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Wavelength dependence of nonsequential double ionization in He

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An enhancement of the He^{2+} yield near the appearance intensity was not observed by linearly polarized KrF laser light (248 nm), whereas a significant enhancement was observed by linearly polarized Ti:sapphire laser light (745 nm). These results are consistent with the quasistatic model proposed by Corkum [*Technical Digest of Short Wavelength V: Physics with Intense Laser Pulses* (Optical Society of America, Washington, DC, 1993), p. 25].

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A significant enhancement of He^{2+} ionization has recently been observed near the appearance intensity by using linearly polarized 614-nm light by Fittinghoff *et al.* [1], and this ionization has been interpreted as evidence of nonsequential double ionization. To explain this phenomenon, they proposed a shake-off mechanism where the second electron left in an excited state is ionized, during the same laser cycle, immediately after the first electron leaves the atom. According to this model, the shake-off mechanism for nonsequential ionization should remain with circular polarization and all optical wavelengths provided the pulse width is short enough [1]. Two-electron ejection should therefore be observed both with circular polarization and with short wavelengths. More recently, however, the same group reported that there was no enhancement of He^{2+} and Ne^{2+} ionization with circular polarization at 614 nm, whereas significant enhancements were observed for both ions with linear polarization [2]. Corkum proposed a different model as the quasistatic model [3], which can explain this enhancement. This model can also explain the cutoff photon energy of the harmonic plateau [4,5] and the high-energy tail in the above-threshold ionization (ATI) spectrum [6]. Schafer *et al.* also described the similar model, as a "two-step" semiclassical model, to explain a cutoff photon energy of the harmonic plateau [7]. In the quasistatic model, there is a significant probability of the electron returning to the vicinity of the ion core within a laser cycle after tunneling ionization induced by using linearly polarized light. The maximum kinetic energy of the returning electron is estimated to be $3.17U_p$ by this model, where U_p is the ponderomotive potential. At a longer laser wavelength, for example 614 or 745 nm, the maximum kinetic energy becomes well above the ionization potential of He^+ (54.4 eV), and the electron-impact ionization of He^+ occurs to an extent that results in two-electron ejection within one laser cycle [8]. If this model is valid, two-electron ejection from He should not occur with circular polarization because the ionized electron would never return to the vicinity of the ion core. And this is consistent with the experimental results in Ref. [2]. At shorter laser wavelengths, for example 248 nm, the maximum kinetic energy ($3.17U_p$) becomes less than the ionization potential of He^+ and two-electron ejection from He would not occur even if linearly polarized light were used.

In this study, we observed the yield of He^+ and He^{2+} in high-field ionization of He using ultrashort KrF and Ti:sapphire lasers. When a linearly polarized Ti:sapphire laser (745 nm) was used for ionization of He, a significant enhancement of He^{2+} near the appearance intensity was observed. There was, however, no enhancement when a linearly polarized KrF laser was used. This result is consistent with the quasistatic model proposed by Corkum [3].

Two kinds of ultrashort pulse lasers were used in this study. One was a KrF laser, with a typical energy and pulse width of 200 mJ and 440 fs. The other was a Ti:sapphire laser, with a typical energy and pulse width of 45 mJ and 200 fs. Details of these systems are reported in Ref. [9]. The Ti:sapphire laser was operated at 745 nm and the KrF laser at 248 nm. Typical spot diameters were 7 μm at 248 nm, by using an achromatic lens ($f/300$ mm), and 26 μm at 745 nm, by using a planoconvex lens ($f/300$ mm). The peak intensity in the experiment was estimated as $0.61E/(\tau\pi r^2)$, where E is the laser energy, τ is the pulse width [full width at half maximum (FWHM)], and r is the spot radius [half width at half maximum (HWHM)].

