PHYSICAL REVIEW A **68**, 065403 (2003)

Ionization of many-electron atoms by ultrafast laser pulses with peak intensities greater than 10^{19} W/cm^2

K. Yamakawa,* Y. Akahane, Y. Fukuda, M. Aoyama, N. Inoue, and H. Ueda
Advanced Photon Research Center, KANSAI Research Establishment, Japan Atomic Energy Research Institute, 8-1 Umemidai, Kizu,
Kyoto 619-0215, Japan

(Received 10 August 2003; published 23 December 2003)

We report on the superstrong field ionization of complex rare gases (xenon, krypton, and argon) by 800-nm, 10-Hz laser pulses of 25-fs duration with a peak intensity greater than 10^{19} W/cm^2, whose electric field well exceeds the Coulombic binding field in strength. We observed charge states as high as Xe^{26+}, Kr^{19+}, and Ar^{16+}, which correspond to Ni-like Xe, Cl-like Kr and He-like Ar, respectively. We compared integral ionic charge state yields of these atoms with those results from the Ammosov-Delone-Krainov model. We found that ionization at a given intensity depends on the atomic species in the fully relativistic field for the first time to our knowledge.

DOI: 10.1103/PhysRevA.68.065403 PACS number(s): 42.50.Hz, 32.80.Fb, 32.80.Rm

Production of highly charged ions via laser-matter interactions has been a subject of great interest ever since high power laser sources became available. Over the last two decades a number of groups has studied the interaction of strong laser fields with atoms in the regime from multiphoton to tunneling ionization [1]. In the tunneling regime, ionization of atoms exposed to the strong laser fields has been investigated extensively for many atomic gases in the intensities ranging from 10^{13} to 10^{16} W/cm^2 [2–5]. Nowadays, the evolution of reliable, ultrafast, chirped-pulse-amplification (CPA) technologies has allowed systems to produce 10-fs-range, multiple-terawatt peak power pulses [6]. Such lasers can access to focused peak intensities of 10^{20} W/cm^2 at 36 000 shots per hour. These intensities correspond to 60 times the field binding the ground state electron in the hydrogen atom, and inner shell electrons for heavy (high z) atoms cannot be held within the atomic center. At such intensities the electron velocity in the laser field also becomes relativistic. Thus the free electrons move at close to the speed of light and their mass changes dramatically compared to their rest mass. Consequentially, it is now possible to study the ionization dynamics of the atoms in entirely new regimes.

Recently, the ionizations of rare gas atoms were studied in a laser intensity range from 10^{18} to 10^{19} W/cm^2 at different laboratories. Chowdhury *et al.* have compared the measured ion yields for Ar^{9+}-Ar^{16+} with the results form the nonrelativistic Schödinger equation, WKB and ADK (Ammosov-Delone-Krainov) tunneling ionization models [7]. They concluded that the observed ionization rates are in good agreement with the calculated ones using these three rates. Dammasch *et al.* have obtained indirect evidence for suppression of nonsequential multiple ionization of Xe in the relativistic laser field [8]. It was expected to occur due to the relativistic forward drift of the ionized electron, and therefore the electron loses the parent ion and no rescattering will occur. Since a very small number of experimental studies of relativistic laser-atom interactions has been carried out, a further study of the interaction of radiation at the intensity of over 10^{19} W/cm^2 with various complex atoms provides a knowledge of fundamental properties of atoms and ions under such extreme conditions, as well as solutions to control these new physical effects for potential applications, such as the generation of Larmor radiation [9], ultrahigh-order harmonic generation [10], particle acceleration [11], and optical field ionization x-ray lasers [12].

In this paper we report on optical field ionizations of complex rare gas atoms exposed to super-intense 800-nm, 10-Hz laser pulses of 25-fs duration. We have studied multiple ionizations of three rare gases (Xe, Kr, and Ar) and observed charge-states as high as Xe^{26+}, Kr^{19+}, and Ar^{16+} at a laser intensity of up to $\sim 3\times 10^{19}$ W/cm^2 in vacuum. We have compared integral ionic charge state yields of these atoms with those results from the ADK model [13]. We found that the ionization of many-electron atoms is dependent both on the ionization potential and on the atomic species in the fully relativistic field. It is suggested that the nonrelativistic ADK tunneling ionization theory is still valid to describe the bulk of the ion yields of various complex atoms even in the relativistic regime.

