tilde{\boldsymbol{\alpha}})$, and its Fourier coefficients are denoted $\tilde{\boldsymbol{\alpha}}$. The coefficients of the new invariant curve $\boldsymbol{\alpha}$ are such that

$$F_{\delta}(\boldsymbol{\alpha}) = \begin{cases} |\mathbf{a}_1|^2 + |\mathbf{b}_1|^2 - \delta^2 = 0, \text{ if no torus is known yet,} \\ |\boldsymbol{\alpha} - \tilde{\boldsymbol{\alpha}}|^2 - \delta^2 = 0, & \text{ if at least one torus is known.} \end{cases}$$
(D.28)

The condition to compute the first invariant curve of the family is different than the condition to compute the other invariant curves of the family. The condition for computing the first invariant torus ensures the curves to converge far away from the fixed point. The derivatives of the function $F_{\delta}(\alpha)$ with respect to the Fourier coefficients is given by

$$\frac{\partial F_{\delta}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} = \begin{cases} [\mathbf{0}^{\top}, 2\mathbf{a}_{1}^{\top}, \underbrace{\mathbf{0}^{\top}, \dots, \mathbf{0}^{\top}}_{M-1 \text{ times}}, 2\mathbf{b}_{1}^{\top}, \underbrace{\mathbf{0}^{\top}, \dots, \mathbf{0}^{\top}}_{M-1 \text{ times}}], \text{ if no torus is known yet,} \\ 2(\boldsymbol{\alpha} - \tilde{\boldsymbol{\alpha}})^{\top}, & \text{ if at least one torus is known.} \end{cases}$$

In sum, in the autonomous case, to compute the family of invariant curves of a fixed point \mathbf{z}^* , a linear system of N(2M+1)+2 equations and N(2M+1)+1 unknowns must be solved. Among the N(2M+1)+2 equations, N(2M+1) equations correspond to the non-autonomous case given by Eq. (D.24), with the zero function $\mathbf{F}(\boldsymbol{\alpha},\nu)$ given by Eq. (D.20). The frequency of the invariant curve ν is considered as an unknown at each iteration, and Eq. (D.28) is used to maneuver along the family of invariant tori. One extra equation is used to fix the non-uniqueness of the Fourier representation of the invariant curve, and is given by Eq. (D.26). At the *k*th iteration of the Newton's method, for a given set of Fourier coefficients and frequency $[\boldsymbol{\alpha}_k; \nu_k]$, the new set of Fourier coefficients and frequency is given by $[\boldsymbol{\alpha}_{k+1}; \nu_{k+1}] = [\Delta \boldsymbol{\alpha}; \Delta \nu] + [\boldsymbol{\alpha}_k, \nu_k]$, where $[\Delta \boldsymbol{\alpha}; \Delta \nu]$ is solution of

$$\begin{bmatrix} \frac{\partial \mathbf{F}(\boldsymbol{\alpha}, \nu)}{\partial \boldsymbol{\alpha}} & \frac{\partial \mathbf{F}(\boldsymbol{\alpha}, \nu)}{\partial \nu} \\ \frac{\partial F_{\delta}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} & 0 \\ \frac{\partial F_{\mathbf{e}}(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} & 0 \end{bmatrix}_{(\boldsymbol{\alpha}=\boldsymbol{\alpha}_{k}, \nu=\nu_{k})} \begin{bmatrix} \Delta \boldsymbol{\alpha} \\ \Delta \nu \end{bmatrix} = - \begin{bmatrix} \mathbf{F}(\boldsymbol{\alpha}_{k}, \nu_{k}) \\ F_{\delta}(\boldsymbol{\alpha}_{k}) \\ F_{\mathbf{e}}(\boldsymbol{\alpha}_{k}) \end{bmatrix}.$$

