Observation of Electron Energy Discretization in Strong Field Double Ionization

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We report on the observation of discrete structures in the electron energy distribution for strong field *double* ionization of argon at 394 nm. The experimental conditions were chosen in order to ensure a nonsequential ejection of both electrons with an intermediate rescattering step. We have found discrete above-threshold ionization like peaks in the sum energy of both electrons, as predicted by all quantum mechanical calculations. More surprisingly, however, is the observation of two above-threshold ionization combs in the energy distribution of the individual electrons.

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Single ionization of atoms and molecules by a multicycle short laser pulse leads to a sequence of equidistant discrete peaks in the energy spectrum of the ejected electron. These above-threshold ionization (ATI) peaks result from the discretization of the photon field and show that more photons than necessary to overcome the binding can be absorbed from the field [1]. A similar ATI structure is predicted for nonsequential *double* ionization. For double ionization, it is, however, the sum energy of both electrons which is expected to show the discretization [2–7].

At certain laser intensities, strong field double ionization is orders of magnitude more efficient than expected from a sequential scenario, where the atom is ionized by subsequent independent interactions with the laser field [8]. There is overwhelming experimental and theoretical evidence that the mechanism for this efficient double ejection involves an intermediate step of recollision (see Refs. [9,10] for reviews). This scenario is therefore termed nonsequential double ionization. Initially, only one electron is released. It is accelerated by the laser field, driven back, and shares its energy with another bound electron. The details of what exactly happens upon recollision are under discussion. In addition to evidence for a direct knockout of a second electron in an $(e, 2e)$-type collision [9], there are also contributions to the double ionization yield where the second yet bound electron is excited upon recollision with the first one and field ionized later during the laser pulse (recollision excitation with subsequent ionization) [11]. At lower laser intensities, there is the suggestion that an intermediate doubly excited transition state is formed upon recollision [12,13]. One possible pathway to create such a transition state is subsequent multiple, inelastic field-assisted recollisions, which resonantly excite the target ion [14–16].

Models based on classical physics have proven to be highly successful in reproducing most of the observed features [10]. They do, however, neglect the discrete photon nature of the radiation field. Hence, all energies of the electrons are continuous in these classical calculations. All quantum models of strong field ionization, on the contrary, predict a discretization of energy in the final state continuum (see, e.g., Refs. [2–7]). In a time dependent picture, energy discretization arises from the periodicity of the ionization events in time [6,17]. This concept has been generalized to the two electron case [6] where the sum energy of both electrons shows discrete peaks. The ATI structure on the electron sum energy was predicted already in the first one-dimensional quantum simulations of strong field double ionization [2,7]. It naturally occurs in all quantum models when more than two cycles of the field are taken into account. The energy of these ATI structures in double ionization is predicted to be [2,6]

$$E_1 + E_2 = nh\nu - I_{p1} - I_{p2} - 2U_p. \quad (1)$$

Here, E_1 and E_2 are the continuum energies of the two electrons, and n is the number of absorbed photons with an energy of $h\nu$. I_{p1} and I_{p2} are the Stark shifted ionization potentials of the neutral and the singly charged atom, respectively (for argon, $I_{p1} = 15.76$ eV and $I_{p2} = 27.63$ eV). The ground state Stark shift is usually small. U_p is the ponderomotive potential, which is the ac stark shift of the continuum.

On the experimental side, however, the predicted discretization of the electron sum energy in nonsequential double ionization has evaded observation so far. One possible reason for this is the focal averaging in the experiment. As U_p changes with intensity across the focus, the focal averaging smears out the discrete peaks [see Eq. (1)]. To circumvent this problem, we have chosen 394 nm light for our present study. This doubles the spacing between ATI peaks compared to the fundamental Ti:sapphire output and at the same time reduces U_p. The 394 nm pulses were generated in a 200 μm thick beta barium borate crystal by frequency doubling of the 788 nm output of a Ti:sapphire laser system (100 kHz, 100 μJ, 45 fsec, Wyvern-500, KMLabs). A Cold Target Recoil Ion Momentum Spectrometer reaction microscope [18] was used

