Production and transport of long lifetime excited states in pre-equilibrium ion solid collisions[a]

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A complete experimental study on the production and transport of long lifetime excited states has been done for Ar^{18+} on solid C targets, at a velocity of 23 a.u. and for a range of thickness allowing to vary the transport conditions from single collision to equilibrium (3.5 to 201 μg/cm2). A systematic determination of Ar^{17+} Rydberg ℓ and 2s state populations has been performed using X-ray spectroscopy technique. Results are compared with predictions of different transport simulations (either developed on a quantum or classical phase space), which take into account multiple collisions and the strong polarization induced by the incoming ion (the wake field). Using Continuum Distorted Wave approximation for modeling the initial capture process, very good agreement is found between experimental Rydberg state populations and theoretical

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approaches limited to the effect of multiple collisions. On the contrary, the transport of the metastable 2s exhibits strong sensitivity to Stark mixing induced by the wake field. Limitation of each theoretical approach is discussed with respect to the different experimental observables.

34.50.Fa, 39.30.+w, 34.10.+x

I. INTRODUCTION

Researches on ion-matter interaction cover a large domain from the study of elementary physical processes involved in ion-atom collisions to the radiation damage in inert or biological materials. Beside some fundamental aspects, atomic collision studies are useful to understand the effects associated with energy deposition in matter[1]. In this frame, transport of electrons in excited states of fast ions through solids is a subject of current investigation. Indeed, a single electron bound to a nucleus allows to achieve experimentally a simply tunable system to probe directly the ion-solid interaction by a proper choice of the ion velocity v_P and the atomic number Z_P. Two specific effects of the solid media have been evidenced: the enhanced production of Rydberg states when compared to single ion atom collision, and the mixing of the internal states of the projectile by the wake field induced by the large charge of the projectile nucleus. So far, however, the production and transport of inner-shells and Rydberg projectile states were determined through experiments performed separately and the behavior of the state populations with respect to different transport processes were interpreted in each case by different theoretical approaches. This led to controversial interpretation of the transport effects of projectile electrons.

On the one hand, several authors observed high ℓ populations in Rydberg states. In their last studies, Betz and collaborators[2, 3] demonstrated that Coulomb capture of target electrons from the last layer of atoms in the solid (just when the ion exits into vacuum) could not explain the observed ℓ populations of Rydberg states. Provided target thickness is thin enough to avoid equilibrium in the state population, they shown that Rydberg states of fast ions are not formed predominantly by direct capture of target electrons. They found, instead, a connection between projectile core electrons and final Rydberg states. They invoked here strong contribution of ionized electrons (over all bound projectile electrons), which have large probabilities to undergo transitions into Rydberg states. On the contrary, results of ℓ Rydberg states populations (O^{2+} (v_p = 9 a.u.) on C, 20µg/cm² [4, 5]) have been interpreted as transport of projectile electrons with a classical Monte Carlo simulation taking into account properly multiple scattering effects. This classical model, pioneered by Burgdörfer and collaborators[6, 7], describes the motion of the projectile electron on a classical orbit perturbed by a stochastic force chosen to represent multiple scattering by the surrounding medium. However, in that case, the understanding of the projectile electron transport was far to be complete: i) target was thick enough to reach equilibrium for the charge state distribution and ii) capture of target electron and excitation or ionization of the 1*s* projectile initial state are of the same order of magnitude. Thus, different processes are involved in the "primary" production of excited states and part between "initial" excited state population and transport effects cannot be distinguished.

On the other hand, for inner-shell populations, extensive studies have been performed on charge state distribution, on *np* populations as well as on fine structure substate ($n\ell_j$) populations for several collisional systems[8]. A range of target thickness from single collision condition to equilibrium has been investigated. Different theoretical approaches have been developed and applied to explain the transport of core projectile excited states, which exhibit different degree of sensitivity to various processes experienced by the projectile electron in the solid. Namely, transport simulations account for multiple collisions, the strong target polarization induced by the incoming ion (the wake field), and the radiative decay inside the foil when it competes. Theoretical works have been carried out to determine the population of a given state at the exit of the foil. They are all based on the Liouville equation to describe the dynamical evolution of the ion during the interaction, with various degrees of approximations, classically or quantum mechanically. The Liouville equation is solved either directly as a matrix equation (the so called *master equations* approach)[9, 10] or by Monte Carlo simulations[9, 11, 12, 13, 14]. From a comparison with experiments, one can draw up an overview of the main results. Evolution of charge state distributions, involving only core state populations, is well reproduced by a model based on *master equations* taking into account only collisional processes (i.e. the *rate equations model*[15]). On the contrary, collisional approach cannot explain alone experimental data concerning transport of $n\ell$ inner-shells through solid foils. Combined effect of the wake field induced by the ion in the solid and multiple collisions has found to play a major role in explaining the evolution of *np* inner-shell populations (signed by the prompt deexcitation) with target thickness for different collisional systems: namely, Ar^{18+} at v_p = 23 a.u. on C [12 16]; Kr^{36+} at v_p = 35 a.u. on C and Cu [17 18]; Kr^{35+} at v_p = 47 a.u. on C and Al [9 12 14 19]. Furthermore, the behavior of the $3\ell_j$ populations of Kr^{35+} (for all cases mentioned above), as a function of ion transit time through the solid, has demonstrated very strong sensitivity to the effective value of the wake field. It even exhibits remaining oscillations of the induced Stark mixing between substates.

In this paper, we present an experiment where both Rydberg and core excited state populations of 23 a.u. Ar^{17+} ions after transmission through thin carbon foils have been measured. We focus on the presentation of long lifetime excited states, i.e. Rydberg and 2s states. For the first time, a complete analysis has been made over a wide range of target thickness (from 3.5 to 201 μg/cm^2) in order to vary extensively the number of collisions the projectile electron suffers in the solid (from ≈ 0.6 to 60 collisions). Moreover, absolute cross sections have been determined for X-ray production/incident ion of the Rydberg state decays and for the population of the 2*s* state. Great care was taken on the choice of the collisional system. Indeed, decisive questions remain: one concerns generally the effect of transport on the population of Rydberg states; the other is relevant to the sensitivity of the population of long lifetime states to the wake field during the transport. Therefore the system has been chosen to allow tests of transport effects, fulfilling the following criteria. First, using incoming Ar^{18+} ions, the primary production process of excited states - single capture - is well identified and multiple capture probabilities through successive collisions can be neglected even for the thickest target (probability ≤ 3%). Secondly, the transport of the initial excited states is mainly due to excitation and ionization processes. Finally, classical description of Ar *n* level is still valid down to quite low *n* value[5] and radiative decay inside the target itself can be neglected (only ≈ 4% of excited states -mainly 2*p* state- decay inside the thickest carbon target). This last remark suggests that a comparison of experimental data to classical approaches might be meaningful even for inner-shells. Therefore, as already discussed for the *np* states[12], quantum and classical transport theories can be applied as well for this particular system.

