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Multiply Charged Ions Formed by Multiphoton Absorption Processes in the Continuum

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Singly, doubly, triply, and quadruply charged krypton ions are formed by multiphoton absorption processes in krypton atoms. They are induced by a 50-psec laser pulse at 1.064 μm in the 10^{13}–10^{14}-W-cm^{-2} intensity range. The percentage ratio between the numbers of doubly and singly ionized Kr atoms is 10% at 8×10^{13} W cm^{-2}. It is shown that Kr^{2+} ions result from a direct 33-photon absorption process. At 1.5×10^{13} W cm^{-2}, the 33-photon absorption rate is only about 60 times less than the 13-photon absorption rate which gives singly charged ions.

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Multiphoton ionization processes are generally described by the lowest-order perturbation theory. In this picture an atom with an ionization energy E_i is ionized by absorbing N photons with energy $h\nu$. The number N is defined as the first integer exceeding the ratio $E_i/h\nu$. However, if an atom can be ionized through an N-photon absorption process, it can also be ionized through the absorption of $N+1, N+2, \ldots$ photons of the same energy. Such processes are present in every multiphoton ionization, but being of a higher order, they require higher laser intensities to be observed. The absorption of one additional photon was observed for the first time in an experiment on six-photon ionization of xenon by measuring the energy spectrum of electrons produced in the ionization.[1] More recently, two experiments on eleven-photon ionization of xenon exhibited absorption which exceeded the minimum number eleven by seven photons[2] and ten photons.[3] The electron energy spectrum consists of series of peaks evenly spaced in photon energy. These experiments showed that ionization must be described in terms of competing N-photon, $(N+1)$-photon, $(N+2)$-photon, ... transitions from the ground state of atoms up to different final states above the first ionization limit. These measurements suggest the possible generation of doubly charged ions when the laser intensity is sufficiently high.

In this Letter, some preliminary results on the first observation of multiply charged ions produced through the multiphoton ionization of krypton atoms are presented. The laser used in the present experiment has been described elsewhere.[4] It is a mode-locked Nd-doped–yttrium-aluminum-garnet oscillator using a Kodak 9740 saturable dye. A single pulse is isolated from the train by the well-known Pockels switch method. This single pulse is amplified by two Nd-glass preamplifiers and then passes through a spatial filter before entering a three-stage Nd-glass amplifier. The laser radiation is linearly polarized and is centered about 10 643.5 Å with a linewidth of 0.5 Å. Pulse characteristics are measured using diagnostics described in previous papers.[5,6] The laser pulse is bandwidth limited

and has a duration of about 50 psec. Fluctuations in pulse duration are controlled by measuring the spectral distribution associated with each laser shot. This procedure is much easier than using a streak camera.

Multiphoton ionization of krypton atoms was realized with use of an experimental arrangement similar to that employed in previous work,[5,7] a modification being the use of a time-of-flight spectrometer to separate the multiply charged ions. Briefly, the laser pulse is focused into a vacuum chamber by an $f/3$ aspheric lens corrected for spherical aberrations. The vacuum chamber is pumped down to 10^{-8} Torr and then filled with spectroscopically pure Kr (5×10^{-5} Torr). At this pressure, no complications from charge-exchange reactions are expected. The ions resulting from the laser interaction with the atoms in the focal volume are extracted with a transverse electric field of 1000 V cm^{-1}, separated by a time-of-flight spectrometer and then detected in an electron multiplier. The time-of-flight system has a length of 20 cm. Its resolution is quite adequate to resolve the Kr^+, Kr^{2+}, Kr^{3+}, and Kr^{4+} peaks as shown in Fig. 1.