to measure both electrons in coincidence with the doubly charged Ar ion. The gas density in our jet was adjusted to reach a count rate of 13 kHz on the electron detector and an ion count rate of 2.5 kHz, 70% of which was argon. We used momentum conservation between the two detected electrons and the ion to suppress events coming from false coincidences. This approach, however, does not eliminate false coincidences completely due to the finite momentum resolution. One kind of false coincidences that remains is when one of the two detected electrons originates from ionization of a second atom in the same pulse. According to our estimations, these events account for less than 20%. We have generated a false correlated electron energy spectrum of these false coincidences and subsequently subtract it from the raw data. The laser intensity in the interaction region has been determined by measuring the shift of the sum energy of electron and proton from dissociative ionization of H_2. This shift, which is given by U_p, has been found to be linear with laser power. We estimate the accuracy of our calibration to be better than $\pm 20\%$.

We first confirm recollision to be responsible for double ionization at the parameters of our experiment. Figure 1(a) shows the intensity dependence of the double ionization probability. It rises only slowly over a range of intensities (1.2–2.2×10^{14} W cm^{-2}, referred to as "the knee" in the literature) because the ejection of the second electron is driven by the electron-electron interaction and not directly by the field. The ratio drops steeply at lower intensities, when the energy of the recolliding electron is insufficient to contribute significantly to ejection of the second electron. These general features of the double ionization probability are very similar to those known for 800 nm. For the experiment, we choose an intensity of 1.3×10^{14} W cm^{-2} [shown with a red arrow in Fig. 1(a)], just at the onset of the nonsequential regime. Under this condition, the Keldysh parameter [19] is 2, indicating a multiphoton regime of ionization. The chosen laser intensity corresponds to $U_p \approx 1.9$ eV; i.e., the maximum energy of the recolliding electron [20] is $3.17 \times U_p \approx 6$ eV. This energy is much below the second ionization potential $I_{p2} = 27.6$ eV and does not even cover the excitation energy of the first excited state ($3s3p^6$), which is 13.5 eV [21]. Thus, for this intensity, the $(e, 2e)$ knockout ionization and recollision excitation with subsequent ionization should be excluded. One of the possible double ionization scenarios in this case is the recapture of the first electron under recollision. The released energy might now be enough for excitation of the second electron, which results in the formation of a doubly excited Coulomb complex or compound state [12,13]. It was shown that under certain conditions, the formation of such a state was the most probable intermediate step in double ionization [13]. Such a Coulomb complex can be ionized subsequently later during the laser pulse [12–16].

A direct proof of the intermediate rescattering step in double ionization is the strong dependence of the double ionization probability on the polarization ellipticity of laser