In the following, the paper is organized in 3 principal parts. In section II, we give a complete description of the experiment and we detail the results analysis. The different theoretical approaches are summarized in section III followed by a discussion on the comparison with the experimental data in section IV. The experimental precision reached allows to enlighten the validity limits of the various theoretical approximations of transport effects without the wake field (collisional models) or with the wake field included. Conclusions and perspectives are reviewed in the last section.

II. EXPERIMENT

Keeping in mind that lifetimes of hydrogen-like $n\ell$ states are proportional to $n^3\ell^2$ for a given projectile, the delayed photon emission observed in the decay of 2*p* and 3*p* states (which have very short lifetimes, i.e. 1.52 10^{-14} s for 2*p* and 5.12 10^{-14} s for 3*p*) comes from the Rydberg state population through cascade contribution. Therefore, the observation of the Lyman ($np \rightarrow 1s$) line intensities as a function of the ion time of flight behind the target is a direct signature of high angular momentum Rydberg state populations[2 3 4 20 21]. For indication, the respective contribution of direct and cascade populations of a given *np* state just at the exit of the foil are specified in Table I. These contributions have been estimated through the theoretical approaches described in section III. Whatever the approach used, contributions of cascade on *np* states are of the same order within 10%. Obviously, if instead of looking just at the exit of the foil, the Lyman line intensities are recorded at a given ion time of flight behind the target, the cascade contribution becomes larger. Beam-foil spectroscopy also allows to get information on the population of the 2*s* metastable excited state. This gives, here for the 2*s* state, a direct insight on the evolution of an inner-shell population during ion-solid interaction since cascade contribution is small (Table I). We have thus studied the evolution of the $2s \rightarrow 1s$ emission with target thickness i.e. the transit time of ions inside the solid. In the following, we detail experimental aspects since in this work *absolute* cross sections have been extracted.

A. Experimental set-up

The experiment has been performed on the SME facility (Sortie Moyenne Energie) at GANIL (Grand Accélérateur National d'Ions Lourds, Caen, France). The experimental set-up is schematically depicted in Figure 1. A beam of 13.6 MeV/u Ar^{18+} ions is directed onto thin carbon foils which can be accurately translated along the beam axis.

The delayed ($np \rightarrow 1s$) X-rays emitted by the projectile are analyzed at 90° with respect to the beam direction at different distances behind the foil by two Si(Li) detectors. Energies of the Ar^{17+} Lyman lines are given in Table II. The Si(Li) detectors are placed on the opposite side of the beam, facing each other. Decay lengths (along the beam axis) in the effective range from 0 to 50 mm, corresponding to delay times up to 1 ns, are explored with a 6 ps time resolution. To ensure this resolution a collimation system using slits of 60.4 mm long, 0.3 mm wide and 7 mm high is placed in front of each Si(Li) detector. In order to record two different spectra during a given acquisition time, each slit is shifted with respect to the centre of the collision chamber (i.e. beam axis origin) either by +3.3 mm or -3.3 mm (see Figure 1). Overlap measurements are also insured at a given distance from the foil by the two Si(Li) detectors. Despite the precision of the stepping motor used for target translation along the beam axis (1µm), the "0" position has been scanned for each foil to control the specific shadow effects entailed by the target holders as well as the X-ray auto-absorption, which depends upon the target flatness (see zoom in Figure 1). These two different effects are important and have to be taken into account when comparing absolute experimental values with theoretical predictions (see section III.C).

In addition, a Ge detector placed at 150° with respect to the beam axis recorded the whole emitted X-rays with the target located at the centre of the collision chamber. This has allowed to determine, for each target thickness, the cross section of Lyman line production mainly due to "prompt" emission as presented and discussed previously[12].

Beam intensity (in the 10 – 500 nAe range) is measured by a Faraday cup with a typical precision better than 1%. Seven self-supported carbon foils of thickness ranging from 3.5 ($\pm$27%) to 201 ($\pm$5%) $\mu g/cm^2$ have been used. The thickness as well as the impurity contents are measured by means of Rutherford-backscattering analysis with 2 MeV α particles produced by a Van de Graaff accelerator. We have selected targets where impurities contribute less than 15% to the total capture signal. The most frequent impurity is oxygen, with a relative content varying between 0.3 and 5%.

B. Detection

The intrinsic energy resolution of each Si(Li) detector used is found to be around 7% corresponding to line-widths of $\approx$ 230 eV at 3.273 keV. This allows to identify Ar^{17+} Lyα($2p\rightarrow 1s$), Lyβ($3p\rightarrow 1s$), Lyγ($4p\rightarrow 1s$) and the sum of remaining Lyman lines i.e. $\sum_5^\infty (np \rightarrow 1s)$, as well as the 2E1 decay mode of the 2*s* metastable state since its energy distribution is centered on 1.636 keV (theoretical value), well below the Lyα line. As it is well known, the 2*s* metastable state (with a lifetime of $3.5\ 10^{-9}$ s for Ar^{17+}) has two decay modes: a two-photon mode (2E1) and a single photon magnetic mode (M1) with branching ratios respectively of 97% and 3% in that case[22 23]. But, with Si(Li) detectors, it is obviously not possible to separate the two fine structure components of the Lyα line ($2p_{1/2}$-$2p_{3/2}$= 4.81 eV) and even less the M1 transition from the $2p\rightarrow 1s$ one (the Lamb Shift $2p_{1/2}$-$2s_{1/2}$= 0.158 eV [23]). Thus, the extraction of the 2E1 photon intensity, with a good accuracy, is imperative for both purposes. One is to determine the evolution of the 2*s* population with the target thickness (one task presented here). The other is to correctly subtract the M1 line contribution to the observed Lyman α' (Lyman α' = Lyman α + M1) emission and to determine the "pure" 2*p* state population, especially for large ion times of flight where the contribution of the 2*s* decay becomes important.

