Charge-State Resolved above Threshold Ionization

B. Witzel,* N. A. Papadogiannis, and D. Charalambidis†

Foundation for Research and Technology–Hellas, Institute of Electronic Structure and Laser, Laser and Applications Division, P.O. Box 1527, 711 10 Heraklion, Crete, Greece

(Received 2 February 2000)

Photoelectron energy resolved spectra recorded in coincidence with singly and doubly ionized atoms are reported for the interaction of xenon with 60 fs laser pulses at 800 nm and 1×10^{14} W/cm^2. Double ionization contributions in the spectrum have been established. The appearance of a single ionization plateau in above threshold ionization is verified. A hotter electron distribution is found for the double ionization.

PACS numbers: 32.80.Gc, 32.80.Rm, 39.30.+w

Short pulse high intensity laser atom interactions have recently led to the observation of several novel phenomena of general interest. One of the most frequently discussed is the so-called nonsequential ejection of two or more electrons from an atom or molecule [1]. In single photon ionization double ionization occurs via simultaneous ejection of two correlated electrons. In multiphoton double ionization the question of whether the two electrons are ejected one after the other (sequentially), producing first a singly ionized atom which further ionizes, or simultaneously (nonsequentially), has been an open problem since the 1970's. A few years ago multiphoton nonsequential double ionization again received enhanced attention because of its observation in atomic He [2]. Above threshold ionization (ATI), studied through high-resolution photoelectron spectroscopy [3,4], has been one of the main tools used in this research field. It has been considered to be an appropriate technique for the investigation of the mechanism of double ionization, but it has also led to several other novel observations. In the long wavelength regime nonperturbative effects observed in ATI spectra include the so-called plateaus, which are areas of the electron energy spectrum where the spectrum envelope flattens instead of dropping [5], side lobes in photoelectron angular distributions [6], and conventionally unexpected ellipticity dependence of the electron yields [7]. Most of these effects have been interpreted within the frame of the rescattering model in quantum-mechanical or classical trajectory calculations [8]. Calculations of ATI spectra are mainly in the single active electron approximation [1,2,5], with a few exceptions that include multiple continua but not double ionization [9]. Experiments with rare gases in the short wavelength regime (5 eV photons and 0.5 ps pulse duration) have in contrast shown that changes in the envelope slope of the ATI spectrum [10], but also of the higher order harmonic generation spectrum [11], can be attributed to the contribution of the ion interaction with the field, especially for laser intensities above the saturation intensity of the single ionization. However, the ionic contribution has been only indirectly evaluated in the short wavelength regime. In studies at long wavelengths it is not taken into account, although from ion yield measurements we know that for some atoms the double ionization yield at an intensity around 10^{14} W/cm^2, at which the studies have been performed, is of the order of 10% [12] of the total yield. Other experiments in two valence electron systems have also shown evidence of double ionization contribution in short pulse ATI spectra [13]. Double or multiple ionizations remain in most of the cases hidden and ignored in the ATI spectra. Charge-state resolved ATI measurements are the most direct approach toward distinguishing the contributions of multiple ionization and thus learning about their characteristics and production process. Very little is known presently about the electron energy and angular distribution spectrum that correlates with strong field double ionization in contrast to the rich information that is available for low intensity single-XUV-photon double ionization and its associated electron-electron correlation effects. This type of information is important for the fundamental understanding of the multiple ionization process and the evaluation of existing models. It further provides useful parameters for other fields, such as electron temperatures of the different ionic contributions, which may serve as input to numerical codes modeling short pulse high intensity plasma generation. Earlier interpretations of ATI spectra, ignoring multiple ionization, are subject to verification, and this can be accomplished through charge-state resolved ATI spectroscopy.

In the present work we report electron-ion coincidence measurements in Xe interacting with 60 fs laser pulses at 800 nm. Charge-state correlated energy spectra of photoelectrons clarify the origin of plateau regions in the total ATI spectrum, establish in a direct manner the contribution of the double ionization in the total ionization spectrum, and provide comparative information about the electron temperatures correlating with different charge states, as well as some limited information about the angular correlation of the electrons originating from a double ionization process and its relation to the rescattering model. To our knowledge this is the first charge-state correlated ATI investigation that touches upon all of the above aspects and is complimentary to other detailed and sophisticated investigations based on ion-recoil spectroscopy [14].

