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MULTIPHOTON IONIZATION OF ATOMS

G. Mainfray

Service de Physique des Atomes et des Surfaces, Centre d'Etudes Nucléaires de Saclay, 91191 Gif-sur-Yvette Cedex, France

Résumé - L'ionisation multiphotonique d'atomes à un électron, tel que l'hydrogène atomique et les alcalins, est actuellement un sujet accompli qu'on peut traiter par des modèles théoriques rigoureux. Cet article est consacré à l'analyse de l'ionisation multiphotonique d'atomes ayant plusieurs électrons sur la couche externe, et tout particulièrement les gaz rares. Il en résulte l'émission de plusieurs électrons, et la production d'ions multi-chargés. Dans le xénon, par exemple, on peut créer des ions jusqu'à Xe^{5+} . Les ions doublement chargés sont formés, soit par excitation simultanée de deux électrons de l'atome, soit par l'intermédiaire d'ions chargés une fois, selon l'éclairement et la fréquence du rayonnement laser utilisé. Les processus fondamentaux mis en jeu sont beaucoup plus complexes que dans le cas d'atomes à un électron. Un nouveau modèle théorique devra être élaboré, pour introduire les corrélations d'électrons. Enfin, la dernière partie de cet article est consacrée à une description succincte de la compétition qui apparaît à haute pression entre l'ionisation multiphotonique résonnante et la génération d'une fréquence harmonique impaire du laser utilisé.

Abstract - Multiphoton ionization of one-electron atoms, such as atomic hydrogen and alkaline atoms, is well understood and correctly described by rigorous theoretical models. The present paper will be devoted to collisionless multiphoton ionization of many-electron atoms as rare gases. It induces removal of several electrons and the production of multiply charged ions. Up to Xe^{5+} ions are produced in Xe atoms. Doubly charged ions can be produced, either by simultaneous excitation of two electrons, or by a stepwise process via singly charged ions. This depends on the laser intensity and the photon energy. The basic interaction processes involved are considerably more complicated than for one-electron atoms. A new theoretical model has to be developed to take into account electron correlation effects. Finally, the last part of the present paper will give a brief description of the competition which occurs at high atomic density between resonant multiphoton ionization and generation of odd harmonics of the laser radiation.

I - INTRODUCTION

Collisionless multiphoton ionization of atoms is a typical example of one of the new fields of investigation in atomic physics that lasers have opened up. The different aspects of the multiphoton ionization of one-electron atoms have been well understood these last few years. They can be correctly described by rigorous theoretical models in the framework of perturbation theory when only one electron is assumed to be involved in the ionization / 1-5 /. Alkaline atoms which have only one electron in the outer shell, and of course atomic hydrogen, are the examples that best satisfy this condition. For example the different aspects of multiphoton ionization of cesium atoms now form a well developed field. Accurate measurements of absolute values of the two, three and four-photon ionization cross sections of cesium atoms are in good agreement with calculated values, and clearly emphasize the validity of perturbation theory / 6 /. In the same way, there is also a good

agreement between theoretical and experimental results on resonance effects in multiphoton ionization of cesium atoms in the moderate laser intensity range, 10^7 - 10^9 W.cm⁻² / 7 /. Resonance effects emphasize the important role played by laser-induced atomic level shifts. Destructive interference effects, which give rise to minima in the multiphoton ionization cross sections of atoms, have been investigated with a high degree of accuracy in the two-photon ionization of Cs atoms / 8 /. This effect has been successfully used to check the validity of different calculation models. Furthermore, laser temporal-coherence effects induced by chaotic fields increase the non-resonant N-photon ionization rate by N!, in good agreement with theoretical calculations / 9 /. The resonance curves obtained by incoherent laser pulses are shifted and broadened with regard to those induced by coherent pulses with the same average intensity / 9 /. Finally absorption processes above the first ionization threshold have been investigated recently in Cs atoms at 1064 nm, in the 10^{10} - 10^{11} W.cm⁻² range. Cs atoms which can be ionized by the absorption of 4 photons can also absorb 5 photons. The ratio of the 5-photon to the 4-photon absorption rate, which is about 10^{-2} , is in good agreement with a calculation performed in the framework of the perturbation theory / 10 /.

New trends in multiphoton ionization are now : multiphoton ionization of atoms which have several electrons in the outer shell, multiphoton ionization and dissociation of molecules, and the competition of resonant multiphoton ionization with the generation of odd harmonics at high atomic density. Multiphoton processes in molecules will not be discussed here. The present paper will be devoted to collisionless multiphoton ionization of rare gases. This process induces the removal of several electrons and the production of multiply charged ions. The basic interaction processes involved are considerably more complicated than for one-electron atoms. A new theoretical model has to be developed to take into account electron correlation effects. Finally, the last part of the present paper will give a brief description of the situation when a competition occurs between resonant multiphoton ionization and a third harmonic generation.