The use of Si(Li) at these low X-ray energies requires a detailed study of the efficiency and of the response function connected to the energy resolution together with a precise determination of the solid angle, in order to obtain meaningful *absolute* values of cross sections. Specific studies of the detection efficiency and the response function have been performed with different calibrated radioactive sources providing a large variety of X-ray lines over a wide range of energy. The efficiency for each Lyman line is given Table II. The collimators in front of each detector define a geometrical solid angle, which leads to a global transmission $T_{glob}^{n\ell}$ reported Table II. For the 2E1 decay mode, we determine the *mean* efficiency by the ratio between the integrals of the “experimental” and the well-known theoretical energy distribution[22] of this two-photon decay mode. The “experimental” energy distribution is obtained, as illustrated Figure 2, by taking into account the efficiency and the intrinsic resolution responses of a Si(Li) detector. This permits to go beyond an efficiency value and to get a complete knowledge of the experimental shape of the observed 2E1 decay mode, which is of importance for the data analysis as discussed in the following section.

C. Spectra analysis and extraction of experimental data

108 spectra recorded by the Si(Li) detectors have been analyzed. We present, Figure 3 as an example, three spectra recorded by one of the Si(Li) detectors at various delay times, *t*, and for a given carbon target thickness (3.5 μg/cm^2). We can distinguish the set of the different Lyman lines described above and the 2E1 photon emission. The Lyα' line decreases less rapidly with *t* than the other Lyman lines for two reasons. One is related to the fact that 2*p* state is more populated by very high $n\ell$ excited states (i.e. of long lifetime) through cascades than *np* states with $n \geq 3$. The second comes from the increased fraction of M1 in the Lyα' for large delay.

Spectra, as presented Figure 3, have been analyzed after subtraction of background, which has different behaviors depending on *t*. For *t* less than 9 ps, background has mainly two components. The first one is due to the well-known bremsstrahlung process that has been simulated using estimated cross sections[24] [25] [26] and taking into account the efficiency and the intrinsic resolution of detectors. The contribution of this component to the signal remains reasonably small. We found, for instance, a maximum contribution less than 1% for the Lyα' line and 2.5% for the Lyγ. The second background source comes from the interaction of the ion beam with the target holder (called beam-support background). To have an accurate estimation of its contribution, we have recorded spectra with the ion beam passing through a support without carbon foil. For $t < 9$ps, this contribution does not go beyond 0.5% for the less intense line (Lyγ line). For $t > 9$ps, only the beam-support background remains. It becomes critical when the signal to background ratio decreases (i.e. for large ion time of flight). For instance, at $t = 198$ ps, we got 35% of background contribution under the Ly γ line.

Sources of background subtracted, a complete knowledge of the X-ray line shapes is needed to determine the line intensities. The shape of the recorded 2E1 decay mode has been explained above (see Figure 2), but Lyman peak shapes depend also on the response function of Si(Li) detectors. In the 1-6 keV energy range, this response consists of a dominating gaussian peak, a low energy tail and a step-like function term[27 28]. A clear physical mechanism to explain this last term is not really yet known. Nevertheless, incomplete charge collection effects due to the dead layer of Si(Li) detectors explain the origin of the low energy exponential tail. Left exponential function with a constant width was used to simulate this tail. Its proportion with respect to the principal peak depends on experimental conditions and more particularly on counting rates, which were kept almost constant during the experiment. We find a proportion of 15% for the Lyα' and Lyβ lines. A precise determination of this low energy tail is required to extract an accurate 2E1 intensity, especially for short ion times of flight. The knowledge of all these line shapes allows to decompose spectra as illustrated in Figure 3. Knowing the counting rate of the 2E1 transition, N^{2E1}, N^{M1} is extracted using the well-known branching ratios and from there, $N^{Ly\alpha}$ of the "pure" Lyα line is deduced. The contribution of the M1 deexcitation mode to the Lyα' can be large. For example, a contribution from 10% for the 201 μg/cm^2 target up to 20% for the 3.5 μg/cm^2 target is reached for a delay time of 788 ps.

All the characteristics described above allow to obtain reliable emission intensities values, $I_{n\ell\rightarrow 1s}$. On the one hand, information on Rydberg ℓ state population is related to the evolution of the number of emitted photons per ion for a given $np\rightarrow 1s$ transition

($\widetilde{N}^{np}_{X/ion}$) as a function of the distance behind the solid target. For a certain distance, $\widetilde{N}^{np}_{X/ion}$ is determined using the following equation:

$$\widetilde{N}^{np}_{X/ion} = \frac{I_{np\to 1s}}{N_{proj} \times T^{n\ell}_{glob}} \tag{1}$$

where N_{proj} stands for the number of projectiles and $T^{n\ell}_{glob}$ for the global transmission.

On the other hand, the absolute values of the 2*s* state population per incident ion have been obtained from a spectrum recorded at a given distance *d* behind the target, using the following relation:

$$P_{2s/ion} = \frac{I_{2E1}(d)}{2R^{2E1}_{2s\to 1s} N_{proj} T^{n\ell}_{glob}} \left(\exp\left(\frac{-d}{\lambda}\right)\left[1-\exp\left(\frac{-L}{\lambda}\right)\right]\right)^{-1} \tag{2}$$

where $R^{2E1}_{2s\to 1s}$ is the branching ratio, λ the metastable 2*s* radiative decay length (173.6 mm for Ar^{17+} at $v_p = 23$ a.u.), and L the effective interception length (equal to 0.62 mm equivalent to a resolution time of 6 ps).

III. THEORETICAL APPROACHES FOR TRANSPORT DESCRIPTION

The choice of the collisional system (Ar^{18+} at $v_p = 23$ a.u. on C foils) allows to apply a classical simulation of projectile electron trajectories even for inner-shells (see section I). Therefore, we can confront the whole experimental results (Rydberg and inner-shell populations) with two kinds of treatments –classical and quantum- to interpret effects due to transport of hydrogen-like ions. We used and made specific improvements of the *master equations* approach as well as of the *classical Monte Carlo* method. The main hypotheses of these different approaches have been given elsewhere[8, 10, 12]. For that reason, we focus on the modifications and the specific treatments brought to these models in order to achieve comparison with the experimental data presented in this paper, in particular for the Rydberg states.

