Above-threshold double-ionization spectroscopy of argon

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We have measured the above-threshold ionization (ATI) electron spectrum from nonsequential double ionization of Ar, and compare our results to the single-ionization ATI spectrum. A variation on the coincidence-tagging technique enables the subtraction of contamination associated with single ionization, revealing an excess of higher energy electrons with measured kinetic energies up to approximately $2U_p$. We also find an excess of very low energy photoelectrons, not previously observed. Implications for the rescattering model are discussed.

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Photoelectron spectra from atoms ionized by intense optical-frequency radiation display a rich structure due to the nonperturbative multiphoton process of above-threshold ionization (ATI) [1]. Ion charge-state distributions collected in this ATI regime usually include a small fraction of multiply charged ions along with the majority of singly charged ions. However, the spectrum of electrons coming from these higher-charge states is very difficult to isolate from the stronger single-ionization spectrum.

The doubly charged ion yield as a function of laser pulse energy often displays a distinct knee, or change in slope, below the ATI saturation energy [2], (Fig. 1). This has led to suggestions that there are at least two distinct mechanisms for double ionization: Ordinary ''sequential'' ionization, in which an atom singly ionized in the laser pulse is further ionized; and ''nonsequential,'' or ''direct'' double ionization, where cooperative dynamics leads to simultaneous ejection of two electrons in the laser field. Sequential double ionization is expected to follow the tunneling theory of Ammosov, Delone, and Krainov (ADK) [3], which explains single-ionization rates extremely well. Any excess double ionization beyond the ADK prediction is then customarily called nonsequential double ionization (NSDI).

Recently, momentum spectra of the electrons emitted in double ionization have been obtained with cold-target recoil-ion momentum spectroscopy (COLTRIMS) [4]. High-resolution spectra could help to further illuminate the physics of the multiple-ionization process [5,6]. Here we report high-resolution measurements of above-threshold double-ionization (ATDI) spectra from the double ionization of argon atoms.

Our spectra agree with the COLTRIMS observation that the ATDI spectrum peaks at higher energies than the ATI spectrum. This supports the hypothesis [7] that much of the nonsequential component of ATDI is due to inelastic rescattering of the first photoelectron in the presence of the intense laser field. Our ATDI spectra also display a previously unobserved excess of zero-energy electrons. This low-energy feature, which is not predicted in the most basic rescattering theory, is better characterized by revisions to the model that include multiple rescattering of the first ATI electron [8–11] and inelastic excitation of the ion [8,9].

Argon atoms are ionized at the focus of a 1 kHz titanium-sapphire laser, which delivers 60-fs pulses centered at 800 nm, and the resulting electrons and ions are accelerated in opposite directions toward separate microchannel plates. The laser focus is in a constant electric field of 38.0 V/cm, oriented parallel to the polarization of the laser and along the axis of the time-of-flight tube. The ion detector acts as a mass spectrometer, separating the ion species according to their arrival time, which depends on the charge-to-mass ratio (Fig. 2).

The laser intensity is chosen by observing the Ar^+ and Ar^{2+} ion count rates (Fig. 1). We require that the experiment be conducted below the saturation intensity for Ar^+ to ensure NSDI. The intensity is calibrated by examining the ratio of ion yields Ar^{2+}/Ar^+ at different peak intensities. The onset of the plateau signifying NSDI occurs at the same intensity (within experimenters' uncertainties) in each published argon experiment [12–14]; we use this point to calibrate our intensity with 25% uncertainty. We find that the electron-coincidence experiments were carried out at 2.6×10^{14} W/cm^2, shown in the figure by the dashed vertical line, well within the NSDI regime.