2 - MULTIPLY CHARGED IONS PRODUCED BY MULTIPHOTON ABSORPTION IN RARE GAS ATOMS

2.1 - Experimental results

Recent experiments have emphasized the production of multiply charged ions by multiphoton absorption in rare gas atoms / 11-14 /. A mode-locked Nd-YAG laser is used to produce a 50-psec pulse which is amplified up to 5 GW at 1064 nm. The second harmonic can be generated at 532 nm up to 1.5 GW when needed. The laser pulse is focused into a vacuum chamber by an aspheric lens corrected for spherical aberrations. The vacuum chamber is pumped to 10^{-8} Torr and then filled with a spectroscopically pure rare gas at a static pressure of 5×10^{-5} Torr. At this pressure, no collisional ionization occurs, and no complications from charge exchange reactions are expected. Only collisionless multiphoton ionization occurs. The ions resulting from the laser interaction with the atoms in the focal volume are extracted with a transverse electric field of 1 kV cm^{-1} , separated by a 20 cm length time-of-flight spectrometer, and then detected in an electron multiplier. The laser intensity is adjusted in order to produce 1 to 10^5 ions. The experiment consists of the measurement of the number of ions corresponding to different charges formed as a function of the laser intensity.

2.1.1 - Multiphoton ionization of Xe at 532 nm

Fig. 1 is a typical result of the multiphoton ionization of Xe at 532 nm / 13 /. Up to Xe⁵⁺ ions are formed. Let us analyse the different processes which occur when the laser intensity I is increased. Fig. 1 can be divided into two parts. The first part ($I < 1.5 \times 10^{12}$ W.cm⁻²) is characterized by a laser-neutral atoms interaction, while in the second part ($I > 1.5 \times 10^{12}$ W.cm⁻²) a laser-ions interaction occurs. In the first part, the absorption of 6 photons by an atom leads to the removal of one electron and the formation of a Xe⁺ ion. This process appears in Fig. 1 through experimental points joined by a straight line with a slope 6 because the 6-photon ionization rate varies as I^6 . When the laser intensity

is increased further, approaching the I_S value, the absorption of 15 photons by an atom induces the simultaneous removal of two electrons and the production of a Xe^{2+} ion. This process appears in Fig. 1 through experimental points joined by a straight line with a slope 15. The 6-photon and 15-photon ionization processes deplete the number of atoms contained in the interaction volume. A marked change appears in the slope of the curves for both Xe^+ and Xe^{2+} ions beyond the laser intensity I_S . This saturation is a typical effect which occurs in multiphoton ionization experiments when all the atoms in the interaction volume are ionized. The intensity dependence of both curves of Xe^+ and Xe^{2+} ions just beyond I_S arises from ions produced in the expanding interaction volume when the laser intensity is increased further.

The second part of Fig. 1, for $I > 1.5 \times 10^{12} W.cm^{-2}$, describes the interaction of the laser radiation with ions, because the interaction volume is filled up with Xe^+ ions in place of atoms. A sudden increase in the number of Xe^{2+} ions occurs when the laser intensity is increased further. This comes from the absorption of 10 photons by a Xe^+ ion. This removes one electron from the Xe^+ and produces a Xe^{2+} ion. This appears in Fig. 1 through experimental points joined by a straight line with a slope 10. When the laser intensity is increased further, the 10-photon ionization of Xe^+ ions also saturates and Xe^{3+} , Xe^{4+} and Xe^{5+} ions are formed most likely through stepwise processes. This means Xe^{3+} ions are produced from Xe^{2+} ions by absorbing 14-photons. In the same way, Xe^{4+} ions are produced from Xe^{3+} ions by absorbing 20 photons, and likewise for Xe^{5+} ions. To sum up, Fig. 1 is a clear picture of the response of the electrons of Xe atoms to a high laser intensity. Each step of increased intensity gives rise to the removal of an additional electron.

It should be noted that the 10-photon ionization of Xe^+ ions which produces Xe^{2+} ions can be investigated in a well defined laser intensity range. The corresponding 10-photon ionization cross section is measured to be $10^{-297} \pm 1$. The uncertainty in this measurement comes mainly from the uncertainty of the laser intensity in absolute values. It should be recalled that a generalized N-photon absorption cross section is expressed in $cm^{2N} s^{N-1}$ units when the laser intensity is expressed in number of photons $cm^{-2} s^{-1}$. It is initially amazing to find that a multiphoton ionization cross section of a singly charged ion can be measured so easily while very few examples of measurements of photoionization cross section of ions are found in existing literature. Here the measurement is easy because it is the same laser pulse which produces both a clean Xe^+ ion target with a $10^{12} cm^{-3}$ density, and induces the 10-photon ionization of Xe^+ ions.

2.1.2 - Multiphoton ionization of Ne at 532 nm

Fig. 2 shows in a log-log plot the variation of the number of Ne^+ and Ne^{2+} ions produced as a function of the laser intensity /13/. The Ne^+ ion curve has a slope of ten which is characteristic of a non-resonant 10-photon ionization of Ne atoms. Ne^{2+} ions are produced in a laser intensity range far beyond the saturation intensity value I_S , that is when the interaction volume is filled up with Ne^+ ions and no longer with any Ne atoms. This requires that Ne^{2+} ions are produced through an 18-photon ionization of Ne^+ ions. This is confirmed by the slope $17 + 2$ measured on the Ne^{2+} ion curve. Here, the probability of production of Ne^{2+} ions by a simultaneous excitation of two electrons is much too low to be measured. An interesting comparison can be made between the 10-photon ionization of Ne atoms which produces Ne^+ ions and the 10-photon ionization of Xe^+ ions which produces Xe^{2+} ions. The atomic process requires a laser intensity 3.7 times higher than the ionic process, as shown in Figs. 1 and 2. If the two processes were compared at the same laser intensity, the 10-photon absorption rate in the spectrum of Xe^+ ion would be $(3.7)^{10} \sim 4.5 \times 10^5$ times higher than the 10-photon absorption rate in the spectrum of Ne atoms.