Prior to transport, one electron of the carbon foil is captured by the Ar^{18+} ion. This primary process is accounted for by using the Continuum Distorted Wave[29, 30] (CDW) approximation aware of some limitations in this choice[9]. Let us note that, the capture process enters as a constant term through the target in the different calculations presented below. This approximation implies to regard the source term as independent of the projectile population, which leads to an over-estimation limited to 4% of the 2*p* state population for the thickest target (the worst case).

A. Master and rate equations approach

To describe the transport of hydrogen-like projectile excited states, we use, on the one hand, the *master equations* approach[10, 19] to solve the *quantum* Liouville equation, which governs the time evolution of the projectile electron. This equation contains dissipative and non-dissipative terms. Dissipative terms illustrate the interaction with the environment and include multiple collisions and radiative decay, while non-dissipative terms describe the free time evolution of the electron in the screened Coulomb potential of the ion (i.e. the wake field effect). This quantum approach allows following the evolution of populations of each $n\ell_j m_j$ state and thus coherences between states. All the sub-states of the n = 1-6 shells are included in the treatment. Actually, convergence for calculations is reached around $n = 6$ (changes when going from $n_{max} = 5$ to 6 do not exceed 2% for the studied observable). The number of atomic cross sections one should calculate, for capture, ionization, intra-shell and inter-shell excitation processes (i.e. 4195 cross sections) and coherences terms (i.e. 245 terms) give indeed a limitation to this type of treatment. Therefore, the *master equations* approach is very suitable to interpret the inner-shell populations, in particular for np states up to $n = 4$ and for 2s. For the Rydberg state populations, where higher n values have to be considered, we have simplified the *master equations* to *rate equations*. In fact, neglecting the wake field effect, i.e. taking into account only the collisional processes and the radiative decay, the $n\ell_j m_j$ basis can be reduced to the $n\ell$ basis. In practice, additional $n\ell$ states can be thus easily introduced in the treatment. At the present time, the treatment involving all the cross sections of collisional processes (i.e. radiative decay, capture, ionization, intra ($\Delta n = 0$) and inter-shell ($\Delta n = \pm 1$) excitation) is complete up to $n = 10$. We have already demonstrated that the experimental delayed Lyman intensities for Ar^{17+} ions are, in fact, sensitive to all the excited states up to $n = 30$ even if only 3% of excited states have a principal quantum number $n > 10$ [16]. Hence, in order to overcome this intrinsic limitation, we assume a n^{-3}

law[31] for states with $n > 10$. The predictions of this *rate equations model* neglecting the wake field effect will be compared to the observed Rydberg state populations in section IV.A and section IV.B. The *master equations* approach will be compared with the evolution of the 2*s* state with target thickness (section IV.C).

B. Classical Monte Carlo methods

On the other hand, we simulate the transport of the projectile electron with a *classical Monte Carlo* method [7]. The use of a classical phase space is very suitable to interpret the Rydberg state populations since no limitation in the number of states involved has to be considered. In the present paper, we have especially studied within this approach, the influence of the wake field on ion transport. Indeed, this effect was found to have an important role in the evolution of prompt Lyman intensities as a function of target thickness due to its interplay with multiple scattering[12]. It was also shown that the dynamics of Rydberg states in the wake field is chaotic[32], which leads to a considerable enhancement of the “loss of population” (the so-called “cut-off”). For the system under consideration, this cut-off is excepted to occur for states above $n \approx 15$ and therefore, one should check the sensitivity of the delayed Lyman intensity to the wake field. The inclusion of the wake field in the calculation is technically a challenge. The numerical integration of electron trajectories is computationally much slower than for the pure Coulomb case. It is therefore difficult to achieve a meaningful statistical accuracy of photon intensity for long delay. Previous calculations[12] made use of an approximated uniform wake field, dependent on the Coulomb electron orbital energy, obtained from a dielectric response function based on local density approximation[9 33], but including only valence electrons. In order to have a consistent calculation with the ion slowing down involving also core electrons, we apply the Ashley dielectric function including carbon 1*s* excitation for both the collisional kernel and the wake field calculation. For electron-nuclei vectors ***r*** such like r is smaller than 10 a.u., we use the following expansion of Legendre polynomials $V_{wake}(\mathbf{r}) = \sum_{k=0}^{50} V_k(r) P_k(\cos\theta)$, where P_k is the Legendre polynomial of degree k and θ is the angle with respect to the beam axis. For larger radii we employ a direct numerical integration with Fast Fourrier Transform, which is more precise at larger distance but rather inaccurate at small r. To speed up the calculation, we exploit the secular approximation, which gives the evolution of the angular momentum ***L*** and the Lenz vector ***A***. We restrict its use for orbital energy E lower than E = -18 a.u., i.e. for orbits such like $r < 1$ a.u.. For these low energy orbits, we confine the spherical harmonic expansion to ℓ = 0, 1 and 2. We check that the Stark period is reproduced within 10% depending on the orbit orientation with respect to the projectile velocity $\boldsymbol{v_p}$. When averaged over several orientations, the Stark period is reproduced within a few percent. From only ***L*** and ***A***, the orbit shape evolution can be defined but not the electron position. This information is however of minor importance for our purpose, and a new electron position is obtained by randomly choosing a new eccentric anomaly in a uniform distribution. In the following, the predictions of the *classical approach* with and without the wake field (wake "on" and wake "off") are compared with the various experimental data on core and Rydberg state populations.

C. Inclusion of cascades and instrumental response

The models and the simulation of the post foil interaction cascade processes lead to the number of emitted photons per ion and per time unit for an $n\ell \rightarrow 1s$ transition:

$$\frac{dN^{n\ell}_{X/ion}(t)}{dt} = A^{1s}_{n\ell} U_{n\ell}(t) \,, \tag{4}$$

where $U_{n\ell}(t)$ is the fraction of ions with one electron in a $n\ell$ state and $A^{1s}_{n\ell} = \sum_{n'\ell'} A^{n'\ell'}_{n\ell}$ the $n\ell \rightarrow 1s$ transition probability per time unit. To compare with experiment, we have to keep in mind that the observed intensity $\tilde{N}^{n\ell}_{X/ion}(t)$ is integrated over the time interval determined by the width of the collimator placed in front of the Si(Li) detectors. $\tilde{N}^{n\ell}_{X/ion}(t)$ is then given by:

$$\tilde{N}^{n\ell}_{X/ion}(t) = \int_Z \frac{dN^{n\ell}_{X/ion}(Z+\Delta)}{dZ} \varepsilon_g(Z)\, dZ \tag{5}$$

with $$\frac{dN^{n\ell}_{X/ion}(Z)}{dZ} = \frac{1}{v_p} \frac{dN^{n\ell}_{X/ion}(t)}{dt} \tag{6}$$

Z is the position where $n\ell \rightarrow 1s$ transitions are recorded. The average shift Δ accounts for the X-ray auto-absorption due to the target itself, the target holder and eventually the target curvature. The geometrical efficiency function $\varepsilon_g(Z)$, accounting for specific shadow effects entailed to the target holders, is detailed in the Appendix.