Figure 2 is a log-log plot of the variations in the numbers of Kr^+, Kr^{2+}, Kr^{3+}, and Kr^{4+} ions generated as a function of the laser intensity I. Experimental results are displayed on an expanded logarithmic scale along the laser intensity axis to achieve higher accuracy, especially for the most interesting part of the curve between 0.8 and 1.5×10^{13} W cm^{-2}. As far as Kr^+ ions are concerned, the slope $\partial \log N_i/\partial \log I = 13\pm0.5$ corresponds to the number $N_1 = 13$ defined as the next integer above the first ionization energy of the krypton atom (13.99 eV) divided by the laser photon energy (1.165 eV). It is a typical result for a N_1-photon ionization process. When the laser intensity I is increased beyond the I_S value, there is a marked change in the law of variation for the number of Kr^+ ions as a function of laser intensity. This saturation is also a typical effect which occurs in multiphoton ionization when all the atoms contained in the interaction volume are ionized.[8] The $I^{3/2}$ intensity law beyond the I_S value arises from ions formed in the expanding interaction volume. As far as Kr^{2+} ions are concerned, the slope $\partial \log N_i/\partial \log I = 32\pm5$ before the saturation intensity I_S. This value is so large that it cannot be measured very accurately. However, it is in good agreement with the number $N_2 = 33$ defined as the next integer above the ratio $E_2/h\nu$, where E_2 is the second ionization energy of the krypton atom, 38.35 eV, deduced from the ground state of the atom. Furthermore, a saturation occurs at the same laser intensity $I_S = 1.5\times10^{13}$ W cm^{-2} as for Kr^+ ions.

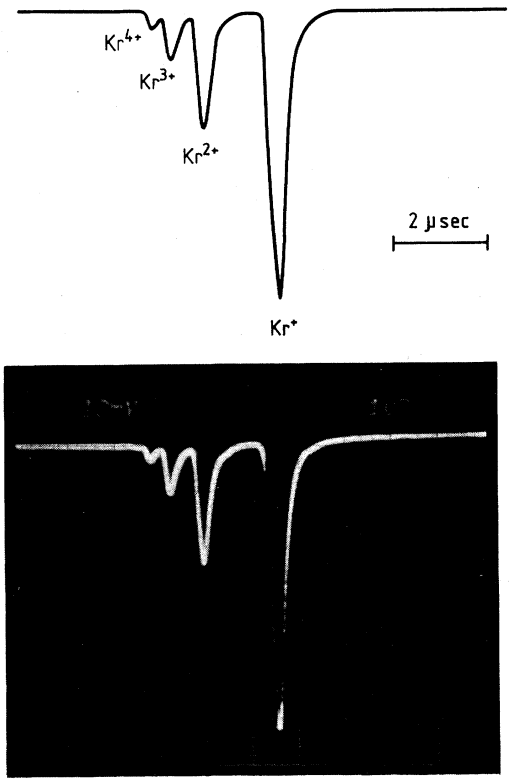

FIG. 1. Time-of-flight spectrum showing, from right to left, Kr^+, Kr^{2+}, Kr^{3+}, and Kr^{4+} ions observed at 10^{14} W cm^{-2}. The oscilloscope photograph has scales of 10 mV and 1 μsec per division.

At first sight, two processes could be responsible for the formation of doubly charged ions: (1) a direct process in which the Kr^{2+} ion is formed from the ground state of the Kr atom. Thirty-three photons are absorbed by the atom to give a doubly charged ion; (2) a stepwise process in which the Kr^{2+} ion results from a 21-photon ionization of the Kr^+ ion. The value 21 is the first integer exceeding the ratio $E_2'/h\nu$ where E_2' is the energy of the second ionization threshold deduced from the ground state of the Kr^+ ion. One of the key results of this work is to have determined that the process involved is most probably a direct process. The Kr^{2+} ion is formed from a 33-photon transition from the ground state of the Kr atom to the second ionization threshold. This conclusion is backed up firstly by the I^{33} dependence of Kr^{2+} ions before the saturation point, and secondly, by the fact that satu-

ration occurs at the same laser intensity $I = 1.5 \times 10^{13}$ W cm^{-2} for Kr^{2+} and Kr^{+} ions. Kr^{2+} ions begin to be formed before saturation occurs for Kr^{+} ions.