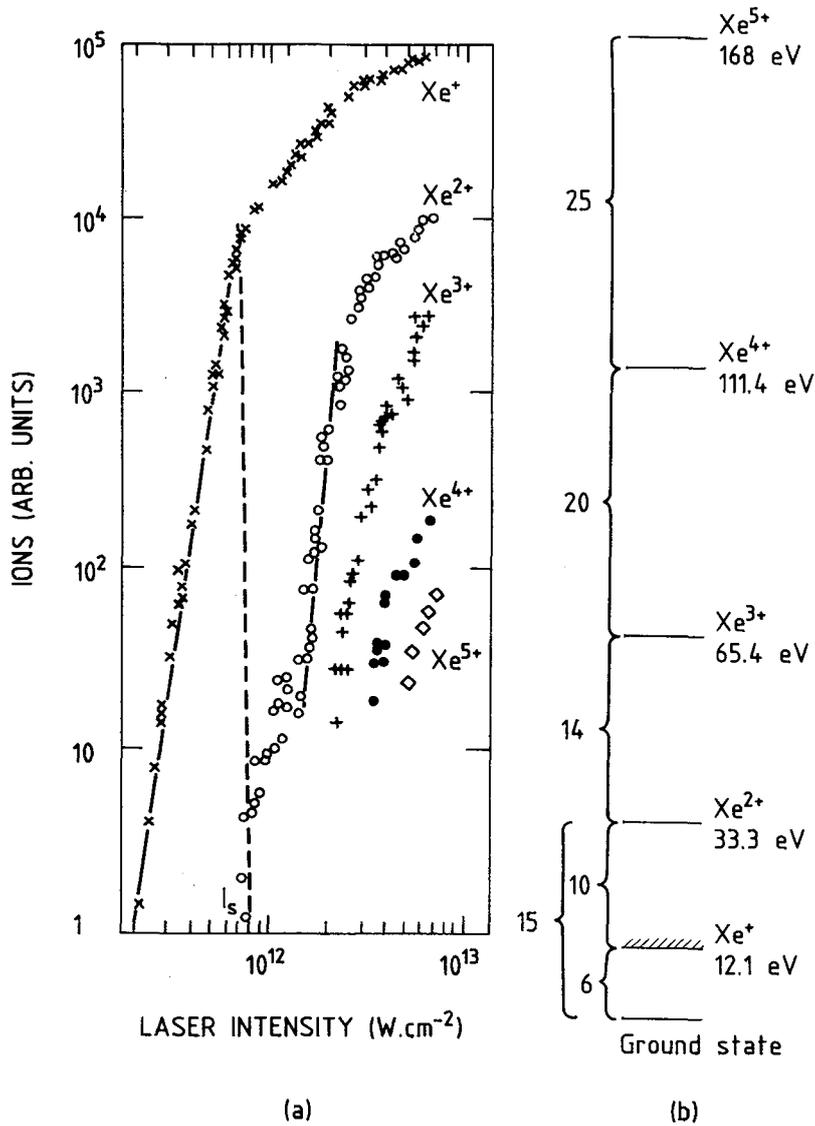


Fig. 1 - (a) A log-log plot of the variation in the number of Xenon ions formed at 532 nm as a function of the laser intensity. (b) Schematic representation of the number of photons involved in the production of multiply charged ions.

2.1.3 - Multiply charged ions produced in rare gases at 1064 nm

The production of multiply charged ions has also been investigated in the five rare gases at 1064 nm / 12 /. Let us consider here the two most different examples : Xe and He. Fig. 3 shows the variation of the number of Xe^+ , Xe^{2+} , Xe^{3+} , Xe^{4+} and Xe^{5+} ions as a function of the laser intensity. The general behavior is similar to that observed at 532 nm, except for two points. First, the two different processes

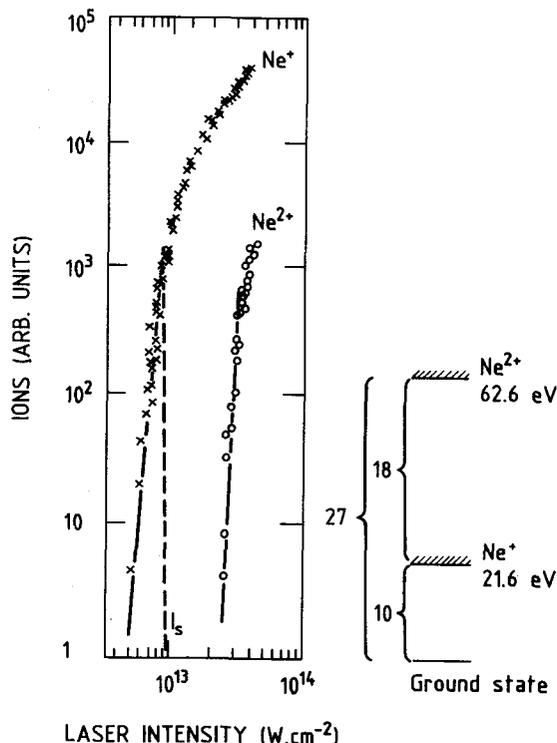


Fig. 2 - The laser intensity dependence of Ne^+ and Ne^{2+} ions formed at 532 nm.