IV. RESULTS AND DICUSSIONS

A. Rydberg state populations

1. Relative evolution with the target thickness

Figure 4 presents the evolution of delayed Lyman ($np \rightarrow 1s$) intensities, for different carbon target thickness, as a function of the distance d behind the foil corresponding to delay times longer than 20 ps. For sake of clarity, we normalized all data to the case of 3.5 µg/cm^2 target at a distance d = 1.05 mm (i.e. $t \approx$ 20 ps).

As found by several authors[4 20 21], the evolution of experimental photon intensities $\tilde{N}^{np}_{X/ion}(t)$ with t is well represented by a power law $\tilde{N}^{np}_{X/ion}(t) \propto t^{-a_{np}}$, where a_{np} is the decay slope of a given state. In these previous studies, performed for one single target thickness, it was observed that $a_{2p} < a_{3p}$. In the present work, decay slopes of n = 2, 3, 4 and $\sum_5^\infty (np \rightarrow 1s)$ are measured for a large range of target thickness –from single collision condition to equilibrium. We found $a_{2p} < a_{3p} < a_{4p} \approx a_{np(n>4)}$ for the whole rang of target thickness. This result simply accounts for cascade effects. Indeed, the cascade contribution decreases when the principal quantum number n increases, as shown Table I, and the less cascades contribute to the *np*-state population, the faster the decay.

Regarding now the *2p*- and *3p*-slope dependence on target thickness (i.e. on the number of collisions the projectile ion undergoes), departure of the experimental data from a binary ion-atom collision proves the sensitivity to transport effects. In particular, we found $a_{2p} = 1.69 \pm 0.13$ at $\delta = 3.5$ µg/cm^2 and $a_{2p} = 1.26 \pm 0.08$ at $\delta = 201$ µg/cm^2. The thickness dependence is smaller for 3*p*. For the $np \rightarrow 1s$ transitions with $n \geq 4$, the a_{np} values are constant within the error bars ($a_{4p} = 2.22 \pm 0.25$ and $a_{np(n>4)} = 1.88 \pm 0.17$), and no deviation from a binary ion-atom collision is observed. The observed decrease of the a_{2p} slope with target thickness shows evidence for high-ℓ Rydberg states population values, which develops during ion transport. In addition, as presented Figure 5, the evolution of a_{2p} and a_{3p} slopes versus foil thickness is well fitted by a decay law, $a_{np} = a_{np}^{sat} + a_0\, exp(-\delta / \delta_l\,)$ with a typical thickness δ_l of 6 µg/cm^2 (± 30%). This shows that a stationary regime is reached quite rapidly at approximately 20 µg/cm^2. In other words, equilibrium in high-ℓ Rydberg states population is established quickly, after typically 6 collisions (i.e. ion transit time in the solid of 2 fs).

Finally, Figures 4 and 5 show that classical transport simulations involving multiple scattering are in close agreement with experimental data. Note that only the “wake off” predictions are plotted, since, as we shall see below, the slopes are rather insensitive to the inclusion of the wake in the calculation.

2. *Absolute Lyman lines behavior with ion time of flight (t)*

Figures 6 to 10 present the *absolute* photon intensities $\tilde{N}^{np}_{X/ion}(t)$ as a function of the delay t for a sample of target thickness, namely 3.5, 8.6, 12.6, 42, and 98 μg/cm^2. In each figure, the experimental data are compared with the *rate equations model* and the *classical* simulation with or without the wake field included, respectively "wake on" and "wake off".

At a first stage, one can see that the three different types of calculation reproduce very well the observed behaviors while comparison addresses to *absolute* experimental measurements on one side and *ab initio* transport calculations on the other side. A more detailed analysis emphasizes different aspects relevant to weak or strong points of each theoretical approaches. Let us first examine the absolute Lyman lines intensities for $t < 9$ ps. In this domain of delay times, the shape of the curves is mostly governed by the geometrical effects detailed in the appendix (obviously affected by the "true" zero position of the target), while the intensity is sensitive to the x-ray auto-absorption. In that respect, for a given thickness, a same average shift of the target has been introduced in the three theoretical approaches. Reasonable values ranging from 5 to 250 μm have been used and allow to reproduce, for the first time, the evolution of the delayed Lyman lines in the "short" delay time region on an absolute scale. As shown Figures 6 to 10, on the one hand, predictions of the *rate equations model* are in very good agreement with the experimental data whatever the target thickness and for the whole Lyman series. On the other hand, the *classical Monte Carlo* simulation gives also realistic results, mainly for Lyman α and β. The very good agreement obtained with *the rate equations model* for short delay, is more likely due to the fact that Lyman lines correspond to prompt deexcitation of *p*-states coming mostly from low *n*-manifolds. In other words, the evolution of the Lyman lines intensities, in this restricted delay time domain, is not connected (and therefore not sensitive) to the Rydberg states population. Considering now the long delay times (i.e. $t > 9$ ps), where neither the geometrical effects nor the X-ray auto-absorption play any role, and, looking first at the evolution of Lyman α and β lines intensities, one can see Figures 6 to 10 that the *rate equations model* provides also very good predictions for targets thinner than 12.6 μg/cm^2. Indeed, for thicker targets, this model leads to an under-estimation of delayed Lyα and β intensities, in particular for long delay. This discrepancy could be ascribed to the empirical treatment used to include the contribution of excited states with $10 < n \leq 30$ (see section III.A), but may also be addressed to the neglected inter-shell excitation processes with $\Delta n > \pm 1$ for $n > 6$, which play a role for thick targets and become as high as those with $\Delta n = \pm 1$ for levels of high n value. For long delay times, the *classical transport* model "wake off" gives very good predictions over the whole range of target thickness for the Lyman β lines and slightly overestimates the delayed Lyα line intensities for target thickness higher than 42 μg/cm^2. Finally, over the whole range of target thickness and delay times, the Lyγ and $\sum_5^\infty (np \rightarrow 1s)$ are better predicted by the *rate equations* model than by its *classical* counterpart, which gives systematically higher values. For these states, the cascades come mainly from upper levels of low ℓ-values and their contribution remains small compared to direct population (see Table I). These lines are thus more sensitive to the evolution of inner-shell populations and a quantum treatment, like the *rate equations* model, is more suitable than a *classical* description.