 0031-9007/00/85(11)/2268(4)$15.00

The experimental setup includes the laser, the beam delivery unit, and the electron-ion coincidence spectrometer. The setup is described in detail elsewhere [15], the only difference being the size of the spectrometer detector. The laser consists of a titanium sapphire oscillator amplified in a regenerative amplifier utilizing a chirped pulse amplification technique [16]. The laser operates at $\sim$1 kHz repetition rate and outputs at maximum 1 mJ of energy. The laser is through a $\lambda/2$ plate linearly polarized parallel to the spectrometer axis. The beam is focused through a 15 cm focal length lens into the interaction region of the spectrometer. For the coincidence measurements a single "time of flight" spectrometer is used. The spectrometer background pressure is lower than 10^{-9} mbar. The same 28 cm flight tube and a 7 cm diam microsphere-plate (MSP) detector are used for the ionic m/q, as well as for the electron energy analysis. The angle of acceptance for the electron measurements is $\sim$14°. This angle allows the detection of a few percent of the total electron yield. Electrons move first field free and are subsequently accelerated by 500 V shortly before they reach the detector biased at 3500 V in order to achieve maximum detection efficiency. 1 μs after the laser atom interaction, a 200 ns long electric voltage pulse applied in the interaction region accelerates the ionic species, which subsequently enter the field-free region of the spectrometer. The pulse height is 8000 V in order to maximize the ion detection efficiency. The absolute detection efficiency of the MSP detector is estimated to be about 50% for the electrons and the ions. With the arrangement used the recorded traces contain both the electron and the ion time of flight spectrum well separated in time. The electron spectrum appears at short times ($<$0.5 μs) before the ion spectrum that starts at times longer than 1 μs. The MSP detector signal passes through a pulse height discriminator and is recorded shot to shot via a multievent time-digitizer card, as a single shot temporal trace. The card has a temporal resolution of 0.5 ns and is connected to a personal computer in which data are trace by trace transferred and stored.

The experimental parameters used in the present work are summarized in the following. Using 30 μJ of the laser energy, the focused intensity in the interaction region is $\sim 1 \times 10^{14}$ W/cm^2. The Xe gas pressure in the spectrometer is maintained at less than 3×10^{-9} mbar. Under these conditions the maximum ionization detection rate is 10 Hz, i.e., one detected ionization event per 100 laser pulses. This is done in order to ensure pure single event detection conditions, which is unavoidable in clean coincidence measurements. This rate assures single event detection conditions [15,17]. It should be noted that in the intensity region at which the experiment is performed the Xe^{++}/Xe^{+} ion yield ratio depends only weakly on the intensity [15], which indicates that the nonsequential process dominates the double ionization procedure [18].

Figure 1 summarizes the experimental results. Figure 1(a) shows the total ATI spectrum (continuous line),

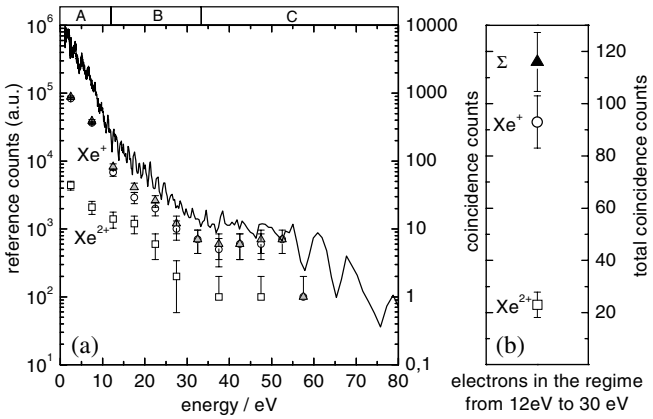

FIG. 1. Above threshold ionization spectra measured at 10^{14} W/cm^2. Total ATI spectrum (continuous line); energy distribution of electrons correlating with single ionization (circles); energy distribution of electrons correlating with double ionization (squares); sum of the last two (triangles) (a). Total number of counts corresponding to electrons with energies between 12 and 30 eV correlating with single ionization (circle), double ionization (square), and the sum of the two (triangle) (b).

energy resolved electrons correlating with single ionization (circles), energy resolved electrons correlating with double ionization (squares), and the sum of the last two categories (triangles). False coincidence can be estimated from the given MSP detection efficiency for the ions and the electrons, and from the measured ratio of the total number of electrons to the total number of ions [17]. The maximum calculated error originating from false coincidences is less than 3% and is included in the error bars of Figs. 1(a) and 1(b), together with the statistical 1 standard deviation.