of production of Xe^{2+} ions, namely the simultaneous two-electron removal from Xe atoms, and the one-electron removal from Xe^+ ions, are not so well separated than at 532 nm. Second, the probability of creating Xe^{2+} ions through a simultaneous two-electron removal from Xe atoms is 30 times larger here than at 532 nm, at the reference intensity I_S . At saturation intensity $I_S = 1.2 \times 10^{13} \text{ W.cm}^{-2}$ at 1064 nm the proportion of Xe^{2+} to Xe^+ ions is 1.5×10^{-2} , whereas it is only 5×10^{-4} at $I_S = 8 \times 10^{11} \text{ W.cm}^{-2}$ at 532 nm.

Fig. 4 shows the variation of the number of He^+ and He^{2+} ions produced as a function of the laser intensity. 68 photons at least have to be absorbed by He to produce He^{2+} ions which most probably comes from a simultaneous excitation of the two electrons. This conclusion is supported by the fact that saturation of both He^+ and He^{2+} ions occurs at the same laser intensity I.

2.2 - Discussion

A theoretical one-electron model has been used successfully to describe one electron removal in multiphoton ionization of atomic hydrogen and alkaline atoms in the past few years. However, such a model cannot be applied by merely extrapolating the lowest order perturbation theory to explain the production of multiply charged ions induced in many-electron atoms. In this respect, the following example of the production of doubly charged ions is very convincing. The one-electron removal in Xe atoms through 11-photon absorption at 1064 nm requires a laser intensity of $10^{13} \text{ W.cm}^{-2}$. The 29-photon absorption corresponding to the production of Xe^{2+} ions at 1064 nm would require a laser intensity of $10^{15} \text{ W.cm}^{-2}$, a value anticipated from the lowest order perturbation theory in the one-electron model. This is at variance

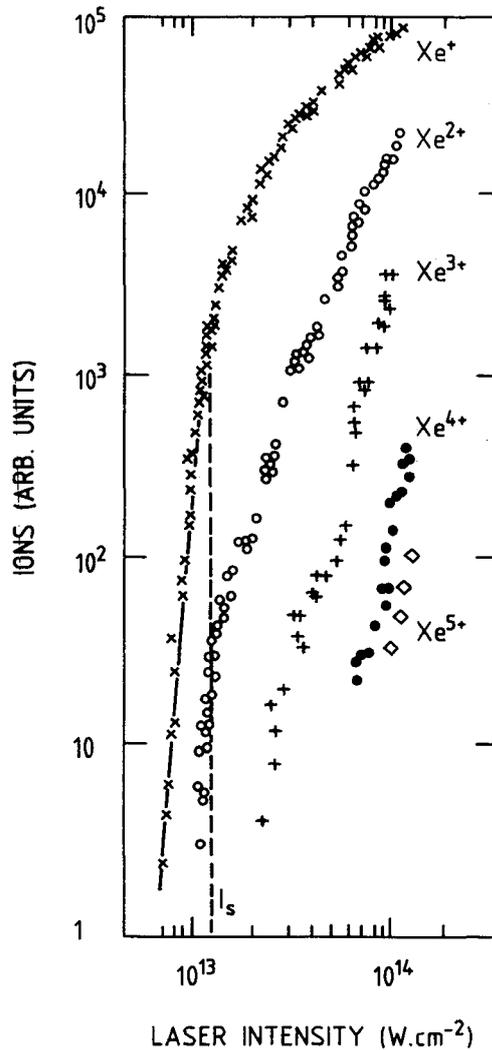


Fig. 3 - The laser intensity dependence of Xenon ions formed at 1064 nm.

with experimental results (Fig. 3) which show that a laser intensity of 1.5×10^{13} W.cm⁻² is enough to produce Xe²⁺ ions. Fig. 3 also shows in other terms that the 29-photon absorption rate giving Xe²⁺ ions is only 100 times less than the 11-photon absorption rate giving Xe⁺ ions at 1.5×10^{13} W.cm⁻².

The production of multiply charged ions through multiphoton absorption emphasizes both atomic properties and laser characteristics such as intensity, photon energy, pulse duration, etc ... Consequently the easy production of multiply charged ions could be explained either in terms of specific properties of many-electron atoms, or in terms of different mechanisms of absorption of photons in a continuum or in the spectrum of ions. Let us consider successively the two independent approaches.