To complete this detailed analysis, the influence of the wake field on Rydberg state populations has been examined within the *classical Monte Carlo transport* approach. As reported Figures 6 to 10, the comparison of the *classical simulation* with or without wake does not exhibit large differences. In particular, the shape of the curve for long delay remains unaffected and the slopes are similar. Regarding the curve behavior, the observed differences between both simulations are too small to be conclusive in spite of good statistics (statistical uncertainty in simulation is lower than 2% at 3.5 $\mu g/cm^2$ and lower than 12% at 98 $\mu g/cm^2$).

From this series of comparisons, we conclude that the *rate equations* model combined with an empirical inclusion of states with *n* above 10 is quite robust to reproduce the whole of experimental variations. The *classical simulation* is obviously much better suited to reproduce the long delayed Lyman intensities originated from high ℓ Rydberg states, even though it is less appropriate for the description of internal states, which provides the bulk of the contribution in the Lyman emission. In particular, the *classical simulation* enables to quantify the number of contributing highly excited Rydberg states in the long delayed Lyman intensities[16]. Furthermore, within this approach, a weak sensitivity of the delayed Lyman intensity to the wake field is found, at least for the system considered here.

3. *The 2s state population*

Figure 11 shows the evolution of the $2s_{1/2}$ state population with the carbon target thickness. Since cascade contribution, coming only from the decay of $\ell = 1$ excited states with $n \geq 3$, is rather small (see Table I), the evolution of this deeply bound *s*-state gives a direct insight of a pure core state population during the transport. As in the case of Rydberg states, departure of the experimental data from a binary ion-atom collision (dotted line in Figure 11) revealed a great sensitivity to transport effects. Simulations including only multiple scattering on the one hand (i.e. the *rate equations* model and the *classical transport* model "wake off") and accounting for both multiple scattering and the wake field influence on the other hand (i.e. the *master equations* model and the *classical transport* model "wake on") are also reported Figure 11.

When comparing with simulations including only multiple scattering (black lines in Figure 11), one can remark that the experimental 2*s* population departs from binary collisions much faster than predicted even though the general trend is quite well reproduces. Note that in this case, the *classical transport* approach gives similar results than the *rate equations* model up to a target thickness of around 20 μg/cm^2. From that point, the *rate equations* model saturates more rapidly to achieve a 40% lower population for the thickest target. To explain this difference between the two approaches, one may examine the effect of the re-capture process, which corresponds to the capture of an ionized electron by the ion. This re-capture process is accounted for in the *classical* approach and not in the *quantum* treatment (in the *rate equations* model, ionized electrons are lost). Nevertheless, the valuation of this process leads to a maximum difference of 20% in the capture population of highly excited states and is far to explain the extra factor of the *classical* approach compared to the *rate equations* model for the 2*s* population, keeping in mind that cascade contribution is there very small. Also, this discrepancy between both transport simulations is very probably due to some limitations in the classical treatment of the 2*s* state since it may be shown numerically[34] that *classical* simulation is not well suited to describe *s*-states transport in the presence of a Coulomb field, while it leads to better results for *p*-states.

The experimental evolution of the $2s_{1/2}$ population may be explained by the wake field influence. In fact, this dynamical screening acts directly on the excited levels of the projectile and induces, in particular, binding energy shifts as well as *mixing* of $n\ell j$ substates by Stark effect [9]. For our system, a substantial $2s_{1/2}$-$2p_{1/2}$ mixing can occur due to both the small Lamb shift in Ar^{17+} (0.158 eV) and the small difference in radiative widths (0.043 eV) compared to the Stark coupling element (0.49 eV). Such a ℓ-mixing is strongly supported by the fact that only a huge enhancement of intra-shell mixing cross sections in the *rate equations* model would be able to account for the observed effect on the prompt Lyman lines [16]. As reported Figure 11, this effect (see grey lines) plays a much stronger role within the quantum calculation (i.e. the *master equations* approach), but has a smaller impact in the case of the *classical* transport. Besides the limitation in the classical treatment, already discussed above, regarding the description of the *s*-states transport, any sensibility to the wake field means that the coherences of the initial sates may play a role and they can not be taken properly into account in a *classical* simulation. As it can be seen Figure 11, if the initial coherences are arbitrary set equal to zero in the *master equation* approach, a perfect matching with the *classical* treatment is reached but only for very thin targets and large differences between these two simulations occur during the transport (see dash-dot-dotted and full grey lines, Fig. 11). Finally, a proper account for CDW phases in the *master equation* approach leads hence to a perfect agreement with the experiment (dashed grey line)

V. CONCLUSION AND PERSPECTIVE

A complete study on the production and transport of long lifetime excited states has been done from the single collision condition to equilibrium. Using X-ray spectroscopy technique, evolution of the population of Ar^{17+} (v_p = 23 a.u.) Rydberg and 2*s* metastable states has been determined over a large range of carbon thickness. A special attention has been taken to extract, for the first time, *absolute* cross sections for X-ray production per incident ion of the Rydberg state decays and for the 2*s* state population. The collision system chosen enables to apply a classical simulation of projectile electron trajectories even for inner-shells. Therefore, the whole experimental results can be compared with the predictions of two different transport simulations developed either on a quantum or on a classical phase space. Using Continuum Distorted Wave approximation for modeling the initial capture process, both theoretical treatments account for transport effects through the multiple scattering contributions and the wake field influence.