The ratio of the number of doubly to singly charged ions deduced from Fig. 2 is 3% at the saturation point: $I_S = 1.5 \times 10^{13}$ W cm^{-2}. However, data in Fig. 2 are uncorrected as far as electron multiplier efficiency is concerned. The sensitivity of an electron multiplier to doubly charged Kr ions is nearly twice that for singly charged ions.[9] The corrected ratio R is now 1.5% at $I = 1.5 \times 10^{13}$ W cm^{-2}. The ratio R increases with laser intensity and reaches 10% at 8×10^{13} W cm^{-2} because, as shown in Fig. 2, beyond the saturation point, Kr^{2+} ions obey a I^3 law instead of the $I^{3/2}$ law of Kr^{+} ions. As far as Kr^{3+} and Kr^{4+} ions are concerned, data shown in Fig. 2 only correspond to a saturated part of the total curve which represents the variations in the number of ions formed as a function of laser intensity. Reference to Kr^{2+} ion data legitimizes the assumption that the Kr^{3+} ion is formed by a direct process from the ground state of the atom to the third ionization threshold, i.e., a 65-photon absorption process. In this assumption the I^{65} intensity dependence should only be observed for a laser intensity $I < I_S$. However, in this intensity range, the ionization rate is too low to allow us to measure Kr^{3+} ions. When the Kr^{3+} ion signal begins to emerge above the sensitivity threshold of the electron multiplier, which is roughly a single ion, the experimental points observed correspond to part of the saturated curve. At a laser intensity $I = 8 \times 10^{13}$ W cm^{-2}, the percentage ratio between the number of triply and doubly charged ions is 6%; the corresponding triply to singly charged ions ratio is 0.6%.

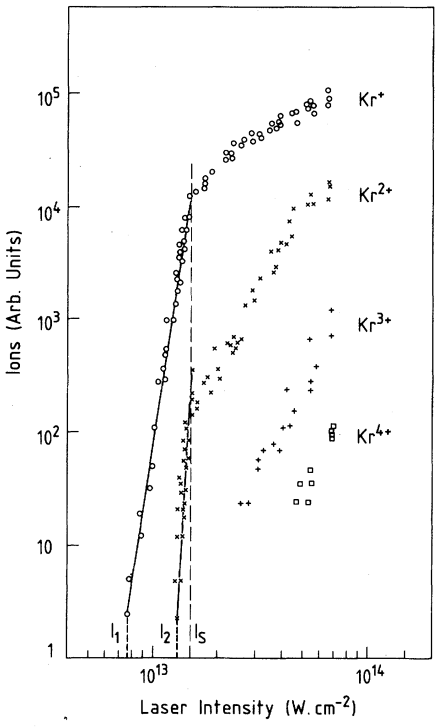

FIG. 2. Log-log plot of the variation in the number of krypton ions formed, N_i, as a function of the laser intensity I. An expanded scale is used for the laser intensity axis to more clearly represent experimental data, especially within the $(0.8\text{–}1.5) \times 10^{13}$-W-cm^{-2} range. Before saturation at $I_S = 1.5 \times 10^{13}$ W cm^{-2}, Kr^{+} and Kr^{2+} ions exhibit, respectively, I^{13} and I^{33} intensity dependence; an $I^{3/2}$ and I^3 dependence occurs beyond I_S. I_1 and I_2 are, respectively, the intensity thresholds for detection of Kr^{+} and Kr^{2+} ions.

Figure 3 shows schematically the processes involved in the present experiment to explain the origin of multiply charged ions. The key point of this work is that the 33-photon process leading to doubly charged ions can be induced with a laser intensity only 40% higher than that required for the 13-photon process which gives singly charged ions, as shown in Fig. 2, in the 0.8 to 1.5×10^{13}-W-cm^{-2} range. The 33-photon absorption rate is

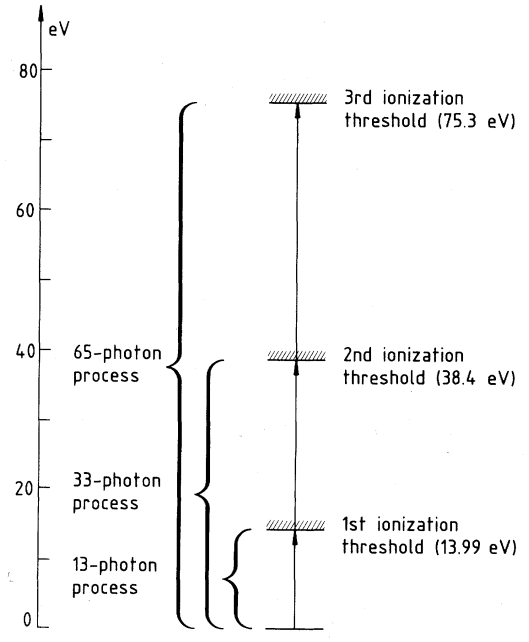

FIG. 3. Schematic representation of 13-, 33-, and 65-photon absorption processes generating Kr^{+}, Kr^{2+}, and Kr^{3+} ions, respectively, from ground-state krypton atoms.