The electrons accelerate 6 mm, then travel 1 m in a field-free flight tube toward the MCP detector. The measured time

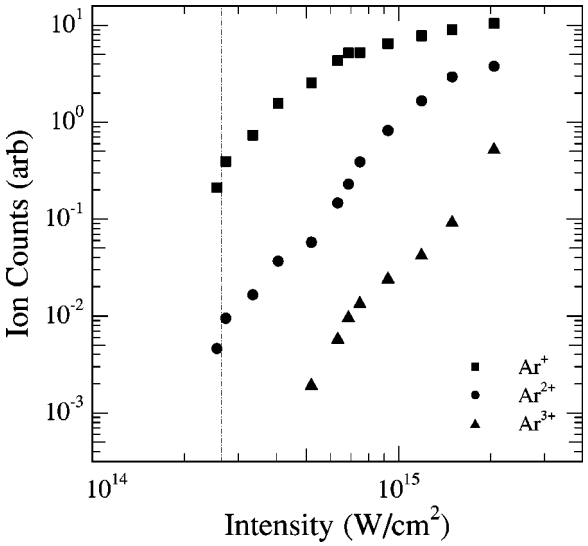

FIG. 1. Multiphoton ionization of Ar using a Ti:sapphire laser (800 nm, 80 fs) shows typical ''knee'' structure in multiply charged ion yields below saturation intensity.

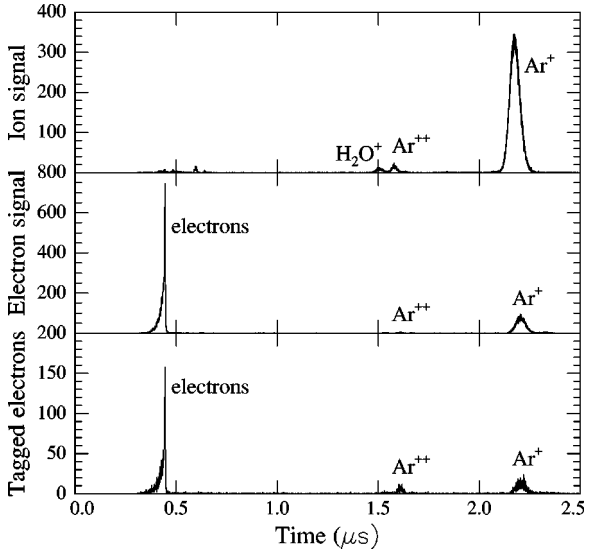

FIG. 2. Ion spectrum (top) shows a dominant Ar^+ peak, a small Ar^{2+} peak, and some background water vapor, with negligible air and $H_2{}^+$. Stray electrons released by the ion MCP arrive at the electron detector with a 20-ns delay with respect to the ion signal, and much later than the photoionization electrons, as seen in the full (middle) and tagged (bottom) electron signals.

of flight is a function of the component of the momentum directed along the flight tube axis. The acceleration field increases the collection efficiency by adding an energy offset, without changing the component of the electron's momentum perpendicular to the field. Electrons with initial momenta in the positive and negative directions with respect to the collection field both arrive at the MCP, but at different times, as indicated by the second axis in electron data figures. This time shift and the slight smearing of arrival times due to electrons' transverse momenta adversely affect the resolution, but the typical ATI structure of peaks spaced by one photon energy is still clearly visible (Fig. 3).

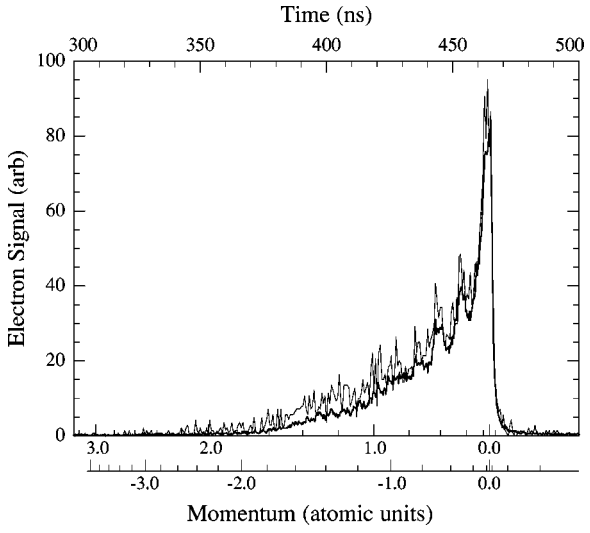

FIG. 3. Coincidence spectrum (thin line) includes only electrons from shots in which a double ion was detected. It is compared to the full spectrum (thick line), scaled as discussed in the text.