To perform an ultrahigh-field atomic phtoionization experiment, we used a state-of-the-art 100-TW, 20-fs, 10-Hz Ti:sapphire CPA laser system which is capable of producing focused laser intensity up to 10^{20} W/cm^2 [14]. In this experiment, the maximum energy and the pulse duration were measured to be 530 mJ and 25 fs, respectively. The diameter of the focal point of the attenuated laser beam was analyzed by an objective lens associated with a CCD camera. Using an off-axis parabolic mirror with a focal length of 161 mm, we obtain a spot diameter of 11 μm at $1/e^2$ with 53% of full energy, which corresponds to the estimated peak intensity of 2.6×10^{19} W/cm^2. The intensity calibration measured with the photoionization of helium was also performed. The discrepancy in the values of the intensities between them is less than $\pm 15\%$. A signal-to-background intensity contrast ratio was also measured to be approximately 10^{-6} at a time scale of ± 2 ns. 500 and 75-l molecular turbo pumps in series are

---

*Email address: yamakawa@apr.jaeri.go.jp

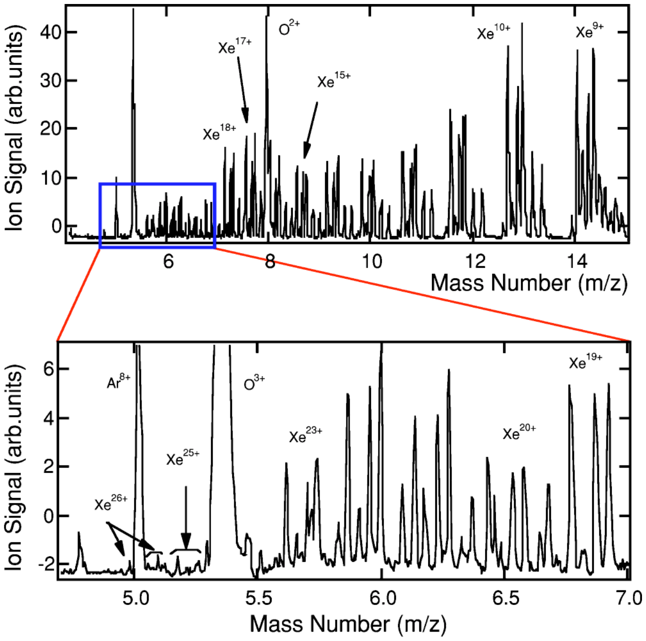

FIG. 1. Time-of-flight ion mass spectrum of xenon at the laser intensity of 2.6×10^{19} W/cm^2. The gas pressure was below 1×10^{-7} Torr. The appearance of Ar^{8+} is due to the result of mixed gases in order to determine the relative laser intensities among the gases.

used to evacuate the ionization chamber to a background pressure below 8×10^{-9} Torr. Rare gases such as Xe, Kr, and Ar were introduced into the chamber with a precision leak valve. Gas pressures in the chamber were controlled typically below 1×10^{-7} Torr in order to reduce space charge and collective effects. The ion species are separated with a 1-m time-of-flight (TOF) mass spectrometer and detected with dual microchannel plates (MCPs). The spectrometer was designed to resolve the isotope structure of neutral Xe. An adjustable slit with a width of 300 μm was placed between the acceleration grids and the MCP detector on the central axis of the flight tube in order to detect the ions produced in the highest intensity focal region while eliminating the lower charged ions and embarrassing noise produced by ionizing contaminants in the large focal volume and low-intensity region. Each data run consists of 3000 laser shots with the laser energy-fluctuations of less than 5%, which was not accessible by using the large-scale, single-shot per hour, high-intensity Nd:glass CPA lasers.

Figure 1 shows the typical TOF mass spectrum of Xe ions produced at the laser intensity of up to 2.6×10^{19} W/cm^2. As shown in Fig. 1, the charge states up to Xe^{26+}, which corresponds to Ni-like Xe have been observed. The appearance of Ar^{8+} is due to the result of mixed gases in order to determine the relative laser intensities among the gases. As for Kr and Ar, the charge states have also been detected as high as19 and 16, which corresponds to Cl-like Kr and He-like Ar, respectively. These are the highest charge states produced by the collisionless ionization of these atoms to our knowledge. It should be noted that all the charge states were produced in the regime of $\gamma\ll1$ (Keldysh γ parameter) [15], which indicated that we performed the experiment exclusively in the tunneling regime. In the nonrelativistic tunneling regime, the ADK model provides a relatively fit to sequential ionization rates of atoms. In order to clarify the ionization dynamics of the different atomic species in the fully relativistic laser field, we first compare the integral ionic charge state yields of Xe, Kr, and Ar with those results from the ADK model. Although no attempt was made to evaluate the ADK model to apply to the various complex atoms into the relativistic regime, our experimental data will provide an important step towards improving our understanding of the superstrong field ionizations of the atoms.

