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Multiphoton Ionization of Rare Gases at Very High Laser Intensity (10^{15} W/cm^2) by a 30-psec Laser Pulse at 1.06 μm

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A bandwidth-limited 30-psec laser pulse at 1.06 μm is used to investigate multiphoton ionization of noble gases at very high laser intensity (10^{15} W/cm^2). For helium atoms interacting with a laser intensity of 1. 1×10^{15} W/cm^2, no departure from a pure 22-photon ionization process is observed, in contradiction with the few theoretical calculations which predict that a tunneling effect should appear in this laser-intensity range.

Multiphoton ionization processes have been investigated especially on 10^{-8}-sec time scales, by using Q-switched lasers.[1-4] It is of interest to investigate multiphoton ionization of atoms with much shorter pulses for various reasons. First, it is only with picosecond pulses that very high laser intensity (10^{16} W/cm^2) can be obtained at the present time. Thus the effect of very high intensity on multiphoton ionization can be investigated with such short pulses. Second, the shortening of the interaction time could greatly influence resonant multiphoton ionization processes which have been observed with 10^{-8}-sec pulses. Finally, a bandwidth-limited picosecond pulse is defined as a pulse completely devoid of intensity or frequency modulation. It has a regular and well-defined temporal behavior, similar to a single-mode laser pulse. Such a pulse has no fluctuation and its Kth-order peak-intensity autocorrelation function, or Kth-order peak-intensity moment, f_K equals 1.[5] Thus it makes for easier comparison between theoretical and experimental multiphoton-ionization probabilities.

The purpose of this Letter is to present some preliminary results of the first investigation of multiphoton ionization of rare gases by a 30-psec laser pulse. The laser used in the present experiment has been described elsewhere.[6] It is a mode-locked Nd-doped yttrium-aluminum-garnet oscillator using Kodak 9740 saturable dye. The TEM_{00} mode is selected with a 1.4-mm pinhole. The mode-locked output consists of a 10-mJ train of ten pulses. A single pulse is isolated from the train, outside the cavity, by the well-known method of a Pockels switch. The single pulse is selected in the rising edge of the train where pulses are usually bandwidth-limited. This single pulse goes through a beam-expanding telescope before entering a three-stage Nd:glass amplifier. The single pulse can thus be amplified to an energy up to 1 J. This laser radiation is linearly polarized and is centered at 10 643.5 Å with a linewidth of 0.8 Å. A linear measurement of the laser pulse shape was carried out by using a picosecond streak camera incorporating a storage-memory video system to visualize instantly and record the shape of the laser pulse. The laser pulse width was measured to be 28 ± 2.5 psec.[6] The corresponding time-bandwidth product had a value $\Delta\nu\,\Delta t = 0.7$.

Multiphoton ionization of five rare gases was performed by using an experimental arrangement identical to that employed in previous works.[5] Briefly, the laser pulse is focused into a vacuum chamber by an $f/3$ aspheric lens corrected for spherical aberrations. The gas under investigation is released into the vacuum chamber at a static pressure of 10^{-4} Torr for which no avalanche effects occur. The ions resulting from the laser interaction with atoms at the focal volume are extracted with a transverse electric field of 400 V cm^{-1} and then detected with an electron multiplier; 10 to 10^4 ions are generally collected. It should be stated that all the multiphoton ions formed at the focal volume can be extracted and detected when the transverse electric field is larger than about 200 V cm^{-1}. Ion data are the same when a transverse electric field of 400 or 500 V cm^{-1} is used to extract ions.

The experiment consists of measurements of the number of ions formed as a function of the laser intensity. In usual multiphoton-ionization

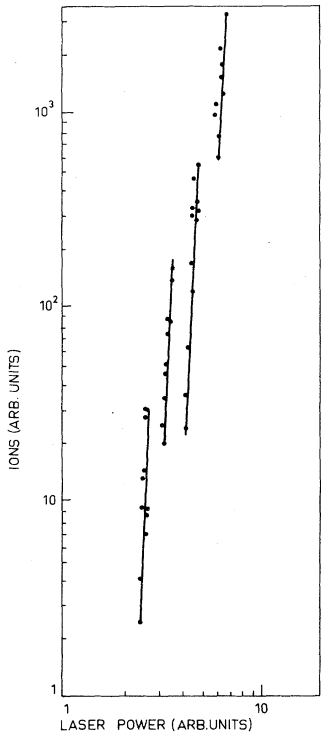

FIG. 1. Log-log plot of the variation of the number of helium ions as a function of the laser power. When the laser power is not intentionally varied, the experimental points form a short straight line as a result of the power fluctuations of the laser from shot to shot. When the laser power is varied by inserting neutral density filters in the laser beam, the effective surface of the focal volume is slightly changed, and experimental points form another short straight line which is shifted compared with the preceding one.