The observed decrease of delayed Lyman line intensities (from 2*p*, 3*p*, 4*p* and $\sum_5^\infty np$ to the ground state 1*s*) with the ion time of flight *t* behind the target is well fitted by a power law characterized by decay slopes a_{np}. In particular, the evolution of the a_{2p} slope as a function of the target thickness shows evidence of a strong population in high ℓ Rydberg states, which develops during the transport and not just at the exit of the foil as claimed previously. Moreover, we have shown that equilibrium in such a Rydberg state population is established very quickly, starting from an ion transit time in the solid of 2 fs. For the first time, the behavior of *absolute* photon intensities $\tilde{N}_{X/ion}^{np}(t)$ with the delay *t* is reasonably well reproduced by the *ab initio* transport calculations we have developed (i.e. the *rate equations model* and the *classical* simulation). Comparisons with theory demonstrate that the evolution of the population of these highly excited states during the transport is mostly governed by multiple scattering. Moreover the *classical* approach is found to be more suited since $\tilde{N}_{X/ion}^{np}(t)$ has been found to be related to the population of excited states with a principal quantum number *n* up to *n* = 30.

The evolution of the deeply bound 2*s* state population with the target thickness, i.e. the ion transit time, is found also to be very sensitive to transport effects. Nevertheless, contrary to Rydberg states, the models accounting solely for multiple scattering cannot predict correctly the experimental evolution of the 2*s* population even if the general trend is obtained. This evolution exhibits a much stronger and faster $2s$-$2p$ mixing than predicted by collisional mixing. To achieve a complete experimental determination of this particular mixing, the $2p_{1/2}$ state population has to be examined within the same experiment. Recently, we have performed new high precision measurements to determine the evolution of the $2pj \rightarrow 1s_{1/2}$ transitions as well. Raw comparisons between the gaseous and solid targets show already an enhancement in the population of the $2p_{1/2}$ substate compared to the $2p_{3/2}$ component for solid targets (see Ref[35] for a complete analysis). This effect is consistent with our present observation on the evolution of the $2s$ population. Besides, the inclusion of the wake field effect in the transport models improves the predictions leading even to a perfect agreement with the experiment for the *quantum* treatment. The remaining discrepancy with the *classical* transport "wake on" is assigned to limitations of a classical treatment in describing transport of deeply bound *s*-states.

Comparisons between *absolute* measurements and *ab initio* simulations presented here allow to improve significantly our understanding of the ion transport in solids at high velocities.

APPENDIX

The observation of Lyman line intensities with detectors located at 90° with respect to the beam axis and collimated by slits demand to take into account two effects so as to calculate $\widetilde{N}^{n\ell}_{X/ion}(t)$ equation (5):

i) the observed intensity is integrated over Δt since we have used slits of 300 μm width,

ii) the solid angle is constant when the target is translated along the beam axis but not the geometrical efficiency $\varepsilon_g(Z)$. Indeed, the target holder entails shadow effects when it is in the intercepted region of detectors.

As mentioned in section II, two cases must be considered to evaluate $\varepsilon_g(Z)$. The first one is the case where the target holder is outside the intercepted region by the collimators. In this case, only delayed Lyman and 2E1 transitions are recorded. If the target holder is now inside the viewing region, one can still observe delayed Lyman lines but also prompt Lyman lines emitted close to the target. Each type of Lyman transitions has to be considered separately. Millimeter units will be used throughout this appendix.

We first examine the **case where the target holder is outside the intercepted region by the collimators**. $\varepsilon_g(Z)$ is then simply represented by the function depicted in Figure 12. Then we can easily deduce $\varepsilon_g(Z)$:

- $Z \geq 0.87$ $\qquad \varepsilon_g(Z) = 0$
- $0.55 \leq Z \leq 0.87$ $\qquad \varepsilon_g(Z) = \dfrac{0.87 - Z}{0.32}$
- $0.25 \leq Z \leq 0.55$ $\qquad \varepsilon_g(Z) = 1$
- $-0.07 \leq Z \leq 0.25$ $\qquad \varepsilon_g(Z) = \dfrac{Z + 0.07}{0.32}$
- $Z \leq -0.07$ $\qquad \varepsilon_g(Z) = 0$

For short ion times of flight, the **target holder is inside the intercepted region** (i.e., in our schemes, for $-0.07 \le Z \le 0.87$). This has two effects: the observation area of delayed Lyman lines is reduced and prompt Lyman lines emitted close to the target can now be observed with efficiency depending on the holder position.

For the delayed Lyman lines, 3 cases have to be considered:

a) *the target position (z) is in the domain* $0.55 \le z \le 0.87$ (see Figure 13). $\varepsilon_g(Z)$ is then given by the same equations than previously but where 0.87 is replaced by *z*.

b) *the target position is in the interval* $0.25 + e \le x \le 0.55$ (see Figure 14). In this case, the hidden area depends on the holder width, called ℓ. The parameter *e* is given by $e = \dfrac{0.55 - 0.25}{124.8} \times \dfrac{\ell}{2}$. Hence, we have:

- $Z \le -0.07$ or $Z \ge z$ $\qquad \varepsilon_g(Z) = 0$
- $z - \delta(z) \le Z \le z$, where $\delta(z) = \dfrac{0.55 - z}{124.8 - \ell/2} \times \ell/2$

$$\varepsilon_g(Z) = \frac{z - 0.25}{0.3} + \frac{0.55 - z}{0.3\,\delta(z)}(z - Z)$$

- $0.25 \le Z \le z - \delta(z)$ $\qquad \varepsilon_g(Z) = 1$
- $-0.07 \le Z \le 0.25$, $\qquad \varepsilon_g(Z) = \dfrac{Z + 0.07}{0.32}$

c) *the target position is in the interval -0.07 + e' ≤ x ≤ 0.25 + e* (see Figure 15). Here, the parameter *e*' is given by $e' = \frac{0.55 + 0.07}{124.8} \times \frac{\ell}{2}$. Finally, we have:

- $Z \leq -0.07$ or $Z \geq z - \delta(z)$ $\qquad \varepsilon_g(Z) = 0$
- $-0.07 \leq Z \leq z - \delta(z)$, $\qquad \varepsilon_g(Z) = \frac{Z + 0.07}{0.32}$

In the experimental conditions used here, prompt Lyman lines can be recorded only if $0.25 \leq z \leq 0.87$ (see Figure 16). In this case, the geometrical efficiency is only a function of *z*:

- $z \geq 0.87$ and $z \leq 0.25$ $\qquad \varepsilon_g(Z) = 0$
- $0.25 \leq z \leq 0.55$ $\qquad \varepsilon_g(Z) = \frac{z - 0.25}{0.30}$
- $0.55 \leq z \leq 0.87$: $\qquad \varepsilon_g(Z) = \frac{0.87 - z}{0.32}$

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TABLES

TABLE I. Respective contribution of direct and cascade population to the total population for different nℓ states and two target thickness. The direct contribution corresponds to the population at the exit of the foil. The cascade contribution corresponds to the population coming from upper levels by radiative decay behind the foil.