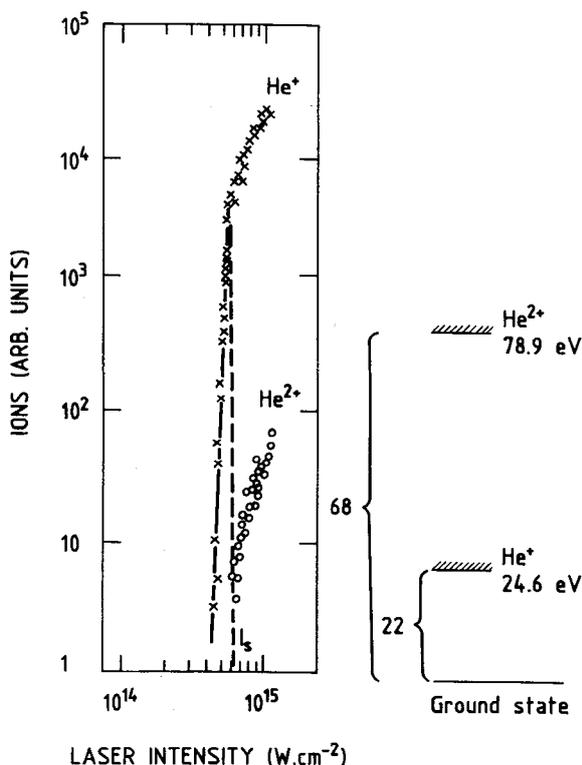


Fig. 4 - The laser intensity dependence of He^+ and He^{2+} ions formed at 1064 nm.

2.2.1 - Multiphoton absorption and laser characteristics

One of the most important questions that faces us is related to the mechanism of absorption of a very large number of photons, for example 68 photons at 1064 nm to produce He^{2+} ions. This problem could be somewhat similar to the multiphoton excitation or dissociation of large polyatomic molecules such as SF_6 by using CO_2 laser radiation. In such molecules, it is well known that 30 to 40 photons can be easily absorbed in the dense multitude of vibrational states. The quasicontinuum model postulates that due to coupling of the various normal modes of a large polyatomic molecule, there will always be quasi-resonant states available at each step of the excitation process. In the same way, the very large N-photon absorption rate giving rise to doubly charged ions at a high laser intensity could be explained in terms of a stepwise incoherent absorption process. At high intensity, one could describe the absorption in the continuum by the use of rate equations.

Some salient features can be drawn from experimental results. In a simplified way (Fig. 5), we have plotted the ionization threshold intensity required to produce singly and doubly charged ions in the five rare gases at 1064 nm, as a function of the number N of photons absorbed. By ionization threshold intensity, we mean the laser intensity which induces the production of a single ion and which is derived from original figures. Curve 5a is related to a one-electron removal from a neutral atom. Curve 5b is related to a one-electron removal from a singly charged ion, and curve 5c to a simultaneous two-electron removal from an atom. As a first observation, let us consider a fixed laser intensity, $1.8 \times 10^{13} \text{ W.cm}^{-2}$ for example, and

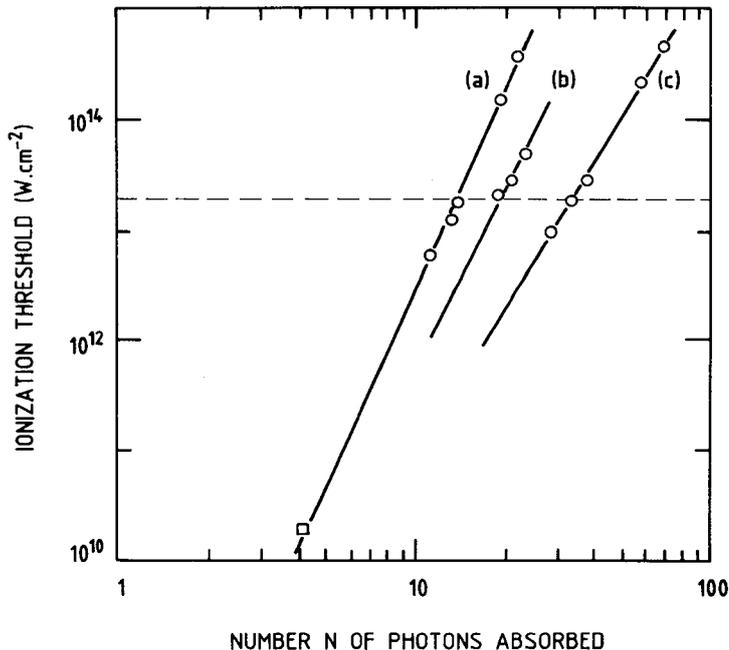


Fig. 5 - The ionization threshold intensity which induces the formation of one ion as a function of the number N of photons absorbed at 1064 nm. Curve (a) relates to a one-electron removal from an atom. (b) relates to a one-electron removal from a singly charged ion, and (c) to a simultaneous two-electron removal from an atom.

let us move along a horizontal line. At this intensity, one electron can be removed from Ar through a 14-photon absorption, one electron can be removed from Xe⁺ ion through a 19-photon absorption, and two electrons can be simultaneously removed from a Kr atom through a 34-photon absorption. In the same way, considerations based on a vertical line would show that the absorption rate of a large number of photons is higher in the discrete spectrum of an ion than in the discrete spectrum of an atom, and is still higher when a simultaneous two-electron removal of an atom occurs.