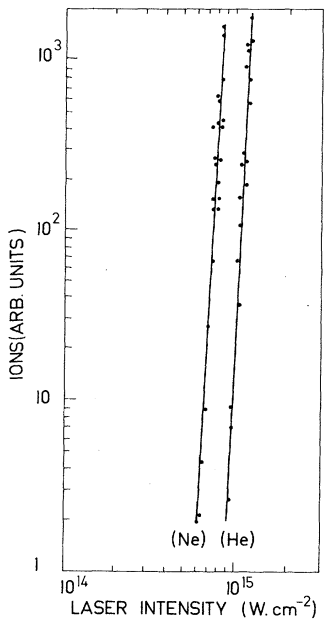

FIG. 2. Log-log plot of the variation of the number of neon and helium ions as a function of the laser intensity at the focal region.

experiments, the laser intensity is varied either by inserting neutral density filters in the laser beam, or by varying the pumping power of the amplifiers. These procedures are not useful in the present experiment. Figure 1 shows, in a log-log plot, the number of helium ions as a function of laser power. When the laser power is not intentionally varied, the experimental points form a short straight line as a result of the power fluctuations of the laser, from shot to shot, within about a 10% range. When the laser power is varied, either by inserting neutral density filters in the laser beam or by varying the pumping power of the amplifiers, the experimental points form another short straight line which is shifted compared with the preceding one, and so on. It seems that every time the laser power is intentionally varied, the effective surface of the focal volume is slightly changed. It should be pointed out that no self-focusing or filament formation has been observed. Both lens aberrations and laser aberrations govern the focused intensity distribution. Lens aberrations were minimized in the present experiment by the use of a corrected aspheric lens. Thus laser aberrations mainly govern the focused intensity distribution. Laser aberrations can be modified by varying the laser power, and can change the effective diameter of the focal region.

We have used a different method more suitable for the present experiment. The laser power is not intentionally varied, and a dye cell (Kodak 9740 in chlorobenzene) is put in the path of the laser beam to enhance the power fluctuations of the laser from shot to shot. The focal-volume measurements were carried out with the dye cell in place in the laser beam. These measurements were made with the full-power beam that was used for taking the multiphoton-ionization data. The focal region was reimaged with 40× magnification outside the interaction chamber and photographed with a film. Isodensitometer processing of film images then gave the laser intensity in the three-dimensional space near the focus. The focal-spot diameter has been measured to be 25 μm, that is, three times the corresponding diffraction-limited diameter. Figure 2 shows, in a log-log plot, the experimental results obtained for neon and helium atoms. The experimental points form a straight line with an exper-

TABLE I. Comparison of K_0 and K_{exp} values for the five noble gases. K_0 is the next integer greater than the ionization energy of the atom divided by the laser photon energy. K_{exp} is the slope of the law of variation of the number of ions as a function of the laser intensity, in log-log coordinates.

Atom	K_0	K_{exp}
Xe	11	11 ± 0.5
Kr	13	13 ± 0.5
Ar	14	14 ± 0.5
Ne	19	20 ± 2
He	22	23 ± 2

imental slope $K_{exp} = \partial \ln N_i / \partial \ln I$. The operating conditions of the laser were unaltered over a long time. Thus over a period of a month, multiphoton-ionization data were checked to be remarkably reproducible as far as the laser intensity, the number of ions formed, and K_{exp} values were concerned.

Table I gives K_{exp} values for the five rare gases, as well as the corresponding K_0 value which is the next integer greater than the ionization energy of the atom divided by the laser photon energy. Within the experimental errors the K_{exp} values are in good agreement with the corresponding K_0 values. This would mean that no resonant multiphoton ionization processes have taken place. Previous experiments have shown that K_{exp} can have values smaller or larger than K_0, depending on the laser wavelength, when there is a quasiresonant multiphoton excitation of an atomic level present.[7,1,4] The question is whether the laser wavelength just happens to be off resonance in these experiments, or whether no resonance occurs because of the very short interaction time or the high laser intensity which broadens the atomic levels and damps the resonance enough to sweep it away. The only way to answer this question is to investigate multiphoton-ionization probabilities as well as K_{exp} values, as a function of the laser wavelength. This is the reason why we are preparing a mode-locked Nd:glass laser which can be tuned over a range of 80 Å, with similar temporal and spectral characteristics as the mode-locked Nd-doped yttrium-aluminum-garnet laser.