	3.5 μg/cm^2 target		98 μg/cm^2 target	
$n\ell$ state	% direct	% cascade	% direct	% cascade
2p	60	40	51	49
2s	74	26	84	16
3p	89	11	86	14
4p	≈ 100		99	1

TABLE II. Energy of each observed line with the associated efficiency and global transmission for one of the Si(Li) detector. The efficiency of the other Si(Li) detector corresponds to 70% of the values given here. E_{Proj} and E_{Lab} correspond respectively to energies in the projectile and laboratory frames. Energies of the Lyman line are taken from Ref[23]. "*" marks energy of the centre of the theoretical distribution of the 2E1 decay mode Ref [22].

Observed Lines	**Energy (eV)**		**Si(Li) detector**	
	E_{Proj}	E_{Lab} (θ_L=90°)	Efficiency	$T_{glob}^{n\ell}$
Lyα	3321	3273	0.83 (±7%)	$8.62\ 10^{-6}$ (±9.5%)
Lyβ	3936	3879	0.88 (±7%)	$9.20\ 10^{-6}$ (±9.5%)
Lyγ	4150	4091	0.90 (±7%)	$9.36\ 10^{-6}$ (±9.5%)
$\sum_5^\infty(\mathrm{np} \rightarrow 1\mathrm{s})$	4362	4299	0.91 (±7%)	$9.50\ 10^{-6}$ (±9.5%)
2E1	1660*	1636*	0.39 (±8%)	$4.01\ 10^{-6}$ (±10.5%)

FIGURES CAPTIONS

FIG. 1. Schematic representation of the experimental set-up. Effects leading to X-ray auto-absorption are displayed in a zoom (see text).

FIG. 2. Energy distribution of the 2E1 decay mode for an Ar^{17+} ion: the theoretical distribution (dotted line), the distribution corrected by the Si(Li) detector efficiency alone (dashed line) and the "experimental" distribution (solid line) i.e. the distribution corrected by the efficiency convoluted with the detector resolution. Si(Li) detector efficiency is also plotted (dash-dotted line) for sake of clarity.

FIG. 3. Spectra recorded by a Si(Li) detector after backgrounds subtraction at various ion time of flight (delay time) behind the 3.5 µg/cm^2 carbon target: (a) t = 0; (b) t = 108 ps; (c) t = 600 ps.

FIG. 4. Normalized evolution of the Lyman lines emission (see text) as a function of the distance behind the target: experimental data (symbols: ● target of 3.5 µg/cm^2, ■ 8.6 µg/cm^2, □ 98 µg/cm^2 and ○ 201 µg/cm^2), binary ion-atom conditions i.e. including cascade contribution without any transport effects accounted for (dotted curves), classical transport "wake off" (solid curves: target of 3.5 µg/cm^2, dashed dotted curves: 201 µg/cm^2). The real distance *d* behind the target has been arbitrary shifted by 0.55 mm to clarify the graph.

FIG. 5. Slope evolution of the delayed Lyman line intensities versus foil thickness; circles with error bars: experiment (symbols with error bars); classical transport (triangles); fit with a decay function given in the text (solid lines). The a_{3p} has been divided by 2 to clarify the graph.

FIG. 6. Evolution with the time of flight of the Ar^{17+} Lyman line intensities (i.e. number of emitted photon per ion) for the 3.5 µg/cm^2 carbon target: experimental results (symbols), classical transport model "wake off" (solid black lines) and "wake on" (solid grey lines), rate equation model (dashed lines). In the simulations, the CDW calculations have been used to account for the primary capture processes. The distance behind the target has been arbitrary shifted by 0.55 mm for sake of clarity.

FIG. 7. Same as Fig. 6 for the 8.6 μg/cm^2 target thickness.

FIG. 8. Same as Fig. 6 for the 12.6 μg/cm^2 target thickness.

FIG. 9. Same as Fig. 6 for the 42 μg/cm^2 target thickness.

FIG. 10. Same as Fig. 6 for the 98 μg/cm^2 target thickness.

FIG. 11. Absolute $2s_{1/2}$ populations of Ar^{17+} at $v_p = 23$ a.u. impact velocity as a function of carbon target thickness: experimental data (symbols); binary ion-atom conditions i.e. including cascade contribution without any transport effects accounted for (dotted black curve); classical transport model "wake off" (solid black line) and "wake on" (solid grey line); rate equations model (dashed black line); Master equations model with complete CDW calculations to describe the initial capture process (dashed grey line) and Master equations model with CDW calculations and with the phases of coherences of the initial states taken equal to zero (dash-dot-dotted grey line).

FIG. 12. Geometrical efficiency function of Si(Li) detectors for $z \geq 0.87$; z is the position of the target holder and Z the position where $n\ell \rightarrow 1s$ transitions are recorded.

FIG. 13. Geometrical efficiency function of Si(Li) detectors for $0.55 \leq z \leq 0.87$; z is the position of the target holder and Z the position where $n\ell \rightarrow 1s$ transitions are recorded. The hatched area is hidden by the support.

FIG. 14. Geometrical efficiency function of Si(Li) detectors for $0.25 + e \leq z \leq 0.55$; z is the position of the target holder and Z the position where $n\ell \rightarrow 1s$ transitions are recorded. The parameters e and $\delta(z)$ are defined in the text. The hatched area is hidden by the support.

FIG. 15. Geometrical efficiency function of Si(Li) detectors for $-0.07 + e' \leq z \leq 0.25 + e$; z is the position of the target holder and Z the position where $n\ell \rightarrow 1s$ transitions are recorded. The parameters e, e' and $\delta(z)$ are defined in the text. The hatched area is hidden by the support.

FIG. 16. Geometrical efficiency function of Si(Li) detectors for $0.25 \leq z \leq 0.87$; z is the position of the target holder and Z the position where $n\ell \rightarrow 1s$ transitions are recorded. The hatched area is hidden by the support.