In order to compare between the experimental and theoretical charge ion yields, we will follow Chowdhury *et al.*, who characterized the degree of agreement between them for highly charged argon ions [7]. We calculate the minimum sum of square differences between the experimental values obtained at the intensity of 2.6×10^{19} W/cm^2 and the calculated yields at the various intensities between 1.0×10^{18} and 3.0×10^{19} W/cm^2 for Xe, Kr, and Ar, respectively. In the calculation, we assume that the focused laser beam is a Gaussian spatial profile with a squared hyperbolic-secant time envelope. Since all the ions discussed here produce within the slit that mitigates the volume effects at the larger and lower-intensity regions, these effects were already taken into account in the ADK calculation.

Figure 2 shows the experimental and calculated ion yields in the case of best agreements, for (a) Xe^{17+} to Xe^{26+}, (b) Kr^{13+} to Kr^{19+}, and (c) Ar^{9+} to Ar^{16+}, respectively. No Xe^{24+}, Ar^{10+}, and Ar^{15+} are observable due to the presence of O^{3+}, O^{4+}, and O^{6+} from residual ionized H_2O in the vacuum chamber. Large differences between the experiments and calculations for Ar^{9+} and Kr^{18+} may be due to the inadequacy of the ADK rates since binding potentials between the charge states 8 and 9, and 18 and 19 are so large. As shown in Fig. 2, the best agreement between the experimental and theoretical yields occurs at the theoretical laser intensities of 3.5×10^{18}, 4.1×10^{18}, and 1.3×10^{19} W/cm^2 for Xe, Kr, and Ar, respectively. The calculated intensity disagrees with the experimental one for Ar by a factor of 2, while a comparison between the theoretical intensities from the ADK model and the experimental ones for Kr and Xe shows rather poor agreement. As a result, the model derived the peak intensities vary by a factor of 3.7. We emphasize that we repeated the measurements for several times and saw no discernible changes in the relative intensities between all these atoms, which means that the disagreements are due to some physical differences in the ionization processes.

In 1989, Augst *et al.* found that the appearance intensity is dependent both on the ionization potential and on the atomic species [3]. The appearance intensity is defined as the intensity at which a small number of ions are produced on a particular charge state. It was observed that there was systematic lowering of the appearance intensity with increasing atomic number. Considering the situation of our results, we first predicted the saturation intensities on each charge state of the three rare gases by the ADK model. The saturation intensity is where the growth in the ion-yield curve levels off to a 3/2 power law in intensity. We use the saturation intensity instead of the appearance intensity, since the saturation

