Observation of a Transition in the Dynamics Of Strong-Field Double Ionization

J. L. Chaloupka,[1,*] J. Rudati,[1,†] R. Lafon,[1] P. Agostini,[2] K. C. Kulander,[3] and L. F. DiMauro[1]

[1]*Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973*
[2]*SPAM, Centre d'Etudes de Saclay, 91191 Gif Sur Yvette, France*
[3]*TAMP, Lawrence Livermore National Laboratory, Livermore, California 94551*

(Received 17 October 2002; published 24 January 2003)

The double ionization of argon and xenon in an intense laser field has been studied in detail using an electron-ion coincidence technique. The observed double ionization electron spectra in xenon show resonancelike structures here resolved for the first time. In argon, the featureless spectra are consistent with rescattering. This represents a clear transition in the dynamics of strong-field double ionization, analogous to the well-known transition between the tunneling and multiphoton regimes in single ionization.

DOI: 10.1103/PhysRevLett.90.033002 PACS numbers: 32.80.Rm, 31.90.+s, 32.80.Fb

The liberation of two electrons from helium following the absorption of a large number of linearly polarized visible photons is one of the fundamental many-body processes in atomic physics. Experiments have observed a dramatic enhancement in the double ionization yields across a relatively low intensity range [1,2], suggesting direct two electron ejection. The enhanced double ion yields are several orders of magnitude larger than those expected from the independent, successive removal of electrons from the ground states of the neutral atom and ion. This so-called ''nonsequential'' ionization is most dramatic in helium, but was first observed in xenon [3], and is universal among all of the rare gases, appearing well below the single ionization saturation intensity.

Single ionization of atoms by strong laser fields is often discussed in terms of either tunneling or multiphoton ionization. The boundary between these two limits can be defined by the ratio γ of the tunneling time to the optical period. Known as the adiabaticity or Keldysh parameter [4], this ratio equals $(I_p/2U_p)^{1/2}$, where I_p is the atomic ionization potential and U_p is the ponderomotive potential [5] associated with the laser field. By varying the laser intensity and the target atom species, a range of γ values can be accessed, thereby helping to identify the important mechanisms of strong-field single ionization [6,7]. For small γ, tunnel ionization is important, and the shape of the photoelectron distribution is largely determined by the classical propagation of the electron in the laser field. For large γ, multiphoton ionization dominates, giving rise to atomic resonance structure and above-threshold ionization peaks in the photoelectron spectra. The validity of this simple Keldysh picture of single electron dynamics has been verified by many experiments as well as by more sophisticated theoretical treatments [8]. Until now, an analogous evolution in ionization behavior has not been observed or tested in double ionization.

Recently, this double ionization process has been studied by several research groups using experimental probes beyond the simple collection of total ionization yields. Using cold target recoil ion momentum spectroscopy (COLTRIMS), the ion recoil momenta of Ar^{2+} [9], Ne^{2+} [10], He^{2+} [11], as well as the electron momenta correlated to the double ionization of argon [12–14], have been compiled. In addition, electron-ion coincidence spectroscopy has been used to measure the electron spectra correlated to the double ionization of xenon [15,16], helium [17], and argon [18], as well as for the rare gases spanning He to Xe [19]. All of these studies have at least partially supported the rescattering scenario [20,21], where one electron tunnels through the distorted Coulomb potential, propagates in the laser field, and then is driven back to the core leading to impact ionization [21]. A large amount of theoretical work has also supported this scenario [22]. While this recent work has created a strong framework for double ionization in the tunneling regime, the mechanisms for multiphoton double ionization have remained unclear. The formation of a complete picture of strong-field double ionization has been hindered by the lack of experimental work performed within the multiphoton regime, as well as by the inadequate energy resolution inherent to recoil spectroscopy.