The solid angle seen by the MCP varies with energy; the size of the plate limits a detected electron's transverse momentum to about 0.06 a.u.. This means that an electron measured with zero (longitudinal) momentum has a small *total* momentum, but at almost any angle, whereas a small transverse component for high-energy electrons means only a small detection angle. In ATI, an electron's initial energy is assumed to be small, resulting in a Gaussian distribution of transverse momenta, with a width independent of the longitudinal momentum gained from the laser field [15–17]. Thus, the solid angle of the MCP and the electron distribution vary in the same way, meaning that the collection efficiency is approximately energy independent.

Our spectrometer geometry has a peculiar feature: When ions strike the detector, the electrons ejected by the front surface of the MCP are at a potential maximum, and may accelerate either into the plate channels, or back towards the electron detector on the other side of the chamber. These electrons, if detected, arrive at times roughly corresponding to the ion's time of flight, producing a duplicate ion-mass spectrum in the electron time-of-flight signal (Fig. 2). We make use of this to calibrate the detectors and to correct the spectrum for false coincidences, as described below.

To reveal the ATDI electron spectrum, it is necessary to separate the electrons released in a nonsequential double-ionization event from a background of 43 times as many single-ionization events. In the low-density limit of a single atom in the interaction region, an ATDI electron can be identified by the simultaneous presence of a double-ionization event on that laser shot. The correlation may be lost if there is more than one ionization event per laser shot. Therefore, the atomic density must be low and the laser focus small so that there is only one atom in the interaction region.

The ionizing laser is expanded and apertured to a 12.5-mm diameter, then focused with a 10-cm mirror, to produce an Airy focus with active focal volume of about $8000\ \mu m^3$. (The active focal volume is the region bounded by the isophote with half of the peak intensity.) The argon gas pressure of 4×10^{-8} torr yields 1.3×10^{-3} atoms/μm^3, so that the average number of atoms in the focal volume is 10.4, but the number of atoms experiencing light intensity within 10% of the maximum is about two.

Since there is likely to be more than one atom in the focal volume on every laser shot, there is a high probability of false correlations. This is exacerbated by the low ion and electron detection efficiencies, measured to be 4% and 1.5%, respectively [18]. Therefore, if a double ion is detected in a given laser shot, it is possible that there were undetected single ions. If one electron is detected, this electron may be from a single ionization, a false coincidence, rather than from the detected double ionization, a true coincidence.

Electron and ion time-of-flight data are examined on every laser shot. When a double ion is detected in the ion detector, a signal is sent to the electron-detection system to mark the laser shot as containing relevant data. The list of arrival times can be sorted to retrieve those marked events, resulting in a spectrum containing only electrons created in the same laser shot as a double ion. This spectrum will not be

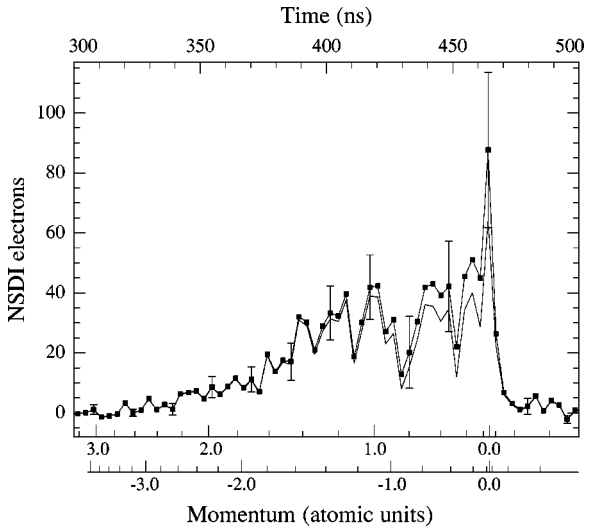

FIG. 4. Double-ionization electron spectrum is found by subtracting the scaled full electron spectrum from the coincidence spectrum in the time domain before converting to momentum. The representative error bars are due to collection statistics, and the two curves represent the scaling uncertainty introduced by the statistics of the number of falsely tagged events.

pure, but will be enriched in comparison with the expectation of less than 3% double ionization in a standard ATI spectrum.

