Tunneling Ionization of Noble Gases in a High-Intensity Laser Field

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(Received 26 June 1989)

Studies of multiphoton ionization of noble gases have been carried out using 1-μm, 1-ps laser pulses with intensities up to the mid 10^{16} W/cm^2. To our knowledge, this work represents the first study of the production of highly ionized noble-gas ions done exclusively in the tunneling regime. It is found in this regime that the ionization at a given intensity depends on both the ionization potential and the charge state of the species. The onset of ionization occurs when the sum of the Coulomb and laser electric potentials causes the electron to be unbound.

PACS numbers: 32.80.Rm, 32.80.Wr, 42.50.Hz

Nonresonant ionization due to intense laser fields can be separated into two regimes by the Keldysh tunneling parameter,[1]

$$\gamma = (E_{\text{ion}}/2\Phi_{\text{pond}})^{1/2}, \tag{1}$$

where Φ_{pond} is the ponderomotive potential of the laser and E_{ion} is the ionization potential of the atom or ion. The ponderomotive potential of the laser is the average kinetic energy of an electron in the laser field given by

$$\Phi_{\text{pond}}(\text{eV}) = e^2\epsilon^2/4m\omega^2 = 9.33\times 10^{-14} I\lambda^2, \tag{2}$$

where ϵ is the electric field strength of the laser and I is in W/cm^2 and λ^2 is in μm.

In the regime of $\gamma > 1$, the ionization can be described as a multiphoton process. An atom or ion simultaneously absorbs N photons:

$$A^{n+} + N\hbar\omega \rightarrow A^{(n+1)} + e^-, \tag{3}$$

where $N\hbar\omega > E_{\text{ion}}$. Most experimental studies of ionization by intense laser fields have been carried out in this regime.[2-8] For example, Rhodes[9] has observed the production of charge states up to Xe^{13+} with a 1-ps, 248-nm laser. Perry *et al.*[3] compared multiphoton ionization in argon, krypton, and xenon and found that the ionization rates for the different charge states were dependent primarily on the ionization potential and less so on the atomic species.

For $\gamma < 1$, the ionization can be described as a tunneling process. Chin and co-workers[10,11] have studied tunneling ionization using a 1-ns, 10-μm, laser and observed the production of charge states up to Xe^{6+}, Kr^{2+}, Ar^{2+}, Ne^{1+}, and He^{1+}.

This Letter reports the first study in the tunneling regime of highly ionized charge states using near optical wavelengths ($\lambda = 1.053$ μm). We have studied the multiple ionization of five noble gases (He, Ne, Ar, Kr, and Xe), and observed charge states as high as Xe^{12+}. For the experiments described here, the laser was operated up to intensities of mid 10^{16} W/cm^2. The ionization occurs for $\gamma < 1$ in all the charge states observed. The intensity at which ionization commences can be well described by summing the Coulomb potential and the electric potential of the laser to find the magnitude of the applied electric field which causes the electron to become unbound.

Our 1.053-μm-wavelength Nd:glass-laser system is based on the concept of chirped pulse amplification.[12] A bandwidth limited, 50-ps, 1.053-μm, input pulse is first produced by a Nd-doped yttrium lithium fluoride oscillator. This pulse undergoes self-phase modulation and group-velocity dispersion in a single-mode fiber and further group-velocity dispersion in a pair of expansion gratings. The dispersed input pulse, with a bandwidth of 30 Å, is a chirped pulse with a duration of 300 ps and an energy of 1 nJ. This pulse is amplified in a three-stage Nd:glass amplifier chain to a final energy in excess of 100 mJ, and is then compressed to $\sim$1 ps by a pair of 1700/mm gratings. The laser can be fired every 30 s.

The laser is focused into a vacuum tank using $f/6$ optics (20-cm focal length lens) which produces peak intensities in the mid 10^{16} W/cm^2 range. The intensity is determined by measuring the energy, pulse width, and focal spot size. The focal spot size is determined by equivalent target plane measurements using 2-m and 15-cm focal length lenses and does not vary significantly on a shot-to-shot basis. The energy of each laser shot is measured by taking a 4% reflection from a beam splitter. Both second- and third-order correlation techniques are used to measure the pulse width.[13-15]

The relative fluctuation of the pulse length is measured on each shot. This is accomplished by frequency doubling a 4% reflection of the main laser pulse. The energy of this green-light signal is divided by the energy of the 1.053-μm signal squared gives a value which is inversely proportional to the pulse length, thus allowing a relative measure of the pulse length on a shot-to-shot basis. The intensities calculated in this manner are consistent with the energy of the ponderomotively accelerated electrons coming from the focus when the focal distribution is taken into account.[16] The uncertainty in the absolute value of the intensity is approximately a factor of 2.