In this Letter, we report on the first observation of the transition from the tunneling/rescattering to the multiphoton regimes in strong-field double ionization. Although it is not feasible to explore this transition in a single atomic species with the coincidence technique used here, by obtaining the spectra from two different atoms the progression can be followed. Therefore through the use of a high-resolution electron-ion coincidence technique, we have compiled the electron momentum spectra correlated to the double ionization of argon and xenon at a total of four different laser intensities. The observed electron momenta from the double ionization of argon agree well with previous experiments and are consistent with the rescattering model. The observed spectra from the double ionization of xenon are not consistent

with rescattering and exhibit distinct multiphoton features. While the details of the multiphoton double ionization transitions are still not uncovered, the observed structures definitely suggest some form of sequential processes at work.

The electron-ion coincidence technique used for this study is described in detail elsewhere [17,19], but is outlined briefly below. The 780-nm, 100-fs output from a titanium:sapphire laser system was focused into an ultrahigh vacuum chamber at a repetition rate of 2 kHz. The laser pulse energy was adjusted for the required peak intensity, and the target gas pressure was set for the appropriate ion count rate. A two-sided, pulsed-plate spectrometer was used to measure the electron time of flight as well as the ion charge to mass for each laser shot. With this technique, the electron spectra correlated to a specific ion species can be generated. Depending upon the detection efficiencies and the ionization count rate, the number of true coincidence events (where the detected ion and electron are positively correlated) versus false coincidence events can be determined [23]. In our study, the ion and electron detection efficiencies were roughly 30% and 1%, respectively, and the electron detection was restricted to a 10° cone angle about the polarization axis. The ionization rates were kept at one ion detection per four laser shots. This gave a calculated ratio of true to false coincidence events of 4:1, meaning that 80% of the detected electrons were positively correlated to the detected ion. By running under reproducible experimental conditions for many hours over several data-taking sessions, it was possible to compile electron spectra from a large number of double ionization events. The high statistics achieved in this study were especially important in determining the presence of the narrow electron energy structures related to multiphoton ionization.

Figures 1(a) and 1(b) show the ratio of double to single ionization yields for xenon and argon, respectively. The telltale "knee" feature in the curves indicates the presence of an additional double ionization rate occurring at low intensities. The shaded areas show the regions of enhanced double ionization, and the arrows indicate the intensities at which data were taken for this study.

Figure 2 shows the electron momentum spectra correlated to single (shaded area) and double (solid lines) ionization. Each spectrum is normalized to unity integrated value and is plotted at a resolution of 0.01 a.u. of momentum. Figure 2(a) shows the spectra taken with xenon at an intensity of 40 TW/cm^2. A total of 4.3×10^8 laser shots were taken, yielding 5276 electron counts correlated to double ionization. Figure 2(b) shows the spectra taken with xenon at 80 TW/cm^2 (2.1×10^8 shots, 12 734 Xe^{2+} counts). Figure 2(c) shows the spectra taken with argon at 100 TW/cm^2 (6.9×10^8 shots, 10 664 Ar^{2+} counts). Figure 2(d) shows the spectra taken with argon at 200 TW/cm^2 (2.7×10^8 shots, 12 110 Ar^{2+} counts). All of the spectra are shown uncorrected for false counts. Correcting for the contribution from false coincidences results in small changes to the details of the double ionization spectra, but does not affect the overall structure and features in the spectra.

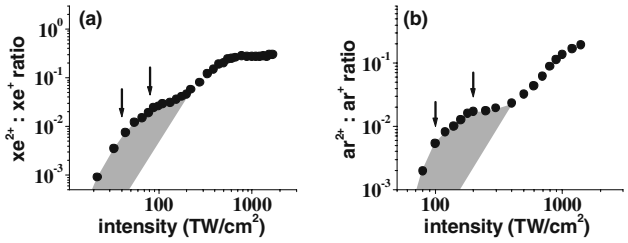

FIG. 1. The ratio of double to single ionization counts as a function of intensity for (a) xenon and (b) argon.

The spectra corresponding to the smallest value of the Keldysh parameter ($\gamma = 0.8$) were taken at 200 TW/cm^2 in argon [Fig. 2(d)]. Here the electrons correlated to double ionization are pushed out to higher momenta with respect to the single ionization electrons. The median energies are 5.2 and 11.1 eV for the electrons from single and double ionization, respectively. This is to be expected within the rescattering picture, and agrees well with results from COLTRIMS experiments [12]. The high-resolution, high-statistics data presented here show the absence of multiphoton structure in the double ionization electron spectrum, while such structure persists in the single ionization distribution.