To look at ATDI electrons without the background of false coincidences, we must subtract a scaled version of the full-electron spectrum from the enriched spectrum, with a precisely determined scaling factor [19]. This scaling factor is measured by the duplicate ion spectrum that appears in the electron detector, as described above. This allows us to compare the ion spectrum for the tagged shots and for the untagged shots. The probability of an Ar^+ ion producing electrons that are detected is the same for any shot, whether or not a double ion caused the shot to be tagged. The number of Ar^+ ions seen in this manner is proportional to the number of electrons from single-ionization events. If we subtract the electron spectra so that the Ar^+ peak cancels, we are subtracting the correct proportion of single-ionization electrons from the enriched spectrum to result in a pure ATDI-electron spectrum (Fig. 3).

The resulting electron momentum spectrum has a distinct structure that is different from the full ATI-electron spectrum (Fig. 4). There is a broad peak around 1.2 a.u. corresponding to electron energies in the range of 15–20 eV, and a second pronounced maximum at zero momentum. Statistical uncertainty is shown with error bars; note that the two features are not clearly separate, but could represent a monotonically decreasing function of momentum. However, we treat the data as two peaks to facilitate discussion of two separate ionization processes.

We now discuss the results in the context of the rescattering model of double ionization in ATI. The premise of this model is that the second electron is liberated in an inelastic collision with a returning ATI electron. This means that the first electron must return to the atom core with kinetic energy greater than or equal to the second ionization energy. If the electron receives this kinetic energy from the laser field, this defines a range of field phases that could result in this collisional double ionization [7].

In a simplified approximation, we can assume that the two electrons have zero energy at the moment they are produced; their measured momentum is thus a result of energy transfer from the laser field, which is entirely dependent on the phase of the field at the moment of recollision. The expected energies fall in a narrow range near the maximum possible energy, $2U_p$, twice the ponderomotive energy. A less approximate solution includes the energy of the first electron in excess of the energy needed to ionize, extending the spectrum above this $2U_p$ cutoff. A nonclassical (S-matrix) approach finds a broad peak centered at energies a little lower than this classical value and extending above it [20]. A low drift velocity (phases near a field maximum) results in a trajectory that approaches the core many times; the effect of this can be seen by comparing the recollision energy distributions found in Refs. [7] and [21]. This suggests that lower energy collisions could become dominant, perhaps introducing more ATDI electrons with lower momenta, as measured here.

For the intensity at which our ATDI spectra were taken, 2.6×10^{14} W/cm^2, an energy of $2U_p$ corresponds to 31 eV, or a momentum of 1.5 a.u. There is a peak in the spectrum for momenta around 1.2 a.u., which is generally consistent with the $E\approx2U_p$ or $p\approx2\sqrt{U_p}$ rescattering prediction, given the more realistic expectation of momenta lower than those given in a strictly classical model. This measured momentum is in rough agreement with COLTRIMS data for both electrons [4] and ions [14] in argon. About 55% of electrons are grouped near this momentum, and can reasonably be associated with simple rescattering. The rescattering model is a classical treatment of the field and the atom; any possible structure in the electron spectrum due to resonances with atomic levels is thus excluded from predictions. The measured spectrum shows some structure, but at the current momentum resolution, it is not clear that these point-to-point variations are meaningful.

The narrow band of very low momentum electrons is interesting because it is not expected in the pure rescattering model, nor is it an artifact of the detection scheme. The angular distribution of ATI electrons in the high-energy plateau region associated with elastic rescattering [17] shows side lobes, which might suggest a similar lowered directionality in the NSDI electron distribution resulting from rescattering. However, the expected transverse momentum gained in Coulomb scattering (the ''first'' electron) decreases with increasing energy. This means that for the Coulomb-rescattered electrons, our detector has a tendency to suppress low-energy detection, so that the low-energy enhancement that we observe might even be an underestimate. On the other hand, the transverse momentum for a collisionally excited electron (the ''second'' electron), is expected to be energy independent as in the case of first ionization. In summary, this analysis suggests that the low-energy electrons cannot come from any energy-dependent collection effects.