only about 60 times less than the 13-photon absorption rate at 1.5×10^{13} W cm^{-2}. It should be pointed out that the 33-photon process consists of a 13-photon absorption in the discrete spectrum and a 20-photon absorption process in the continuum. For comparison purposes, the 22-photon ionization of He atoms corresponding to a 22-photon absorption process in the discrete spectrum leading to He^+ ions has been considered. This process requires a laser intensity of 10^{15} W cm^{-2}.[7] This shows that the multiphoton absorption rate in the continuum is surprisingly high compared to that in the discrete spectrum.

The basic processes involved in the present experiment could be tentatively explained as follows. Figure 2 shows that as the laser intensity increases, multiply charged ions are successively observed. Kr^+ ions are observed when the laser intensity I_1 is reached and Kr^{2+} ions as soon as I_2 is reached. Multiphoton absorption processes starting from the ground state of the atoms induce multiply charged ions. These processes exist at low intensities but with a probability too low to be observable. I_1 and I_2 are the threshold intensities for the production of Kr^+ and Kr^{2+} ions, respectively, in our experiment. The main result of the present work is that multiphoton absorption can induce rather easily the excitation of several electrons. Possible core rearrangements and correlation effects can occur and permit the production of multiply charged ions.[10] This tentative model will be checked in the near future by measuring the electron energy spectrum.

Doubly charged ions formed in the multiphoton ionization of Ba and Sr atoms have already been observed.[11] These atoms have two electrons in the outer shell so the process which induces double ionization could be different, and their authors have explained their results in terms of resonances and autoionizing states. Furthermore, these results were obtained in the 10^{10}–10^{11}-W-cm^{-2} intensity range where multiphoton absorptions in the continuum are expected to play a much lesser role than in the 10^{13}–10^{14}-W-cm^{-2} range.

In conclusion, this work clearly shows that multiply charged ions can easily be formed in the 10^{13}–10^{14}-W-cm^{-2} intensity range. One of the key points is that the 33-photon absorption rate leading to doubly charged ions is only about 60 times less than the 13-photon absorption rate leading to singly charged ions at 1.5×10^{13} W cm^{-2}. Furthermore, as doubly and triply charged ions are easy to obtain, it would be of interest to take advantage of the well-known radiative properties of such ions in the extreme ultraviolet. A tunable-wavelength laser could be used to selectively excite an ionic state and to investigate the extreme ultraviolet emission from this ionic state. This approach has not yet been considered.

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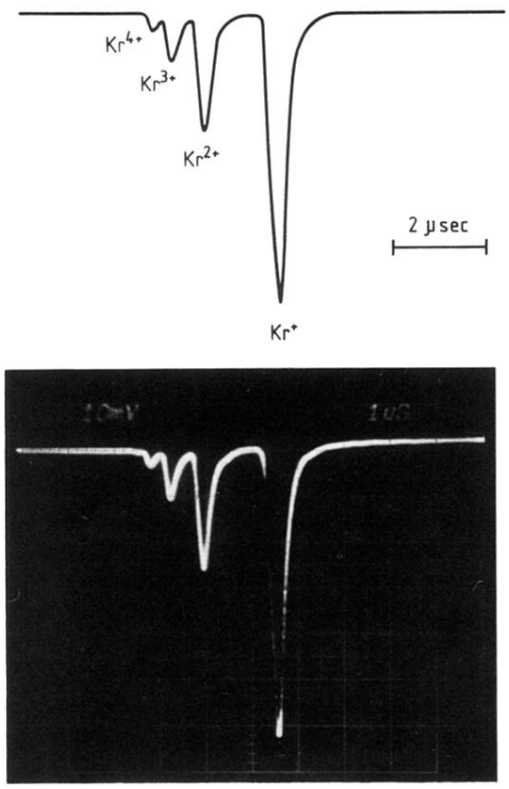

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