This double ionization spectrum convincingly identifies the dynamics involved in tunneling followed by rescattering to free the two electrons. It is the nature of rescattering to broaden the electron energy distributions. Following the inelastic collision with the ion core the first liberated electron is scattered into a broad range of angles. The subsequent propagation of the electron in the laser field will determine the final, observed energy, which will be a strong function of its momentum at impact, the phase of the laser field, and the scattering angle. In the case of impact ionization, the final energy of the second electron will vary in a similar way. The distribution of the excess energy between the two exiting electrons will be continuous, eliminating any structure in the spectra. (We note that the single ionization spectrum in the tunneling regime has been calculated to maintain its ATI structure, but this has not been observed experimentally. This most likely is due to the spatial averaging over the intensity distribution within the focal volume washing out the peaks [24].) The energetic recollisions that produce excitation or ionization of the ion core occur near a node in the field strength. If the collision is inelastic, the excited state produced will be field ionized before the field reaches its next maximum, i.e., within a quarter of a cycle. Thus the uncertainty in its energy will again produce an unstructured distribution. It is the value of the Keldysh parameter associated with the single ionization

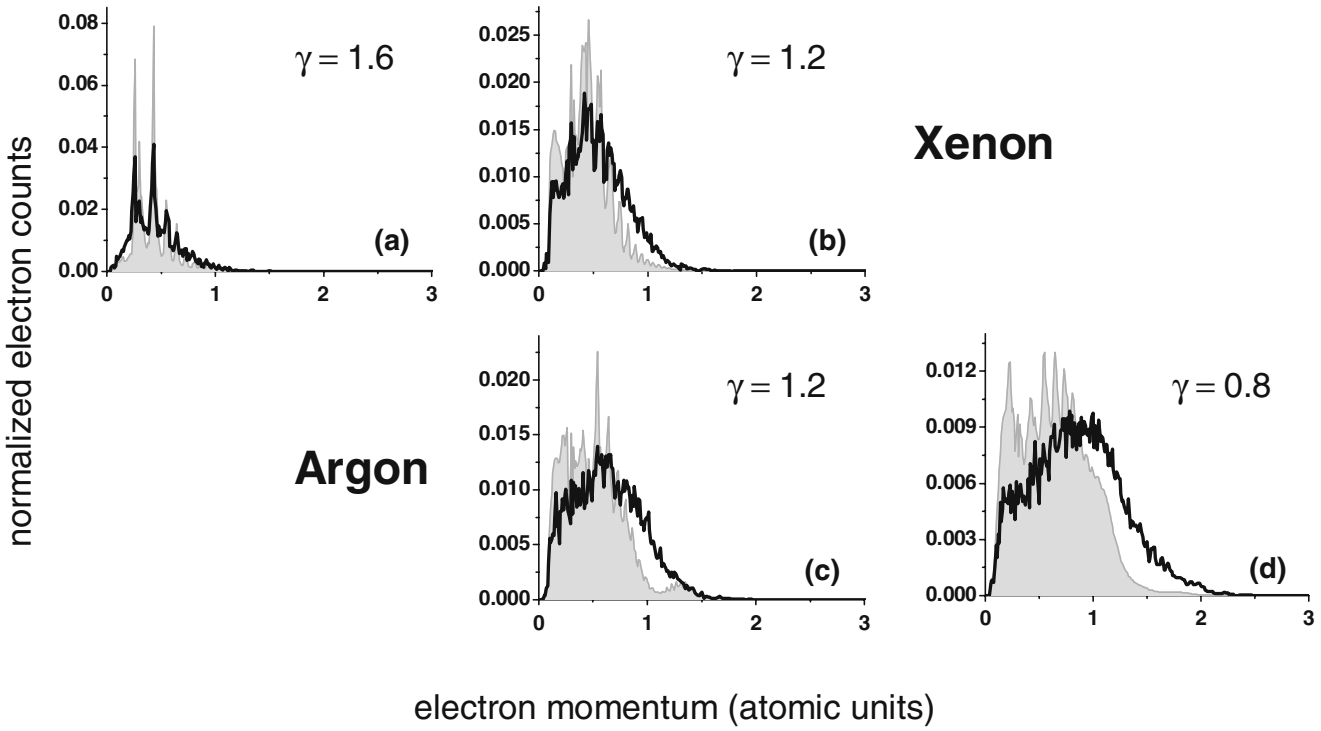

FIG. 2. The electron momentum spectra correlated to single (shaded areas) and double (solid lines) ionization for xenon at (a) 40 and (b) 80 TW/cm^2 and argon at (c) 100 and (d) 200 TW/cm^2. The Keldysh parameter is indicated for each plot.

step, which here indicates that tunneling is becoming dominant, that establishes the dynamics of the nonsequential ionization. The energy spectrum reinforces this conclusion.

Figure 2(a) shows the spectra taken in xenon at 40 TW/cm^2. Here the Keldysh parameter equals 1.6 and represents the data taken closest to the multiphoton limit. There is no apparent enhancement at high electron momenta in the double ionization electron spectra, and clear structure is observed. In fact, the median electron energies are nearly equal for single and double ionization (2.31 eV versus 2.35 eV). This is not consistent with rescattering, which is, in fact, classically forbidden. The first liberated electron has a maximum kinetic energy ($E_{\max}$) equal to $3.2U_p$ when it reencounters the core. Here $E_{\max}$ equals only 7.2 eV, which is not only well below the 21.2 eV ionization potential of Xe^+, it is even below the 11.3 eV Xe^+ first excited state. As a result, the quasiclassical picture of rescattering cannot lead to impact ionization, or even to ionization through excitation. The electron energy distributions from both single and double ionization exhibit the strong energy structure expected from multiphoton ionization.