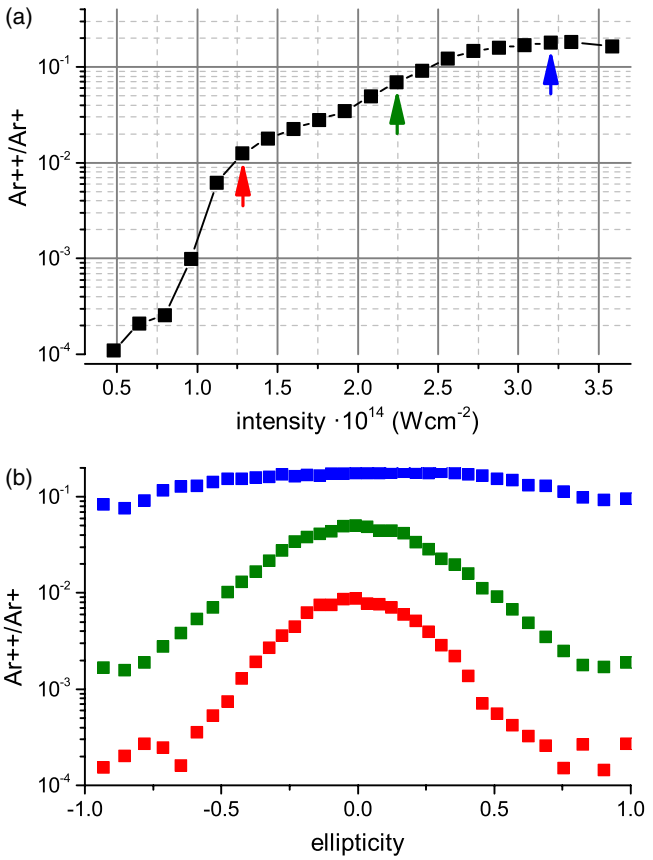

FIG. 1 (color online). Ratio of double to single ionization of Ar by 394 nm, 45 fsec laser pulses (a) as a function of laser intensity and (b) as function of polarization ellipticity at the intensity marked by colored arrows in (a). The intensity shown by the red arrow was used in the experiment (1.3×10^{14} W cm^{-2}). Note the logarithmic scale.

pulses [Fig. 1(b)]. Already, a small ellipticity [Fig. 1(b), red and green curves] steers the trajectories of the electrons away from the parent ion and hence completely suppresses recollision [20,22]. At higher intensities (3×10^{14} W cm^{-2}, Fig. 1, in blue), the double ionization probability is almost independent of the polarization ellipticity, indicating sequential liberation of electrons.

Having established that recollision is the decisive step in double ionization also at 394 nm, we now turn to the predicted ATI structure. Figure 2 shows the electron energy spectrum for single ionization in comparison to our findings for double ionization. The single ionization spectrum is in agreement with previous work [23]. It shows discrete peaks spaced by the photon energy (3.14 eV). In Ar, the first peak shows a fine structure resulting from Freeman resonances [23,24]. Here, the electron is first excited by resonant multiphoton absorption to an excited field dressed Rydberg state. As the pulse rises and falls, the intensity dependent Stark shift moves the Rydberg states at some point in resonance with a multiple of the photon energy. From there, the electron is then ejected by a single photon absorption. The Stark shift of these states is almost identical to the one of the continuum, which