The total spectrum is recorded under no coincidence conditions at a Xe pressure of 10^{-7} mbar in order to obtain a spectrum with good statistics that will be the base of comparisons with the coincident spectra. It can be divided into three energy regimes showing qualitatively different envelopes. The first (region *A*) extends up to 12 eV depicting a rapidly decreasing envelope of the ATI. This is also the only region of the spectrum where ATI peaks separated by one photon energy (1.55 eV) can be found. The second (region *B*) ranges from 12 to 30 eV. Its envelope decreases less rapidly with energy and the ATI structure is not maintained. For energies higher than 30 eV (region *C*) the envelope becomes almost horizontal before it drops to zero above 50 eV. This is a familiar plateau picture. In this region the combination of low statistics, reduced resolution of the spectrometer, and increased ATI peak width does not allow the observation of resolved ATI structure.

The yield of electrons correlating with singly ionized atoms (open circles) is also rapidly decreasing by more than 1 and $\frac{1}{2}$ orders of magnitude up to an energy of 18–20 eV, while for the next 20 eV of kinetic energy ($\sim$18–38 eV) the decrease of the yield reduces to $\frac{1}{2}$ order of magnitude, thus creating a plateau region similar to the one known from total ATI spectra. This is an important

result that unambiguously verifies the existence of different envelope slopes (plateaulike structures) in the ATI of single ionization independently from the contribution of the double ionization process at the given laser parameters. It verifies previous interpretations [5], thus clarifying a question concerning the origin of plateaus in ATI that has been answered unambiguously only for one valence electron systems such as hydrogen [19] and alkalines [20] up to now. Peak structure is observable only in the beginning of the spectrum and will be discussed below.

At the present laser parameter electrons correlating with doubly ionized atoms (squares) have a main contribution to the total spectrum in the energy region between 12 and 30 eV. This contribution is approximately 20% of the total electron production as shown in Fig. 1(b), while between 20 and 30 eV it increases to about 30%. A comparison of the coincident distributions with the total spectrum strongly indicates that electrons correlating with the double ionization are those forming the envelope region *B* in the total spectrum. One could further argue that they are responsible for the absence of regular ATI peaks in this region. Such an absence of ATI peaks in the beginning of plateaus as well as peak positions not corresponding to an exact integer number of photons has been observed in previous works [5]. A further comparison of the two electron distributions is shown in Fig. 2. Here each distribution is normalized to unity. The decrease of the yield with energy in the distribution of the electrons correlating with Xe^+ is more rapid. Electrons correlating with Xe^{2+} appear hotter than those produced via single ionization. The mean electron energies are 6 and 8.6 eV for electrons measured in coincidence with singly and doubly ionized Xe, respectively. Energy transfer from the field to the electrons is more efficient during the double ionization process as would be expected by the fact that a larger percentage of the atoms interacts with higher intensity parts of the laser pulse in order to doubly ionize.

Up to this point the general characteristics of the envelope of the spectra have been presented since this is the main information that can be gained from the coincidence spectra. Detailed peak structure is observable in the coincidence spectra only at their low energy region below 5 eV. This low energy region for the spectra of Fig. 1(a) is shown expanded in Fig. 3. The total spectrum depicts a series of broad peaks separated by the photon energy. Their width can be accounted for through the ponderomotive shift of the ionization threshold. Transient resonances therein cannot be excluded. The two fine structure levels of Xe^+ are not resolved in the spectrum because of this broadening. In addition to this main series there is a well-resolved second series of narrower peaks, the first of which appears at 2.5 eV. This secondary series cannot be attributed to double ionization since its abundance is higher than that of the double ionization and its position does not correspond to any structure in the electron spectrum correlating with double ionization. It can be attributed to transient resonace(s) shifted into resonance. For the electrons emitted through single ionization (circles) the first three ATI peaks are resolved, while for the electrons correlating with double ionization (squares) only one peak is observable in the beginning of the spectrum because of the low number of recorded events. Its position appears slightly shifted with respect to the peak of the single ionization electrons.