The vacuum chamber is uniformly backfilled with one of the five noble gases, or by a mixture of gases to a pressure of typically 5×10^{-6} Torr. The ions produced during the laser-gas interactions are detected using a time-of-flight spectrometer. An electric field of 800 V/cm is applied across the focal region thereby sweeping the ions into a 30-cm field-free drift tube. The ions are detected with a microchannel plate coupled to a digitizing oscilloscope. The temporal resolution is sufficient for charge states as high as Xe^{13+}.

A typical data run consists of ~200 laser shots in an intensity range from mid 10^{13} to mid 10^{16} W/cm^2. In some cases, data runs were repeated many times and the intensity at which a given number of ions was produced was found to differ by less than 25%. Mixed gas targets were also used to help check the relative intensities among the gases. Therefore, the uncertainty in the relative intensities among data runs is less than 25%. Figure 1 shows a result of a typical run. In this figure, the number of argon ions detected is plotted versus intensity. Similar plots have been generated for He, Ne, Kr, and Xe.

In order to compare our results with those of other researchers, we will follow Perry *et al.*,[3] who defined the "appearance intensity" of a particular charge state as the intensity at which a small number of ions is produced. In this work, the appearance intensity of a charge state is the intensity at which ten ions of that species are detected. In our case, the ten-ion level corresponds to an ionization probability of $\sim5\times10^{-3}$ for the lowest charge states. As can be seen in Fig. 1, the slope of the curve representing the number of ions produced versus intensity is very steep when a small number of ions is detected and so the value of the appearance intensity is insensitive to the exact choice of the number of ions used in defining our appearance level.

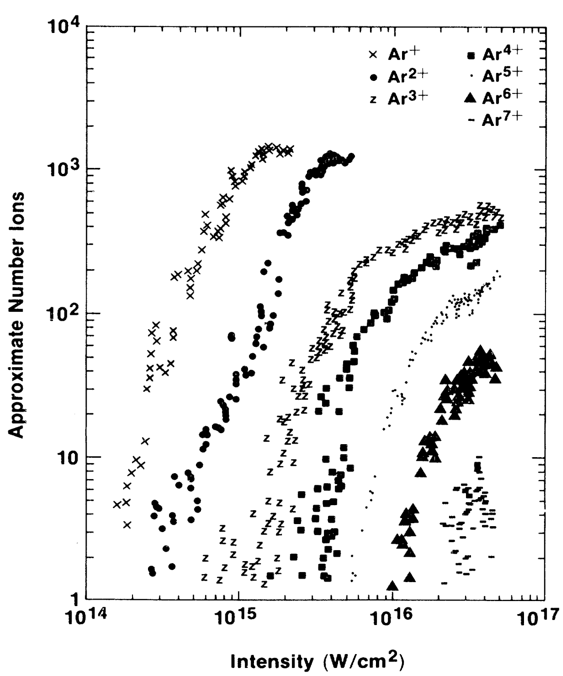

FIG. 1. Approximate number of argon ions detected as a function of peak laser intensity. Similar graphs have been constructed for He, Ne, Kr, and Xe.

Figure 2 shows a plot of the appearance intensities for the observed charge states of ions of the five noble gases versus the ionization potential. There are two points which should be made regarding this graph. First, the appearance intensities show a clear species dependence. The appearance intensity of each of the noble gases follows a smooth curve versus ionization potential, but these curves do not overlap. We conclude that the value of the appearance intensity is dependent both on the ionization potential and on the atomic species. This is in contrast to the observations of Perry *et al.*, who observed that in the nontunneling regime of $1<\gamma<4$, the appearance intensities were dependent primarily on the ionization potential.

The second point of interest is that the observed appearance intensities are much higher than those previously seen at this wavelength.[2] The difference between these data and those shown in Ref. 2 is as much as a factor of 10 depending on the charge state. It should be emphasized that this factor of 10 cannot be due to experimental error as the absolute error in the intensity measurement is estimated to be a factor of 2 at most.

The ionization experiments described in Ref. 2 were performed using a 50-ps bandwidth-limited laser pulse, in contrast to our 1-ps non-bandwidth-limited pulse. By shortening the laser pulse length and increasing the bandwidth, the ionization process is pushed from the multiphoton regime into the tunneling regime. Details

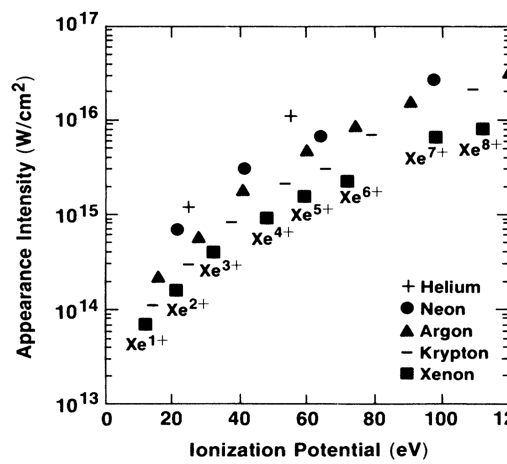

FIG. 2. Ion appearance intensity vs sequential ionization potential for five noble gases. There is a clear species dependence present. The relative error in the laser intensity is approximately the size of the data boxes.

concerning this transitional regime are not yet understood and experiments to study it are under way.