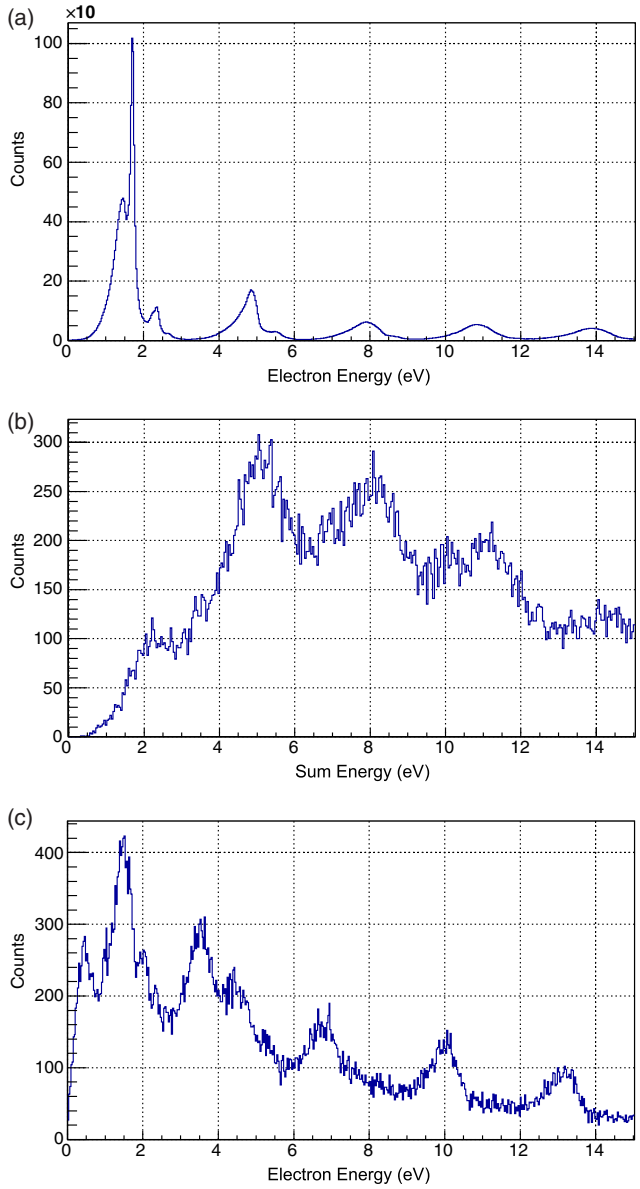

FIG. 2 (color online). (a) Electron energy distribution for single ionization of Ar at 394 nm, 1.3×10^{14} W cm^{-2} (marked with the red arrow in Fig. 1). (b) Sum of the energy of both electrons from double ionization of Ar at the same conditions as (a). (c) Energy of one of the two electrons from double ionization.

leads to intensity independent peak positions in the ATI electron spectrum. Figure 2(b) displays the sum energy of both electrons detected in coincidence with a doubly charged argon ion. The spectrum shows the first experimental observation of a progression of ATI peaks in double ionization. More surprising is, however, the energy distribution of one of the two electrons shown in Fig. 2(c). It shows a pronounced multipeak structure. The discretization of the single electron energy is expected for the case of sequential double ionization. However, under the present experimental conditions, the sequential nature of double ionization can be excluded, as was discussed above (Fig. 1).

To shed more light onto these discrete structures, we plot the energy correlation between the two electrons in Fig. 3(a). The constant sum energy $E_1 + E_2$ is found along diagonal lines in this plot. Any recollision scenario in which the system as a whole absorbs a discrete number of photons [Eq. (1)] whose energy is then continuously shared among the electrons would lead to counts continuously distributed along these lines. In our data, however, the observed electron pairs rather form islands which are clustered along these diagonals of constant sum energy. Thus, in addition to