D.2.4.3 Stability of the invariant curve

the method for computing the stability of an invariant curve is the same in the autonomous and the nonautonomous case. We denote $\Psi(\theta)$ the eigenvector and λ the associated eigenvalue of the invariant curve $\mathbf{x}(\theta)$. We consider a small displacement along the invariant curve $\delta \mathbf{x}(\theta)$. The invariance equation (D.17) becomes $\mathcal{P}(\mathbf{x}(\theta) + \delta \mathbf{x}(\theta)) = \mathbf{x}(\theta + \nu) + \delta \mathbf{x}(\theta + \nu)$. We perform a Taylor expansion on the left-hand side of the invariance equation, we obtain $\mathcal{P}(\mathbf{x}(\theta) + \delta \mathbf{x}(\theta)) \approx \mathcal{P}(\mathbf{x}(\theta)) + \mathbf{C}(\theta)\delta \mathbf{x}(\theta)$, where $\mathbf{C}(\theta) = \partial \mathcal{P}(\mathbf{z})/\partial \mathbf{z}|_{\mathbf{z}=\mathbf{x}(\theta)}$. The eigenvalue problem is (see Ref. [79])

$$\mathbf{C}(\theta)\boldsymbol{\Psi}(\theta) = \lambda\boldsymbol{\Psi}(\theta + \nu). \tag{D.29}$$

We consider the discrete value problem for 2M + 1 angles θ_j defined in Eq. (D.19). The Fourier representation of the eigenvectors is denoted

$$\Psi(\theta;\underline{\alpha}) = \mathbf{A}_0 + \sum_{k=1}^{M} \left[\mathbf{A}_k \cos(k\theta) + \mathbf{B}_k \sin(k\theta) \right],$$

where the coefficients of the eigenvector are $\underline{\alpha} = [\mathbf{A}_0, \dots, \mathbf{A}_M, \mathbf{B}_1, \dots, \mathbf{B}_M]^{\top}$. The rotation matrix of an angle ν is denoted \mathbf{H}_{ν} and is such that $\mathbf{H}_{\nu}\Psi(\theta; \underline{\alpha}) = \Psi(\theta + \nu; \underline{\alpha}) = \mathbf{A}_0 + \sum_{k=1}^M \{[\mathbf{A}_k \cos(k\nu) + \mathbf{B}_k \sin(k\nu)] \cos(k\theta) + \mathbf{B}_k \sin(k\nu)\}$

 $[-\mathbf{A}_k \sin(k\nu) + \mathbf{B}_k \cos(k\nu)] \sin(k\theta)\}.$ Therefore,

$$\mathbf{H}_{\nu} = \begin{bmatrix} \mathbf{I}_{N} & \mathbf{0}_{N} & \dots & \mathbf{0}_{N} & \mathbf{0}_{N} & \dots & \mathbf{0}_{N} \\ \mathbf{0}_{N} & \mathbf{I}_{N} \cos \nu & \dots & \mathbf{0}_{N} & \mathbf{I}_{N} \sin \nu & \dots & \mathbf{0}_{N} \\ \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{0}_{N} & \mathbf{0}_{N} & \dots & \mathbf{I}_{N} \cos(M\nu) & \mathbf{0}_{N} & \dots & \mathbf{I}_{N} \cos(M\nu) \\ \mathbf{0}_{N} & -\mathbf{I}_{N} \sin \nu & \dots & \mathbf{0}_{N} & \mathbf{I}_{N} \cos \nu & \dots & \mathbf{0}_{N} \\ \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{0}_{N} & \mathbf{0}_{N} & \dots & -\mathbf{I}_{N} \sin(M\nu) & \mathbf{0}_{N} & \dots & \mathbf{I}_{N} \cos(M\nu) \end{bmatrix}.$$

The matrix \mathbf{H}_{ν} is of dimension $(2M+1)N \times (2M+1)N$. In Fourier representation, the matrix $\mathbf{C}(\theta)$ reads

$$\mathbf{C} = \frac{\partial \mathbf{Z}(\mathcal{P}(\mathbf{X}(\alpha)))}{\partial \mathcal{P}(\mathbf{X}(\alpha))} \frac{\partial \mathcal{P}(\mathbf{X}(\alpha))}{\partial \mathbf{X}(\alpha)} \frac{\partial \mathbf{X}(\alpha)}{\partial \alpha}.$$
(D.30)