For the intermediate Keldysh values ($\gamma = 1.2$), there are some indications of both rescattering and multiphoton effects. The spectra taken at the higher intensity in xenon [Fig. 2(b)] and the lower intensity in argon [Fig. 2(c)] both show an enhancement at higher electron momenta in the double ionization spectra, as well as some indications of energy structure. The enhancement at high momenta is weaker than in the high intensity case of argon since the maximum return energy of the electrons is smaller. In the case of argon at 100 TW/cm^2, $E_{\max}$ equals 18.0 eV. Even though this is insufficient for direct impact ionization, it is large enough to lead to excitation to the 13.5 eV first excited state of Ar^+. Any excess energy can be carried away by the first electron, and the second electron, in the excited state, can then be readily field ionized. The specifics of the energy sharing and subsequent propagation in the laser field can lead to a broad range of final electron momenta. A similar scenario can occur in the case of xenon at 80 TW/cm^2. Here $E_{\max}$ equals 14.4 eV, which is below the 21.2 eV ionization potential of Xe^+, but above the 11.3 eV first excited state.

While simple energy arguments can exclude the contribution from rescattering in the double ionization of xenon at the low intensity [Fig. 2(a)], the details of the ionization process are still unclear. The most likely multiphoton scenario is simply resonantly enhanced sequential ionization. Just as the production of Xe^+ is enhanced by Stark-induced Rydberg (Freeman) resonances, the production of Xe^{2+} could be enhanced by ionic Freeman resonances. Here the double ionization electron spectrum would represent an even mix from the uncorrelated "first" and "second" electrons. The observed structure would reflect the resonances of the ground state of

the neutral atom as well as of the ion. However, identifying the intermediate states can be difficult. The enhancement via high-lying (Rydberg) resonances in the ion decreases significantly because of the large number of photons, N, required to reach them. For small N, the resonant enhancement of a $(N-1)$ process is large. As a result, a resonance through a high-lying state can have a significant impact on the total ion yields. As N increases (e.g., for double versus single ionization), the effect of a $(N-1)$ resonance is reduced, and may, in fact, become negligible. A second type of resonant enhancement involving an ion core transition can be low enough order to strongly affect the sequential rate [25]. In Ar there is no suitable candidates for this mechanism, but in Xe, the $5s^2 5p^5 \rightarrow 5s5p^6$ lies approximately seven photons above the ground state of the ion and could very well be responsible for the observed enhancement and the narrow structures in the energy distributions.