First we measured the ion yield of He^+ and He^{2+} for the Ti:sapphire laser. A time-of-flight (TOF) analyzer (R. M. Jordan Co.) was used for this measurement. Helium (^4He) backfilled the target chamber to a pressure of 4.7×10^{-7} Torr after a turbomolecular pump had evacuated the chamber to below 5×10^{-9} Torr. The acceleration field was 900 V/cm and the drift length was 1.4 m. The ions were detected with a two-state microchannel plate (MCP) that had a total gain of about 10^6. The time resolution was good enough to clearly resolve $H_2{}^+$ from He^{2+} even though their mass-to-charge ratios were both close to 2 (2.0013 for He^{2+} and 2.0164 for $H_2{}^+$). The ion spectra were recorded by a digital signal analyzer (Tektronix DSA 602). When we measured the ion yield of He^{2+}, the time window was carefully set to the He^{2+} position in order to exclude the $H_2{}^+$ signal [1]. The intensity dependence of He ionization by Ti:sapphire laser light with linear polarization is shown in Fig. 1. Solid curves are predicted by the quasistatic model [3] for each charge state at 745 nm. In this model, the ionization probability at the "first step" is assumed to be given by Ammosov-Delone-Krainov (ADK) tunneling theory [10]. After tunneling, the electron motion in the field is given by the classical theory. The electron-impact ionization cross section of He^+ is given by Ref. [8]. And the wave function was assumed to have a Gaussian probability distribution for the impact parameter with a radius of 1.5 Å [3]. Our calculation for He^{2+} was checked against the values in Ref. [3] at 600 nm. Of course the curve for He^+ comes purely from ADK theory. The dotted curve was calculated by ADK tunneling theory for He^{2+}, assuming sequential ionization. Open circles fit very well with the quasistatic model. Below 6×10^{15} W/cm^2 there is a clear difference between experimental plots and ADK theory, but the experimental plots agree well with the quasistatic model. A significant enhancement of He^{2+} near the appearance intensity can be seen in Fig. 1, in agreement with the results at 614 nm [1,2]. The upper and lower horizontal axes respectively show the intensities fit to the quasistatic model or ADK theory and to the measured intensity. These scales differ by a factor of 2. This overestimate of the measured intensity is due to the incompleteness or to the underestimate of focal spot or both. The full energy may not completely concentrate on the central Gaussian profile. So an uncertainty factor of 2 would be inevitable in estimating the absolute intensity.

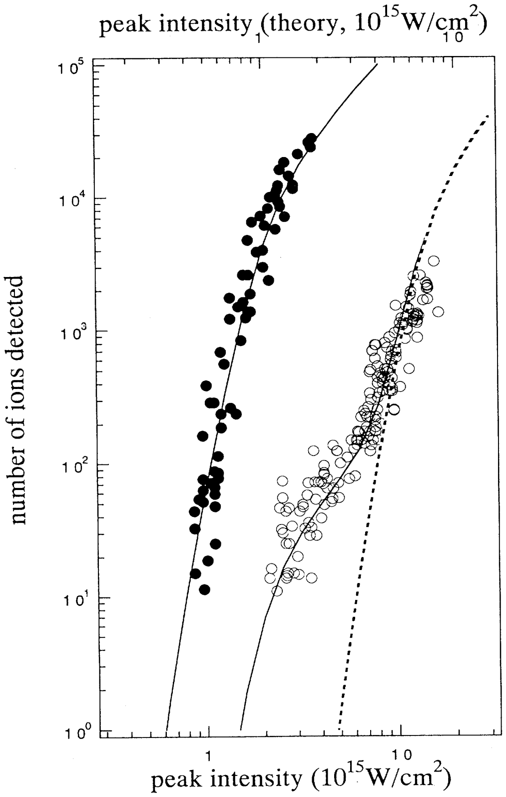

FIG. 1. Numbers of He^+ (filled circles) and He^{2+} (open circles) ions as a function of the peak intensity of Ti:sapphire laser light (745 nm). Solid curves show values predicted by the quasistatic model. The dotted curve is from ADK theory for He^{2+}, where sequential ionization is assumed. The upper and lower horizontal axes show the intensities fitted to the theoretical and raw experimental values, respectively. They differ by a factor of 2.