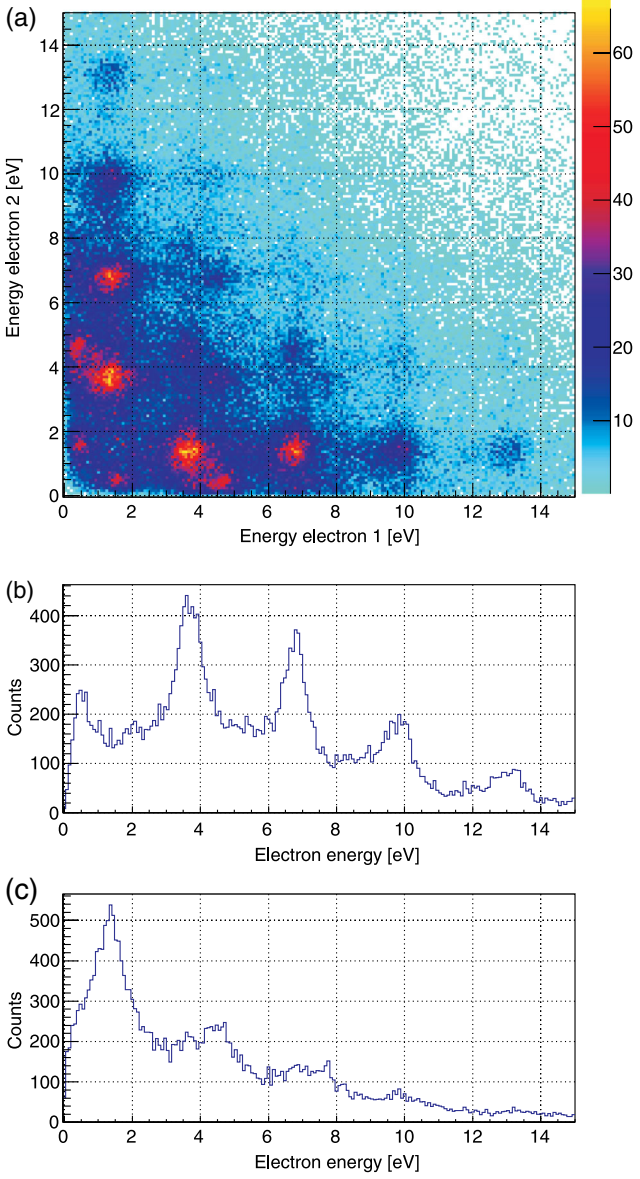

FIG. 3 (color online). Double ionization of Ar by 394 nm, 45 fsec pulses. Same laser conditions as in Fig. 2. (a) Energy of one electron versus the energy of the other electron. The electrons are integrated over all angles. (b) Energy of one of the electrons if the other electron has an energy between 1.0 and 1.6 eV, i.e., projection of (a) in the range 1.0–1.6 eV. (c) Same as (b) for the range 3.0–3.8 eV.

quantization of the sum energy, sharing of the energy between the electrons is not continuous either.

We now discuss the possible mechanism of the observed double ionization. The first step, as was already discussed above, is formation of a Coulomb complex upon recollision of the first electron. Information about its preparation is believed to be lost due to a very strong electron-electron interaction upon recollision [12,13]. The second step is liberation of both electrons from this excited compound state. The discrete islands in Fig. 3(a) rule out all scenarios where the electron pair is set free into the continuum simultaneously, i.e., during the same laser half-cycle. In any such scenario, the electrons would originate spatially close to each other. In this case, the electron-electron repulsion would add a continuous kinetic energy to both electrons. For instance, the potential energy stored in the electron-electron repulsion at a distance of 5 a.u. would already translate to an energy of 2.7 eV on each of the electrons, sufficient to wash out any narrow peaks. It is known from single photon double ionization that the nearly simultaneous birth of the electron pair leads to a smooth energy spectrum [25].

If, however, the two electrons were ejected sequentially from a Coulomb complex, then the two emission steps would each produce their own ATI progression. Each such ATI spectrum would have the same spacing between the peaks but could have different offsets. Close inspection of the pattern in Fig. 3(a) shows, indeed, that it consists of mainly two ATI spectra. We obtain these two ATI spectra by projecting horizontal slices from the data shown in Fig. 3(a). This procedure yields electron spectra of electron 1 for the fixed energy of electron 2. These two ATI spectra show a different offset and a different envelope. All islands in Fig. 3(a) are located at points corresponding to one electron being from the ATI sequence in Fig. 3(b) and the other electron from the shifted one in Fig. 3(c). Therefore, one of the plausible explanations of the present results would be an out-of-phase scenario, as predicted by the classical simulation [14], where the electrons are liberated at different phases of the laser field after multiple recollisions. In this case, the electrons are emitted in opposite directions, as was experimentally observed on argon under similar laser field conditions ($3.17 \times U_p \approx$ 7.4 eV), however, at a wavelength of 800 nm [15].

Presently, light driven double ionization is discussed heavily in three rather different regimes of complexity: the single photon case, which is mostly well understood in atoms [26] and molecules (see, e.g., Ref. [27]), the two photon case as a central topic at free electron laser science (see, e.g., Ref. [28]), and finally the strong field case discussed here. The observed energy discretization shows a direct link between these fields. A discrete energy of the photon field is one of the quantum effects common to all three problems. It is related to a second quantum effect, which is the symmetry of the many particle wave function in the continuum. This is governed by the Pauli principle. To access the role of symmetry, the photon number and hence the total parity of the final state have to be known [26]. For the one and two photon cases, this symmetry is the most important ingredient shaping the two electron continuum (see Refs. [26,29]). The ability to count the photons even for a multiphoton multielectron process paves the road to explore these symmetries in the future.

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