Although the pure rescattering model predicts only electrons around $2U_p$, the zero-energy electron peak might be in

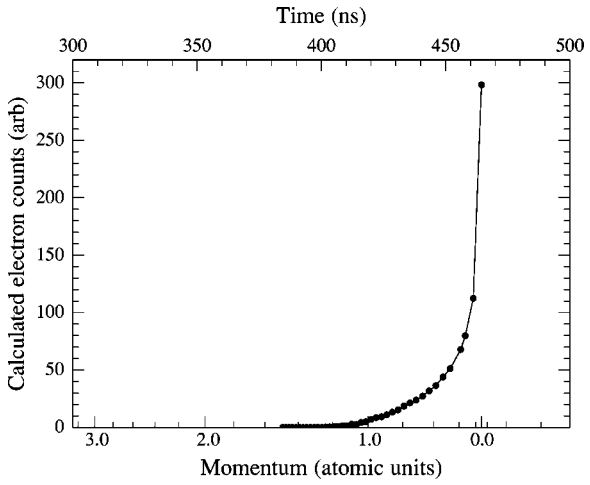

FIG. 5. The ADK ionization rate of excited Ar^+ is combined with the expected drift velocity of the resulting electron to predict an electron-energy spectrum, which was converted to the time domain for easy comparison with data.

agreement with recent modifications to the picture of rescattering. If we assume that electrons produced at field phases described above are the only electrons that result in NSDI, the double-ion yield is underestimated. The pure rescattering model overlooks the propensity of excited atoms or ions to field ionize [8,9], and does not include the Coulomb potential of the ion, which leads to multiple interactions with the core [8–11].

If the electron returns to the atom with slightly less kinetic energy than the ionization energy, then a second electron cannot be released, but can be raised to an excited state. The ADK tunneling ionization rate equations show that excited atoms are very likely to ionize, most probably at the field maxima of subsequent laser cycles. An electron produced at the peak of the cycle is measured as zero energy, as it does not receive any drift velocity from the field phase. We combine the ADK ionization rate over time with the energy gained from the field by electrons produced at these times. As expected, the resulting electron-energy spectrum has a sharp maximum at zero, and is insignificant beyond $1U_p$ (Fig. 5). Forty-five percent of measured ATDI electrons are at these low energies; it is reasonable to associate them with recollision resulting in excitation, rather than ionization.

The simplest recollision theory predicts no zero-energy electrons at these laser intensities [20]. COLTRIMS measurements of the argon ion [14] and electron [4] momentum spectra reveal a significant number of low-momentum products, though not a distribution sharply peaked at zero. To get the calculated ion yields to match experiment [8–11], recollision must result in more excitation than ionization; this would result in at least as many electrons near zero energy as are in the peak at $2U_p$. The data, taken as it stands, approaches these equal numbers. However, if the NSDI electrons are emitted in a less directional manner than the ATI electrons, the detection efficiency is impaired at high energies. If an isotropic distribution is assumed, the low-energy peak is suppressed, but the data is likely overcorrected; the true spectrum would fall between the isotropic correction and the raw data.

When the electron returns toward the ion, it is likely that the thinly spread electron wave packet will miss the ion core. It is possible that the electron will be caught in the ion's Coulomb potential so that over the laser's next few cycles, the electron trajectory will intersect the ion. This orbiting for several cycles in the combined laser and ion potentials is called Coulomb focusing [10,11]. The final electron energy would depend on the energy and time (phase) of collision, which is not easily predicted. This means that these electrons will not necessarily fall near the $2U_p$ expectation, and could have rather low energies, which could affect the 45% excitation to 55% ionization ratio estimated above. Additionally, this ratio would be reduced if high-energy electrons are being undercounted due to an unexpectedly large transverse momentum, perhaps related to the quantum nature of the rescattering problem.

In conclusion, we have measured the electron spectrum from nonsequential double ionization of Ar. We find that in addition to the high-energy peak observed in previous measurements, there is a low-energy feature, which could be attributed to inelastic excitation of the Ar^+ ion, or to multiple returns of the first electron.

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