It should be pointed out that multiphoton ionization of helium atoms was observed six years ago by using a 30-nsec multimode laser pulse at 10 586 Å with a laser intensity 30 times lower. This difference in laser intensities could be explained by considering the two most important parameters which govern multiphoton ionization processes, that is, resonance and laser temporal-coherence effects. The number of ions induced by a multimode pulse is nearly $K!$ times larger than that induced by a laser pulse of the same average intensity without fluctuation.[5] Now a bandwidth-limited-30-psec laser pulse has no fluctuation. Consequently, for a 22-photon process, multiphoton ionization of helium atoms with a bandwidth-limited 30-psec pulse should be observed for a laser intensity $(22!)^{1/22}$ (that is, 9) times larger than with a multimode 30-nsec pulse. The remaining difference in laser intensities could be explained by the fact that resonant processes occur at 10 586 Å, but do not seem to take place at 10 644 Å.

An important conclusion can be derived from the experimental results obtained for high laser intensity. Figure 2 shows, in a log-log plot, the experimental results obtained for neon and helium atoms at a laser intensity up to 10^{15} W/cm^2. The magnitude of the 22-photon ionization rate of helium atoms is $10^{10 \pm 1}$ sec^{-1} for a laser intensity $(11 \pm 4) \times 10^{14}$ W/cm^2. No lowering of the slope is observed. Consequently, a possible tunneling effect does not play any role at this laser intensity. Ionization of helium atoms seems to result only from a "pure" 22-photon process. It should be pointed out that an earlier work on the breakdown of two rare gases gave evidence of multiphoton ionization by 50-psec laser pulses.[8] It has been shown by Keldysh[9] that the behavior of the multiphoton ionization is related to the parameter γ defined as

$$\gamma = (\omega/eE)(2mI_0)^{1/2},$$

where ω and E are the angular frequency and peak electric field of the laser radiation, respectively. I_0 is the ionization energy of the atom, and e and m are the charge and mass of the electron, respectively. This parameter γ has been generally considered as a characterization parameter between multiphoton processes, $\gamma \gg 1$, and a tunneling effect, $\gamma \ll 1$. This is the first multiphoton ionization experiment in which the range $\gamma < 1$ was investigated. $\gamma = 0.32$ in our helium case, and $\gamma = 0.36$ for neon. The ratio of the laser field E to the atomic field E_0 is ~ 1 for neon. Thus multiphoton ionization processes are still observed in the intermediate region characterized by $\gamma \lesssim 1$ and $E/E_0 \sim 1$. This region does not seem to be adequately described in the theories.[9,10] The question is how far is it necessary to go be-

yond the atomic field in order to observe a departure from the multiphoton process in the ionization of an atom with very high laser intensity. We hope that these experimental results will stimulate further theoretical work in this direction, especially now that with high power lasers developed for laser-fusion experiments, $E/E_0 > 1$ can be attained.

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Photon-Correlation Effects in Resonant Two-Photon Ionization

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The effect of photon correlations (coherence) on the transition probability of resonant two-photon processes is discussed. It is shown that coherence influences not only the transition probability but also the linewidth of the resonance. The implications for higher-order multiphoton ionization are discussed.

Two relatively recent experiments[1,2] have brought again to the forefront one of the most interesting aspects of multiphoton processes; the dependence of the total transition rate on the coherence (correlation) properties of the light source. Krasinski *et al.*[1] have shown that the nonresonant two-photon absorption rate with a chaotic (incoherent) source is about 1.52–1.86 times higher than with an approximately coherent (Glauber state) source. Theoretically it should have been larger by a factor of 2, but the difference is readily understood since what was assumed to be a pure coherent state in fact was not.[3] In addition, a single-photon component may have existed in the signal. This experiment resolves an ambiguity stemming from contradictory results of two earlier experiments[4,5] performed under essentially identical conditions. The pulsed lasers used in those experiments were in all probability in a more or less chaotic state (because of the presence of many modes), and thus further randomization should have produced no additional effect. Therefore, the result obtained by Carussotto, Polacco, and Vaselli[5] is what one should have expected.

The most dramatic demonstration of coherence effects was provided by a beautiful experiment[2] by Lecompte *et al.* who showed that eleven-photon nonresonant ionization of Xe atoms with a multimode laser (100 modes $\approx$ chaotic state) is more efficient by a factor of $10^{6.9\pm0.3}$ than with a single-mode laser. Theoretically, the difference between perfectly incoherent (chaotic) and purely coherent light is $11! = 3.99\times10^7$.