A final point of speculation: the observed double ionization spectrum at the low intensity in xenon [Fig. 2(a)] shows the presence of a broad "continuum" beneath the sharp structures. The origin of this feature is not obvious. The classical picture of rescattering cannot contribute simply due to the very low electron return energies. A modified rescattering model, or perhaps some other form of electron correlation, may be used instead. The size, shape, and position of this feature, as well as of the sharp peaks, should aid in distinguishing between the viability of future models.

In conclusion, the electron spectra correlated to the single and double ionization of xenon and argon have been recorded. The use of a high-resolution electron-ion coincidence technique reveals distinct multiphoton structure in xenon, observed for the first time in strong-field double ionization. Along with the broad featureless double ionization spectrum observed in argon, these results illustrate a dramatic and previously unknown transition in double ionization dynamics, analogous to the single ionization evolution related to the Keldysh parameter. The double ionization of argon is consistent with rescattering, while the double ionization of xenon appears to involve a more complex, multiphoton process.

The experiments were carried out at Brookhaven National Laboratory under Contract No. DE-AC02-98CH10886 with the U.S. Department of Energy and supported by its Division of Chemical Sciences, Office of Basic Energy Sciences. J. R. acknowledges support from the W. Burghardt Turner Foundation.

*Present address: Department of Physics, College of William & Mary, Williamsburg, VA 23187-8795.
†Present address: Department of Physics, SUNY at Stony Brook, Stony Brook, NY 11794-3362.

[1] D. N. Fittinghoff *et al.*, Phys. Rev. Lett. **69**, 2642 (1992).
[2] B. Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994).
[3] A. l'Huillier *et al.*, Phys. Rev. A **27**, 2503 (1983).
[4] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[5] U_p is the cycle-averaged kinetic energy associated with a free electron in a laser field. U_p measured in eV is given by $0.093 I\lambda^2$ where I is the intensity in TW/cm^2 and λ is the wavelength in μm. U_p is 5.66 eV for 0.78 μm excitation at 100 TW/cm^2.
[6] E. Mevel *et al.*, Phys. Rev. Lett. **70**, 406 (1993).
[7] B. Walker *et al.*, Phys. Rev. A **48**, R894 (1993).
[8] See, for example, *Atoms in Intense Laser Fields*, edited by M. Gavrila (Academic, Boston, 1992), and references therein.
[9] Th. Weber *et al.*, J. Phys. B **33**, L127 (2000).
[10] R. Moshammer *et al.*, Phys. Rev. Lett. **84**, 447 (2000).
[11] Th. Weber *et al.*, Phys. Rev. Lett. **84**, 443 (2000).
[12] Th. Weber *et al.*, Nature (London) **405**, 658 (2000).
[13] B. Feuerstein *et al.*, Phys. Rev. Lett. **87**, 043003 (2001).
[14] M. Weckenbrock *et al.*, J. Phys. B **34**, L449 (2001).
[15] B. Witzel, N. A. Papadogiannis, and D. Charalambidis, Phys. Rev. Lett. **85**, 2268 (2000).
[16] J. L. Chaloupka *et al.*, Acta Phys. Pol. A **101**, 337 (2002).
[17] R. Lafon *et al.*, Phys. Rev. Lett. **86**, 2762 (2001).
[18] E. R. Peterson and P. H. Bucksbaum, Phys. Rev. A **64**, 053405 (2001).
[19] J. L. Chaloupka *et al.*, Opt. Express **8**, 352 (2001).
[20] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. **70**, 1599 (1993).
[21] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[22] See, for example, the special issue of Opt. Express **8** (2001), and references therein.
[23] V. Stert *et al.*, Eur. Phys. J. D **5**, 97 (1999).
[24] B. Sheehy *et al.*, Phys. Rev. A **58**, 3942 (1998).
[25] D. Charalambidis *et al.*, Phys. Rev. A **50**, R2822 (1994).