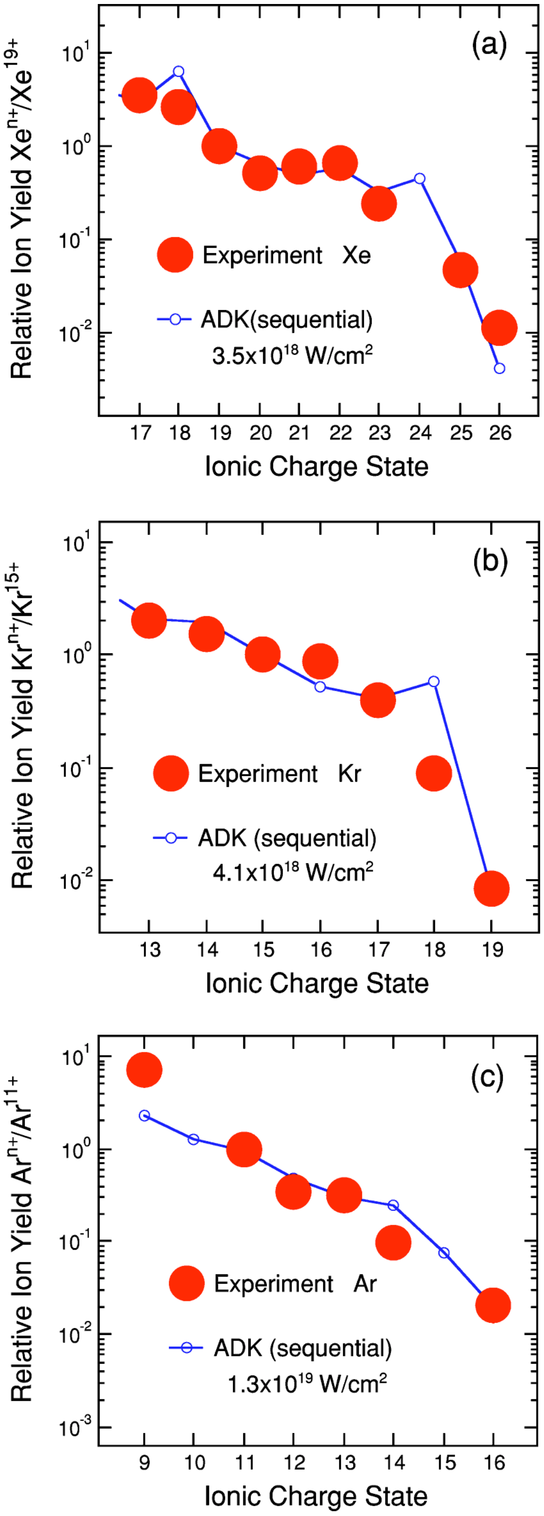

FIG. 2. Multiply charged ion yields for (a) xenon, (b) krypton, and (c) argon obtained at a laser intensity of 2.6×10^{19} W/cm^2. The closed and open circles are the experimental and calculated results, respectively. The laser intensities obtained from the numerical calculation based on the Ammosov-Delone-Krainov (ADK) model, in which the best fits to the experimental data, by calculating the minimum sum of squares differences, are also indicated. The experimental points shown in each figure are normalized to the calculated Ar^{11+}, Kr^{15+}, and Xe^{19+} yields, respectively.

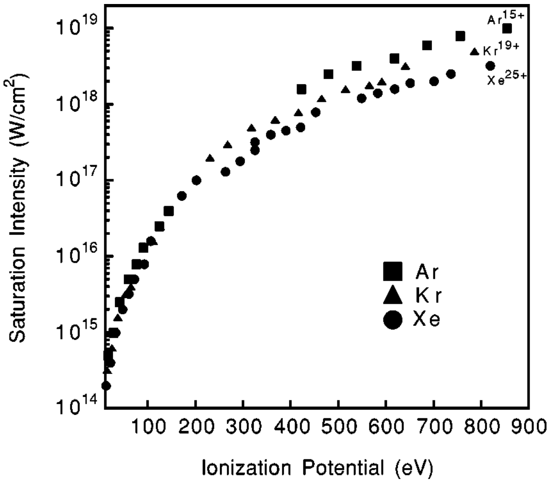

FIG. 3. Calculated saturation intensity vs ionization potential for three rare gas atoms.

threshold is independent of the gas pressures and therefore eliminates an experimental uncertainty somewhat. Figure 3 shows a plot of the calculated saturation intensities for the each charge state of ions of the three rare gases versus the ionization potential. The saturation intensities of Xe, Kr, and Ar shown in Fig. 3 separate into a group of curves. The curves of smaller atomic numbers lie above those of larger atomic numbers. This ordering means that the laser fields remove electrons from higher valence shells more easily than from lower valence shells for a fixed ionization potential. Because the ion yields of Xe^{25+}, Kr^{19+}, and Ar^{15+} are close to or above saturations at the highest laser intensities, the saturation intensities of these charge states were calculated to be 3.2×10^{18}, 5.0×10^{18}, and 1.0×10^{19} W/cm^2, respectively. Across all the three rare gases the calculated peak intensities is differ by a factor of 3.1, indicating a validation of relative agreement between the experimental data and the calculated yields as shown in Fig. 2.

The ADK model is based on a quasi-classical ac-tunneling theory. The rate of tunneling ionization of the complex atoms in a linearly polarized field with the effective principal quantum number n^*, the orbital quantum number l, and the magnetic quantum number m is given by [14]

$$W_{ac}=\omega_A C_{n^*l}^2 f(l,m) E_i \left(\frac{3E_L}{\pi(2E_i)^{3/2}}\right)^{1/2} \times\left[\frac{2}{E_L}(2E_i)^{3/2}\right]^{2n^*-|m|-1} \exp\left[-\frac{2}{3E_L}(2E_i)^{3/2}\right], \tag{1}$$

where ω_A is the atomic unit of frequency, E_L is the electric field of the laser in atomic units, E_i is the atomic ionization potential, and the effective principal quantum number n^*, and the factors f and C are given by

$$n^*=\frac{Z}{(2E_i)^{1/2}}, \tag{2}$$

$$f(l,m)=\frac{(2l+1)(l+|m|)!}{2^{|m|}(|m|)!(l-|m|)!}, \tag{3}$$

and

$$C_{n^*l}=\left(\frac{2e}{n^*}\right)^{n^*}\frac{1}{(2\pi n^*)^{1/2}}, \tag{4}$$