Fig. 5 also illustrates laser intensity effects through the three lines a, b, and c which diverge from each other as N increases or as the laser intensity increases. Let us give an example drawn from Fig. 5 by a direct comparison of experimental data shown in Figs. 1 and 4. In a moderate intensity of 10^{12} W.cm⁻² let us consider the one-electron removal from an Xe atom through a 6-photon absorption at 532 nm. The laser intensity has to be increased by a factor of 4 at the ion threshold detection to induce the simultaneous removal of two electrons from the Xe atom through a 15-photon absorption. In contrast, at a very high intensity of 6×10^{14} W.cm⁻², let us consider the one-electron removal from a He atom through a 22-photon absorption at 1064 nm. Increasing the laser intensity by 40 %, at the ion threshold detection, is enough to induce the simultaneous removal of the two electrons of He atom through a 68-photon absorption.

The photon energy seems to play an important role in the production of doubly charged ions through the simultaneous excitation of two electrons. For example, the probability of production of Xe²⁺ ions through a simultaneous two-electron removal from Xe atoms is 30 times less at 532 nm for 10^{12} W.cm⁻² than at 1064 nm for 10^{13}

W.cm^{-2} , as shown by a comparison of Figs. 1 and 3. This result can most likely be explained rather in terms of laser wavelength than in terms of laser intensity, as exemplified by Fig. 2. This figure shows that at 532 nm and at high laser intensity ($10^{13} \text{ W.cm}^{-2}$) no Ne^{2+} ions are produced by a simultaneous excitation of two electrons. Such a wavelength dependence in the simultaneous excitation of two electrons looks like the well known wavelength dependence in the photoionization cross section of Rydberg atoms. In contrast, the sequential production of doubly charged ions through singly charged ions does not seem to depend so strongly on the photon energy.

2.2.2 - Atomic properties

In multiphoton ionization of a many-electron atom, one can no longer consider the interaction of the laser radiation with a single electron, as one could for a one-electron atom. Firstly two electrons can be simultaneously excited and removed. Experimental results have shown that such a process is very sensitive to the laser wavelength, and is especially important when a long wavelength laser radiation is used. Secondly, electron correlation effects can lead to a collective response of the outer shell irradiated by an intense laser pulse. In a closed shell, several electrons could be excited while the first or two first electrons are removed from the shell. This could explain why an additional electron can be removed by increasing further the laser intensity by a small amount.

Fig. 6 shows the variation of the maximum ion charge states produced at 1064 nm and 193 nm in the five rare gases, as a function of the atomic number Z . The 193 nm data used here are derived from a recent work by Luk et al. / 15 /. Fig. 6 shows that He^{2+} and Ne^{2+} ions are produced at 1064 nm and not at 193 nm. This result could be explained by considering that He^{2+} and Ne^{2+} ions are produced at 1064 nm through a simultaneous excitation of two electrons. It was shown previously that such a process is very sensitive to the laser photon energy and could have probability too low at 193 nm to be observed. Furthermore, a large amount of energy can be transmitted to a many-electron atom through multiphoton absorption processes. For example, 250 eV when Xe^{6+} ions are produced.

As is well known, inner shells have to be considered, especially in Kr and Xe, in multiple photoionization by using Synchrotron Radiation at about 100 eV. Here, it is not possible to draw any conclusion on the possible contributions of an inner shell in the production of multiply charged ions at 1064 nm, or 532 nm. It could be assumed that one goes gradually from a situation close to multiple photoionization when high energy photons (5-10 eV) are used to produce multiply charged ions with consequently few photons absorbed at moderate laser intensity, to a very different situation when low energy photons (1 eV) are used with a very large number of photons absorbed by atoms at very high laser intensity. Generally speaking, we have to consider how inner shell contribution varies in the response of an atom which absorbs one 100 eV photon or 100 photons with 1 eV each.

To conclude this discussion, it should be pointed out that resonance effects in multiphoton ionization rates of one-electron atoms have been investigated in detail over the past few years / 10 /. These resonances are due to selective multiphoton excitation of an atomic state. An obvious question arises here. Is it possible, in the same way, to observe selective multiphoton excitation of high lying energy ionic states? If yes, is it possible to get a large population in a specific state inducing a population inversion and a stimulated emission in the extreme ultraviolet with a significant gain? Measurements of the electron energy spectrum and of possible extreme ultraviolet radiation will be performed in the production of multiply charged ions in the near future to answer the afore-mentioned question.