One further attempt of the present experiment concerns the correlated electron angular distribution in the double ionization of Xe. Setting the single trace coincidence condition to be two electrons and one Xe^{2+} ion, it is possible to investigate whether the two electrons produced by the double ionization are emitted in the same direction, within the acceptance angle of 14° of the spectrometer or not. If more than 50% of the emission of both electrons would be in the same direction and within the above acceptance angle, the number of the multievent time-digitizer card traces satisfying this coincidence condition should not be smaller than 13, which is about 13% of the number of

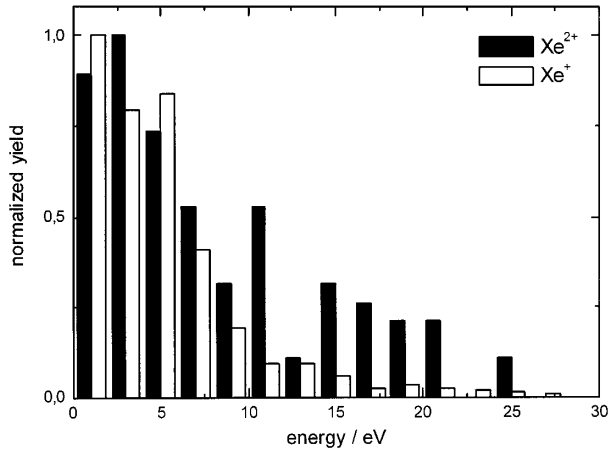

FIG. 2. Energy distribution of electrons correlating with single ionization (open bars) and double ionization (black bars) normalized to unity. The later distribution appears hotter.

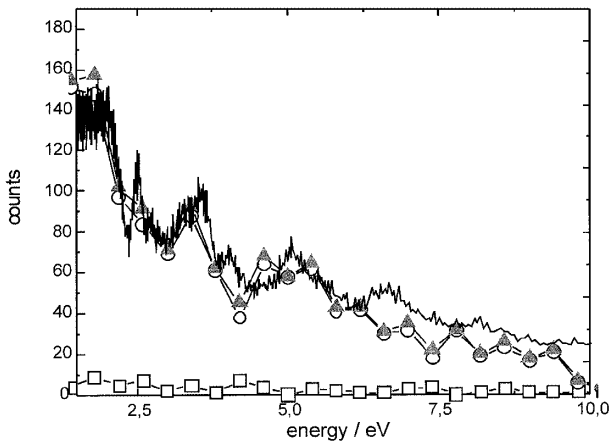

FIG. 3. As in Fig. 1(a) for the expanded low energy region up to 15 eV, where peak structure is observable.

traces containing one electron and one Xe^{2+} ion, for the given MSP efficiency of the electrons and the ions. The measured number of traces satisfying the above triple coincidence condition are 3, i.e., less than 3% of the traces containing one electron and one Xe^{2+} ion. This result indicates that, with the present experimental conditions, for the majority (~88%) of the double ionization processes the electrons are emitted in directions lying more than 14° apart, but the statistics do not allow conclusive statements about the angular correlation of the two emitted electrons.

In conclusion, the present electron-ion coincidence measurements in the long wavelength regime give unambiguous evidence for the existence of single ionization ATI plateau structures, in contrast to the ATI at shorter wavelengths (248 nm) and longer pulse durations (0.5 ps) where plateaulike regions appear above the saturation of the single ionization due to sequential double ionization. The present work verifies, on the basis of experimental evidence, the earlier interpretation of such features as a nonperturbative effect in the short pulse ionization process. However, double ionization produces a hotter than single ionization electron distribution, which is visible in the total ATI spectrum. It contributes to the general picture of the ATI spectrum producing additional changes of the ATI envelope, in the form of intermediate plateaus.