where Z is the charge of the ion. It is known form Eq. (1) that the species dependence of the ionization rates to be a dependence on the effective principal quantum number n^* of the valence shell from which the electron is removed. This dependence on the quantum number n^* arises in the complex atoms because the rate at which an atom or ion ionizes in the model depends not only on the ionization energy of the electron but also on the initial state of the electron. It is consistent with our experimental data, although we only consider tunneling ionization in the frame of nonrelativistic quantum mechanics. It is therefore suggested that the nonrelativistic ADK tunneling ionization theory is still valid to describe the bulk of the ion yields of various complex atoms even in the relativistic regime. Though our result shows that the initial ionization process is nonrelativistic even in the relativistic field, an ejected electron should obtain the relativistic energy in the final continuum state that is on the order of its rest energy mc^2. For example, the ponderomotive energy of the free electron is on the order of 1 MeV at a laser intensity of over 1×10^{19} W/cm^2, and therefore the electron velocity becomes fully relativistic. Further experiments are currently underway to measure a forward electron momentum due to the relativistic quiver motion and kinetic energy of electrons [16] and relativistic high-order harmonics [10] in a superstrong laser field. Although our calculations ignore nonsequential ionization processes [8,17–19], more detailed investigations are also in progress by measuring the ion yields as a function of laser intensity and comparing these results with the ADK model.

In conclusion, we have studied tunneling ionization of complex rare gas atoms (Xe, Kr, and Ar) by using a 100-TW, 20-fs, 10-Hz Ti:sapphire laser system. Highly charged ions as high as Xe^{26+}, Kr^{19+}, and Ar^{16+} have been observed at a laser intensity up to 2.6×10^{19} W/cm^2. We have found that the ionization of many electron atoms at a given intensity is dependent both on the ionization potential and on the atomic species in the fully relativistic field for the first time to our knowledge. Furthermore, laser peak intensities can be reasonably determined by using a nonrelativistic ADK model even in the relativistic field. The experimental data presented here will provide an important step towards improving our understanding of the atoms and ions exposed to relativistic laser fields. The study presented in this paper should also lead to a better understanding of relativistic plasmas of atoms, molecules, and clusters in all of their complex interactions for future studies.

The authors acknowledge T. Utsumi for his computational support and T. Kimura, T. Tajima, and Y. Kato for their encouragement.

---

[1] See, for example, *Multiphoton Processes*, edited by L. F. DiMauro, R. R. Freeman, and K. C. Kulander, AIP Conf. Proc. No. 525 (AIP, Melville, NY, 2000).

[2] F. Yergeau, S. L. Chin, and P. Lavigne, J. Phys. B **20**, 723 (1987).

[3] S. Augst *et al.*, Phys. Rev. Lett. **63**, 2212 (1989).

[4] E. Mevel *et al.*, Phys. Rev. Lett. **70**, 406 (1993).

[5] S. Larochelle, A. Talebpour, and S. L. Chin, J. Phys. B **73**, 1201 (1998).

[6] K. Yamakawa and C. P. J. Barty, IEEE J. Sel. Top. Quantum Electron. **6**, 658 (2000).

[7] E. A. Chowdhury, C. P. J. Barty, and B. C. Walker, Phys. Rev. A **63**, 042712 (2001).

[8] M. Dammasch *et al.*, Phys. Rev. A **64**, 061402 (2001).

[9] Y. Ueshima *et al.*, Laser Part. Beams **17**, 45 (1999).

[10] S. X. Hu and C. H. Keitel, Phys. Rev. A **63**, 053402 (2001).

[11] T. Tajima and J. M. Dawson, Phys. Rev. Lett. **43**, 267 (1979); V. Malka *et al.*, Science **298**, 1596 (2002).

[12] N. H. Burnett and P. B. Corkum, J. Opt. Soc. Am. B **6**, 1195 (1989); S. Sebban *et al.*, Phys. Rev. Lett. **86**, 3004 (2001).

[13] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **9**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].

[14] K. Yamakawa *et al.*, Opt. Lett. **23**, 1468 (1998).

[15] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].

[16] C. I. Moore, J. P. Knauer, and D. D. Meyerhofer, Phys. Rev. Lett. **74**, 2439 (1995).

[17] D. N. Fittinghoff *et al.*, Phys. Rev. Lett. **69**, 2642 (1992).

[18] B. Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994).

[19] Th. Weber *et al.*, Nature (London) **405**, 658 (2000).