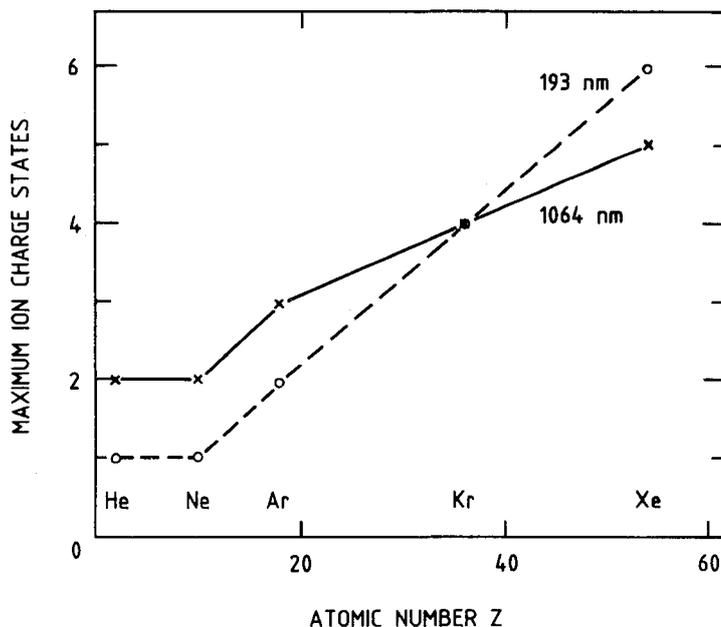


Fig. 6 - The maximum ion charge states produced in the five rare gases at 193 and 1064 nm as a function of the atomic number Z .

2.3 - Additional data

2.3.1 - The production of doubly charged ions and the production of energetic electrons

A many-electron atom irradiated by an intense laser pulse can absorb a very large number of photons, where "very large" should be understood by comparison to the minimum number of photons required to reach the first ionization threshold. For example, the Xenon atom should absorb at least 29-photons to produce Xe^{2+} ions at 1064 nm, at least 57 photons to produce Xe^{3+} ions ... , while the absorption of 11-photons is enough to reach the first ionization threshold. The excess energy, compared to single ionization energy, is used to remove additional electrons and to produce multiply charged ions. An excess energy can also be used to generate energetic electrons, as shown in previous experiments / 16-19 /. The energy spectrum of the photoelectrons produced by absorption of photons above the first ionization threshold shows a series of peaks evenly spaced by an amount equal to the photon energy. These experiments have been performed mainly on Xe at 1064 and 532 nm. At 1064 nm, Xe^{2+} ions produced by simultaneous excitation of two electrons of Xe atoms as shown in Fig. 3, and the absorption of about ten additional photons above the first ionization threshold occur simultaneously at the same laser intensity just below the saturation intensity I_S . Consequently, an obvious question arises. Are these two processes independent or not? A unified theoretical model should be developed if these two processes are not independent.

2.3.2 - Doubly charged ions produced by multiphoton ionization in alkaline-earth atoms

Previous experiments on the multiphoton ionization of alkaline earth atoms have emphasized the production of doubly charged ions / 20, 21 /. A striking similarity

can be seen between Fig. 3 in Xe in the present paper and Fig. 2 of Feldmann et al. / 21 / on Sr atoms, both obtained at 1064 nm. In both cases, doubly charged ions appear before the saturation of singly charged ions. This supports the assumption of a production of Sr^{2+} ions through the simultaneous excitation and removal of the two electrons of the outer shell. Experimental results obtained in Sr also emphasized that the simultaneous excitation of the two electrons is very sensitive to the photon energy. For example at 532 nm, Sr^{2+} ions are produced in a laser intensity range far beyond the saturation intensity, that is when the interaction volume is filled up with Sr^+ ions and no longer with Sr atoms. This requires that Sr^{2+} ions are produced solely through a multiphoton ionization of Sr^+ ions at 532 nm. In consequence, the probability of the simultaneous excitation of the two electrons decreases dramatically when the laser wavelength decreases by a factor of two. In conclusion, a striking similarity is observed in the production of doubly charged ions by multiphoton absorption in rare gases and alkaline earth atoms. Other atoms are expected to behave similarly.

3 - MULTIPHOTON IONIZATION AND ODD HARMONIC GENERATION

Collisionless multiphoton ionization of atoms has been well described by the perturbation theory when atomic density is low enough so that atoms can be considered to interact independently with the laser field. However, the description of multiphoton ionization is more complex when the atomic density is increased so much that odd harmonics are generated, and consequently should be included in theoretical treatment of multiphoton ionization.

Recent experiments performed in Xenon / 22, 23 / and mercury / 24 / clearly exhibit that a third harmonic wave can be produced in five-photon ionization when a three-photon resonance takes place on the 6s state in Xe or the $6p\ ^1P_1$ state in Hg. Third harmonic generation strongly affects the ionization rate. Fig. 7(a) is a scheme for five-photon ionization in Hg near a three-photon resonance on the $6p\ ^1P_1$ state. A sample result is shown in Fig. 7(b). The global ion production, which should be linear versus pressure at low pressure, saturates and then decreases. Conversely, a VUV emission corresponding to the third generation is observed in the forward direction at a pressure as low as 10^{-4} Torr. In Xenon, a disappearance of the ion signal has even been observed when the pressure is increased beyond 1 Torr. Furthermore, the resonant ionization profile and the line shape of the third harmonic generation progressively broaden and are blue shifted as the pressure is increased. Different theoretical models have been developed to explain the observed behaviour / 25-30 /. Briefly, experimental results are explained in terms of an interference between two different coherent channels to the resonant state : the absorption of three laser photons and absorption of one third harmonic photon. These two amplitudes can exactly cancel each other. The resonant level is no longer populated, and the ionization signal disappears. In each model, collisional effects are neglected because they make a negligible contribution over the range of pressures relevant to these experiments.