The appearance intensities are predicted quite well by a simple theoretical model which consists of a superposition of the Coulomb potential with a static electric field.[17] As the external field strength is increased, the height of the Coulomb barrier is suppressed. Ionization occurs when the amount of suppression is equal to the ionization potential of the atom or ion. The superimposed potential is written as

$$V(x) = -Ze^2/x - e\epsilon x\,. \tag{4}$$

The position of the barrier maximum, $x_{\max}$, is found by setting $\partial V(x)/\partial x = 0$. Equating $V(x_{\max})$ to the ionization potential, E_{ion}, yields an expression for the critical electric field strength

$$\epsilon^2 = E_{\text{ion}}^4/16e^6Z^2\,. \tag{5}$$

Setting this static electric field equal to the peak electric field of the laser, we obtain a corresponding intensity which can be considered the appearance intensity,

$$I_{\text{app}} = cE_{\text{ion}}^4/128\pi e^6Z^2\,. \tag{6}$$

The species dependence is contained in the known values of the ionization potentials which, in a purely Coulombic system, would be a function of Z alone.

Figure 3 shows a plot of these calculated appearance intensities versus the experimental appearance intensities. Using no free parameters, excellent agreement is obtained over almost three decades of intensity for all charge states. Furthermore, the fits produced by this simple model predict the appearance intensities for all five gases more accurately than complicated theories which are commonly used to describe high-intensity laser-atom interactions such as Keldysh theory[1] and modified Keldysh-Faisal-Reiss theory.[3]

The appearance intensities were also compared with the complex-atom tunneling ionization theory of Ammosov, Delone, and Kraĭnov.[18] While all of the appearance intensities are matched within the factor of 2 absolute uncertainty in the experimental intensity, the deviation between experimental and predicted intensities systematically increases with decreasing atomic weight.

In summary, we have shown that for a laser wavelength of 1 μm and pulse length of 1 ps, the ionization of noble gases occurs in the tunneling regime. The ionization is species dependent and the appearance intensities are as much as a factor of 10 higher than those seen with pulse lengths of 50 ps at the same wavelength. Further, the appearance intensities can be quite accurately predicted using a simple Coulombic-barrier model. Future work will involve an investigation of the transition regime between 50 and 1 ps where the ionization changes from a multiphoton process to a tunneling process.

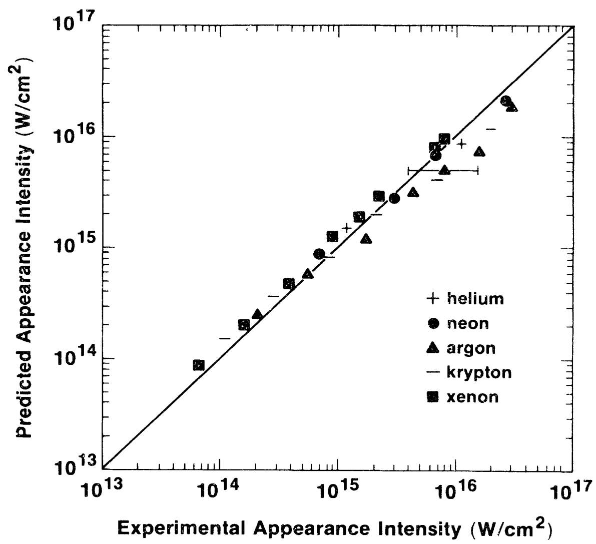

FIG. 3. Comparison of experimentally determined and theoretically predicted appearance intensities using a simple classical theory. The solid line corresponds to exact agreement.

The authors would like to acknowledge the assistance of S. Batha, C. I. Moore, and M. C. Richardson in the experiments, and useful discussions with J. Knauer, A. Szöke, M. D. Perry, P. B. Corkum, and N. H. Burnett. We also would like to acknowledge the work of G. Mourou, P. Bado, and P. Maine in the development of the laser system. This work was supported by the National Science Foundation Grant No. PHY-8822730. Additional support was provided by the U.S. Department of Energy Division of Inertial Fusion under Agreement No. DE-FC03-85DP40200 and by the Laser Fusion Feasibility Project at the Laboratory for Laser Energetics which has the following sponsors: Empire State Electric Energy Research Corporation, New York State Energy Research and Development Authority, Ontario Hydro, and the University of Rochester.

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