When using the KrF laser, we tried to detect the signal of He^{2+} the same way we did when using the Ti:sapphire laser, but there was only one peak near the position of He^{2+} or $H_2{}^+$ in the TOF spectra. And the same signal was detected even in vacuum. This signal can be attributed to $H_2{}^+$ created from H_2O in the evacuated target chamber. This signal is plotted in Fig. 2 for the vacuum pressure of 3×10^{-8} Torr. (The solid curves shown for reference were determined by fitting ADK theory to the experimental data of He^+ obtained at the same vacuum pressure and a He backfill pressure of 3×10^{-6} Torr.) The signal level certainly decreased with improving vacuum, but a significant He^{2+} signal was still not found. All the signals of the H_2O-related ions (H^+, $H_2{}^+$, O^{2+}, and H_2O^+) become stronger in KrF laser light than in Ti:sapphire laser light because a single photon (5 eV) of KrF laser light can dissociate H_2O. We therefore used ^3He instead of ^4He so that we could completely discriminate it from the background $H_2{}^+$ signal. The $^3He^{2+}$ signal level was still low because of the MCP saturation due to scattered KrF light and the signal of H^+. Therefore a gate voltage of 800 V with a 1-μs duration was applied,

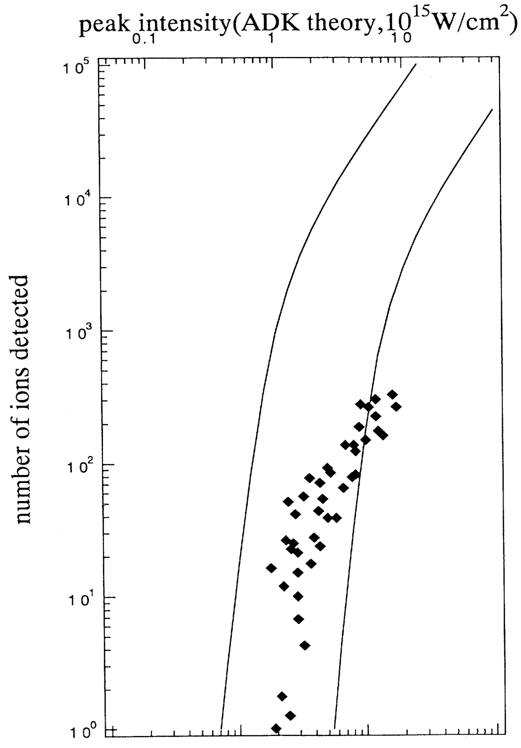

FIG. 2. Number of $H_2{}^+$ ions detected as a function of peak intensity of KrF laser light (248 nm) under a vacuum (3×10^{-8} Torr). Solid curves calculated from ADK theory for He are shown for reference. Theoretical curves are fitted to experimental data obtained for the same vacuum pressure and a He backfill pressure of 3×10^{-6} Torr.

with an appropriate delay, to one stage of the MCP in order to select only ^{3}He$^+$ or ^{3}He^{2+} signals. The total gain of the MCP was slightly lower in a gate mode than in a dc mode because in a dc mode 960 V were applied to each stage. With these improvements, we successfully obtained the ^{3}He^{2+} data. Of course, exactly the same intensity dependence of ^{3}He$^+$ and ^{3}He^{2+} was obtained in a gate mode as in a dc mode at 745 nm. The intensity dependence of ^{3}He$^+$ and ^{3}He^{2+} ion yields in KrF laser light with linear polarization is shown in Fig. 3. The difference of MCP gain between both modes is not taken into account in these data. The solid curves were calculated by using ADK theory because the two-step process does not contribute to ionization. In this intensity range, the ionization by KrF laser light is also in the tunneling regime [11,12], whereas the Keldysh γ parameter is larger than unity. And there is no enhancement of He^{2+} near the appearance intensity.

According to the model in Ref. [1], a short pulse width is essential for two-electron ejection from He. According to the quasistatic model, however, a large kinetic energy (the maximum of which is $3.17U_p$) is essential for two-electron ejection. Two-electron ejection should therefore occur when the intensity is high enough. This means that the absence of the enhancement with KrF laser light is not due to a pulse width of 440 fs, although it is slightly longer than that of Ti:sapphire (200 fs). Actually, the same enhancement in the appearance intensity for He^{2+} was observed with a 1-ps chirped pulse at 745 nm. This result contradicts Ref. [13] where there was no enhancement with a 1.5-ps pulse at 1.06 μm. The reason for this difference is unclear. When the pulse width is much longer, the ionization occurs at a lower intensity because multiphoton ionization dominates and the $3.17U_p$ energy is less than 54.4 eV because of the low ionization intensity. As a result, two-electron ejection should not be observed [14].