This work has been carried out in the Ultraviolet Laser Facility of FORTH with partial support from the TNR Program of the European Union (Contract No. ERBFMGECT950017). Helpful discussions with G. G. Paulus and C. Kalpouzos are greatly acknowledged. One of us (B. W.) gratefully acknowledges support from the Alexander von Humboldt–Stiftung No. 11461.

*Current address: University of Freiburg, Department of Molecular and Optical Physics, Hermann-Herder-Strasse 3, D-79104 Freiburg, Germany.
Email address: witzel@physik.uni-freiburg.de
†Also at Department of Physics, University of Crete, P.O. Box 2208, 71003 Heraklion, Crete, Greece.

[1] P. Lampropoulos, P. Maragakis, and J. Jhang, Phys. Rep. **305**, 203 (1998), and references therein; M. Protopapas, C. H. Keitel, and P. L. Knight, Rep. Prog. Phys. **60**, 389 (1997), and references therein; A. Becker and F. H. M. Faisal, J. Phys. B **32**, L335 (1999).
[2] B. Walker, B. Sheehy, L. F. DiMauro, P. Agostini, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **73**, 1227 (1994); A. Talebpour, C-Y Chien, and S. L. Chin, J. Phys. B **29**, 5725 (1996).
[3] P. Agostini, F. Fabre, G. Mainfray, G. Petite, and N. K. Raman, Phys. Rev. Lett. **42**, 1127 (1979).
[4] L. F. DiMauro and P. Agostini, Adv. At. Mol. Opt. Phys. **35**, 79 (1995).
[5] G. G. Paulus, W. Nicklich, Xu Huale, P. Lambropoulos, and H. Walther, Phys. Rev. Lett. **72**, 2851 (1994).
[6] G. G. Paulus, W. Nicklich, and H. Walther, Europhys. Lett. **27**, 267 (1994); V. Schyja, T. Lang, and H. Helm, Phys. Rev. A **57**, 3692 (1997).
[7] G. G. Paulus, F. Zacher, H. Walther, A. Lohr, W. Becker, and M. Kleber, Phys. Rev. Lett. **80**, 484 (1998).
[8] W. Becker, M. Kleber, A. Lohr, G. G. Paulus, H. Walther, and F. Zacher, Laser Phys. **8**, 65 (1998); P. Corkum, Phys. Rev. Lett. **71**, 1994 (1993); A. Becker and F. H. M. Faisal, J. Phys. B **29**, L197 (1996).
[9] Jian Zahng and P. Lambropoulos, Phys. Rev. Lett. **77**, 2186 (1996).
[10] C. J. G. J. Uiterwaal, D. Xenakis, and D. Charalambidis, Z. Phys. D **38**, 309 (1996).
[11] S. G. Preston, A. Sanpera, M. Zepf, W. J. Blyth, C. G. Smith, J. S. Wark, M. H. Key, K. Burnett, K. Nakai, M. Neely, and A. Offenberger, Phys. Rev. A **53**, R31 (1996).
[12] A. Talebpour, C-Y Chien, and S. L. Chin, J. Phys. B **29**, 5725 (1996).
[13] D. Xenakis, N. E. Karapanagioti, D. Charalambidis, H. Bachau, and E. Cormier, Phys. Rev. A **60**, 3916 (1999).
[14] Th. Weber, M. Weckenbroich, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, F. Afaneh, G. Urbasch, M. Vollmer, H. Giessen, and R. Doerner, Phys. Rev. Lett. **84**, 443 (2000); J. Phys. B **33**, L127 (2000); H. Rotke, C. Trump, M. Wittman, G. Korn, K. Hoffmann, and W. Sandner (to be published).
[15] B. Witzel, N. E. Papadogiannis, and D. Charalambidis, Eur. J. Phys. D (to be published).
[16] D. Strickland and G. Mourou, Opt. Commun. **56**, 219 (1985).
[17] V. Stert, W. Radloff, C. P. Schulz, and I. Hertel V, Eur. Phys. J. D **5**, 97 (1999).
[18] C. Guo, M. Li, J. P. Nibarger, and G. N. Gibson, Phys. Rev. A **58**, R4271 (1998).
[19] G. G. Paulus, W. Nicklich, F. Zacher, P. Lambropoulos, and H. Walther, J. Phys. B **29**, L249 (1996).
[20] B. Sheehy and L. F. DiMauro (to be published).