The example which has been described should not be considered as a rule. The pressure at which third harmonic generation becomes an important channel to include in the ionization process depends strongly on physical conditions. In a previous experiment the four-photon ionization of mercury atoms through the resonant three-photon excitation of the $7p\ ^1P_1$ state has been investigated / 24 /. In this experiment, no third harmonic generation has been observed when the Hg pressure is increased, even at 10^{-1} Torr, and the ionization signal does not depart from a linear pressure dependence. This could be because the resonant $7p\ ^1P_1$ state is much more strongly coupled (by a one-photon process) to the continuum than to the ground state. This strong coupling of the resonant state to the continuum does not favour the generation of a third harmonic.

In conclusion, multiphoton ionization of atoms has been mainly investigated so far at low gas pressures less than 10^{-4} Torr, while nonlinear optics have been investigated at pressures typically higher than 10 Torr. An overlap between multiphoton

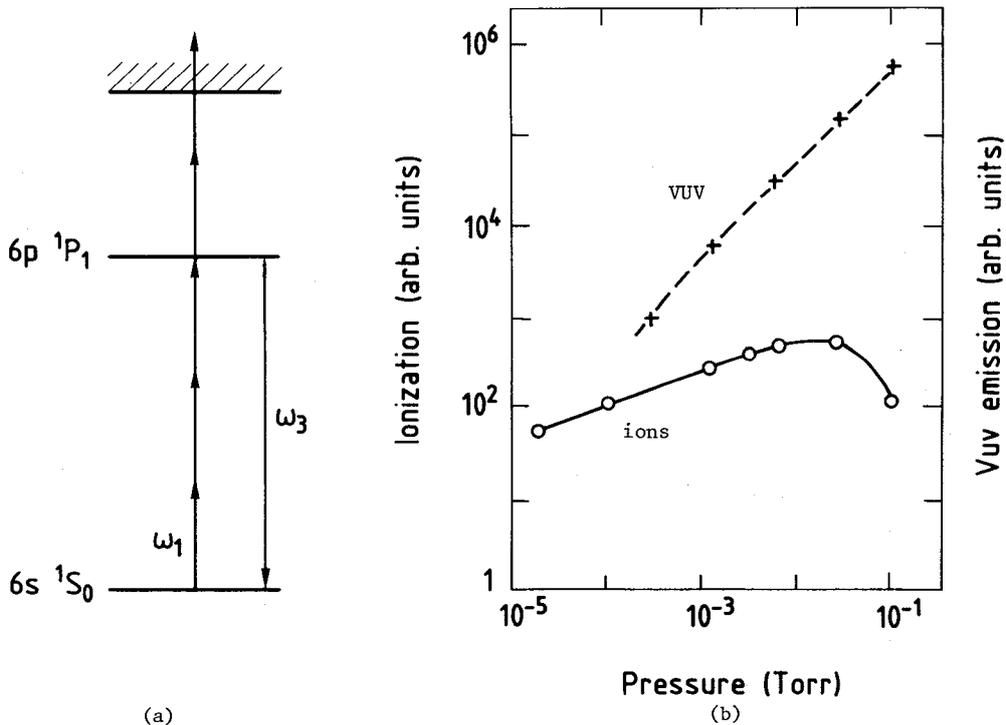


Fig. 7 - (a) Schematic representation of the five-photon ionization of Hg through the three-photon excitation of the $6p \ 1P_1$ state. (b) The variation of the ionization signal and the VUV emission (185 nm) corresponding to third harmonic of the laser wavelength (555 nm), as a function of Hg pressure.

ionization and odd harmonics generation can occur in the broad intermediate pressure range (10^{-4} to 10 Torr). The pressure limit which determines a "pure" multiphoton ionization process strongly depends on physical conditions.

4 - CONCLUSION

The interaction of an intense laser radiation with a many-electron atom is one of the few main topics which remains to be investigated in atomic physics. It is quite an open field and is of great interest from a basic point of view. It leads to the challenge of calculating multiphoton ionization rates for a many-electron atom. The theoretical one-electron model which has been used successfully to describe multiphoton ionization of atomic hydrogen and alkaline atoms in the past few years cannot be applied to describe the production of multiply charged ions induced by a collective response of an atomic shell irradiated by an intense laser pulse. A simultaneous excitation of many electrons of the outer shell could occur. Another model has to be developed to take into account electron correlation effects and possible electron rearrangement during the multiphoton absorption. Furthermore, the process of absorption of a very large number of photons by a many-electron atom is not yet understood and has yet to be investigated.

Recent experiments described in the present paper have been performed using an ion detector. These experiments will be completed in the near future, first by measurement of energy spectrum of electrons generated in the production of multiply

charged ions, and second by fluorescence measurements. Both methods are expected to give very valuable information on the internal energy states of the ions produced. An increase in the data concerning atomic and ionic spectroscopic behaviour can be expected. Finally, one of the most important questions is related to the possibility of a selective multiphoton excitation of a high lying energy ionic state, by tuning the laser wavelength. At high atomic density, it would then be possible to get a large population in a specific ionic state inducing a population inversion, and an extreme ultraviolet generation.

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