We also investigated the ionization with circular polarization at 745 nm in 200 fs. No enhancement of He^{2+} was observed as in Ref. [2]. The quasistatic model proposed by Corkum is thus supported both by the wavelength dependence and the polarization dependence.

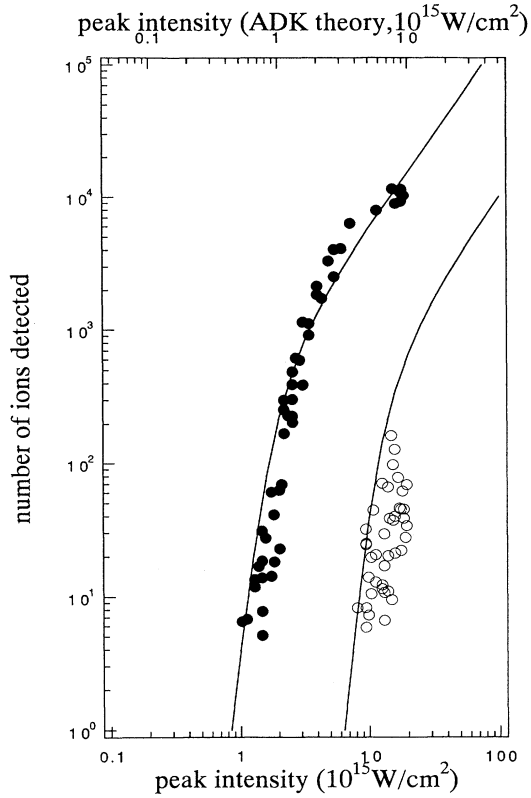

FIG. 3. Numbers of He$^+$ (closed circles) and He^{2+} (open circles) as a function of the peak intensity of KrF laser light (248 nm). Theoretical curves are calculated from ADK theory. (The two-step process does not contribute in this case.) The upper and lower scales differ by a factor of 2.

In summary, we have measured He^{2+} ion yield while varying the peak intensity of the Ti:sapphire and KrF lasers. When we used near-infrared linearly polarized light, we observed a significant enhancement of He^{2+} near the appearance intensity. When we used linearly polarized ultraviolet light, however, we saw no enhancement. These results are consistent with the quasistatic model.

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*FAX: 03-3401-5169.

[1] D. N. Fittinghoff, P. R. Bolton, B. Chang, and K. C. Kulander, Phys. Rev. Lett. **69**, 2642 (1992).

[2] D. N. Fittinghoff and P. R. Bolton, in *Technical Digest of Short Wavelength V: Physics with Intense Laser Pulses* (Optical Society of America, Washington, DC, 1993), p. 29.

[3] P. B. Corkum, in *Technical Digest of Short Wavelength V: Physics with Intense Laser Pulses* (Ref. [2]), p. 25.

[4] J. L. Krause, K, J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).

[5] J. J. Macklin, J. D. Kmetec, and C. L. Gordon III, Phys. Rev. Lett. **70**, 774 (1993).

[6] U. Mohideen, M. H. Sher, H. W. K. Tom, G. D. Aumiller, O. R. Wood II, and P. H. Bucksbaum, in *Technical Digest of Short Wavelength V: Physics with Intense Laser Pulses* (Ref. [2]), p. 32; U. Mohideen, M. H. Sher, H. W. K. Tom, G. D. Aumiller, O. R. Wood II, R. R. Freeman, J. Bukor, and P. H. Bucksbaum, Phys. Rev. Lett. **71**, 507 (1993).

[7] K. J. Schafer, B. Tang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. **70**, 1599 (1993).

[8] H. Tawara and T. Kato, At. Data Nucl. Data Tables **36**, 167 (1987).

[9] Y. Nabekawa, N. Sarukura, K. Sajiki, K. Kondo, and S. Watanabe, Opt. Lett. (to be published).

[10] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].

[11] G. Gibson, T. S. Luk, and C. K. Rhodes, Phys. Rev. A **41**, 5049 (1990).

[12] K. Kondo, T. Tamida, Y. Nabekawa, and S. Watanabe (unpublished).

[13] J. Peatross, B. Buerke, and D. D. Meyerhofer, Phys. Rev. A **47**, 1517 (1993).

[14] A. L'Huillier, A. Lompre, G. Mainfray, and C. Manus, Phys. Rev. A **27**, 2503 (1983).