The two last terms on the right-hand side of the latter equation are given by Eq. (D.21) and Eq. (D.23). The first term on the right-hand side of the latter equation corresponds to the derivatives of the inverse Fourier transform with respect to the points in the Cartesian frame. They read

$$\frac{\partial \mathbf{Z}(\mathbf{X})}{\partial \mathbf{X}} = \frac{1}{2M+1} \begin{bmatrix} \mathbf{I}_N & \dots & \mathbf{I}_N \\ 2\mathbf{I}_N \cos(\theta_0) & \dots & 2\mathbf{I}_N \cos(\theta_{2M+1}) \\ \vdots & \ddots & \vdots \\ 2\mathbf{I}_N \cos(M\theta_0) & \dots & 2\mathbf{I}_N \cos(M\theta_{2M+1}) \\ 2\mathbf{I}_N \sin(\theta_0) & \dots & 2\mathbf{I}_N \sin(\theta_{2M+1}) \\ \vdots & \ddots & \vdots \\ 2\mathbf{I}_N \sin(M\theta_0) & \dots & 2\mathbf{I}_N \sin(M\theta_{2M+1}) \end{bmatrix}.$$
(D.31)

The latter matrix is of dimension $(2M+1)N \times (2M+1)N$. Finally, in Fourier representation, the eigenvalues λ and eigenvectors $\Psi(\theta; \underline{\alpha})$ of the invariant curve $\mathbf{x}(\theta; \alpha)$ are solution of the eigenvalue problem

$$\mathbf{H}_{\nu}^{\top} \mathbf{C} \boldsymbol{\Psi}(\boldsymbol{\theta}; \underline{\boldsymbol{\alpha}}) = \lambda \boldsymbol{\Psi}(\boldsymbol{\theta}; \underline{\boldsymbol{\alpha}}). \tag{D.32}$$

An example is shown in Fig. D.6 for a non-autonomous map. There are 2M + 1 eigenvalues. For invariant curves of Hamiltonian systems and M = 4, there are only two real eigenvalues. These two real eigenvalues correspond to the eigenvalues of the invariant curve. One is smaller than unity and corresponds to the eigenvalue associated with the stable direction of the invariant curve, and the other is greater than unity and corresponds to the unstable direction of the invariant curve.

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List of publications

List of publications in the frame of the thesis:

- S. A. Berman, C. Chandre, J. Dubois, M. Perin, T. Uzer, *Classical versus quantum views of intense laser pulse propagation in gases*, J. Phys. B: At. Mol. Opt. Phys. **52**, 125601 (2019).
- J. Dubois, S. A. Berman, C. Chandre, T. Uzer, *Inclusion of Coulomb effects in laser-atom interactions*, Phys. Rev. A **99**, 053405 (2019); featured in the Kaleidoscope images of Phys. Rev. A.
- J. Dubois, S. A. Berman, C. Chandre, T. Uzer, *Guiding-center motion for electrons in strong laser fields*, Phys. Rev. E **98**, 052219 (2018).
- S. A. Berman, C. Chandre, J. Dubois, F. Mauger, M. Perin, T. Uzer, Variational formulation of classical and quantum model for intense laser pulse propagation, Ann. Phys. **399**, 66 (2018).
- J. Dubois, S. A. Berman, C. Chandre, T. Uzer, *Capturing photoelectron motion with guiding centers*, Phys. Rev. Lett. **121**, 113202 (2018); featured in Physics Buzz (2018).
- S. A. Berman, J. Dubois, C. Chandre, M. Perin, T. Uzer, *Coherent buildup of high harmonic radiation:* the classical perspective, Phys. Rev. A 97, 061402(R) (2018).
- J. Dubois, S. A. Berman, C. Chandre, T. Uzer, Single and double ionization of magnesium by electron impact: A classical study, Phys. Rev. A 95, 022713 (2017); featured in the Kaleidoscope images of Phys. Rev. A.