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Two-photon double ionization of helium by attosecond laser pulses: Evidence of highly correlated electron motion

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We apply a recently developed *ab initio* numerical framework to investigate the angular distributions of the emitted electrons in the immediate proximity of the threshold for the two-photon double ionization of helium. Provided one of the electrons is emitted perpendicular to the laser polarization direction, it is found that the angular distribution of the other electron is characterized by three lobes. The results are similar to those recently reported for the corresponding process in the hydrogen negative ion [R. Nepstad and M. Førre, Phys. Rev. A **84**, 021402(R) (2011)], and provide further evidence of highly correlated electron dynamics in the vicinity of the double ionization threshold.

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The problem of direct (nonsequential) two-photon double ionization of helium has been studied extensively in recent years, as exemplified by numerous theoretical [1–18] and experimental [19–24] works. This breakup process is fundamental in the sense that it is one of the simplest processes in nature where electron correlations are exhibited, manifested by a rather complex interplay between the electrons. As such, a complete understanding of it will pave the way for further investigations of the role of correlations in few-photon and multiphoton multiple ionization processes in atoms and molecules.

In the present Brief Report, we investigate the direct two-photon double ionization process of helium in the near vicinity of the lower threshold (i.e., for 40 eV photons), and with particular emphasis on the direction of ejection of the photo-electrons. In a recent work [25], it was found that the corresponding process in H^- is characterized by a strong backward-forward asymmetry in the sense that if one electron is emitted perpendicular to the (linear) laser polarization direction then the other electron is emitted most preferably in the opposite direction, forming three characteristic lobes in the angular distribution. Similar features have also been observed theoretically in H_2 [26–28].

Solving the time-dependent Schrödinger equation numerically for helium [14], the conditional angular distribution is obtained for both short (500 and 1000 as) and long (4 fs) linearly polarized laser pulses. We examine the case where one of the electrons is emitted perpendicular to the laser polarization direction, and integrate over the energy of both electrons. It is found that the direction of emission of the other electron is characterized by three lobes, concordant with the observations in H^- [25]. Furthermore, with increasing pulse duration, the lobe pointing in the backward direction, representing electrons being emitted back-to-back, becomes relatively more important. The "backward" lobe is most distinct at lower photon energies, and already at a photon energy of 42 eV it loses its significance [11]. We therefore anticipate that the presence of the structure at lower photon energies is a signature of a competing double ionization mechanism that becomes suppressed at higher photon energies.

Figure 1 depicts our results for the angular distributions in the double ionization process. In the figure, the left-right arrow indicates the laser polarization direction. The direction of emission of one of the electrons is assumed to be upward (thick arrow), and the lobes in the distributions represent the (conditional) differential probability of emission (angular distribution) of the other electron. In the calculations, the laser pulse is assumed to be of sine-squared shape with a central frequency corresponding to a photon energy of 40 eV. The intensity of the pulse was set to 10^{13} W/cm^2, which is weak enough that three-photon processes are of less importance. Three different (total) pulse durations were considered, 0.5 (upper panel), 1 (intermediate panel), and 4 fs (lower panel). The angular distributions were extracted by projecting onto $Z = 2$ Coulomb waves. The wave packet is propagated some time, 3 (upper panel), 4.5 (intermediate panel), and 1 fs (lower panel), after the pulse before the projections are performed.

The variation of the shape of the angular distributions with increasing pulse duration can be attributed to the very short pulses used in the calculations, the shortest one corresponding to only five optical cycles. This means that the spectral width of the pulse is large and overlaps significantly with the double ionization threshold (positioned at 39.4 eV). As such, we expect the mechanisms responsible for the three-lobe shape of the distributions to become more distinct with increasing pulse durations.

Figure 2 shows the convergence of the conditional angular distribution as a function of the number of angular momenta included in the basis expansion and for the case of the longest pulse. We find a similar convergence behavior for the two shorter pulses used in Fig. 1 (not explicitly shown here). It turns out that the distributions are well converged already at $l_{\max} = 5$, but we have nevertheless used $l_{\max} = 7$ in the calculations in Fig. 1. Furthermore, we found that the value $L_{\max} = 2$ is sufficient to obtain converged results. For further details about the calculations, see Ref. [14].

The angular distributions in Fig. 1 clearly show that there is a strong backward-forward asymmetry. The bending of the left and right (symmetric) lobes has already been well documented

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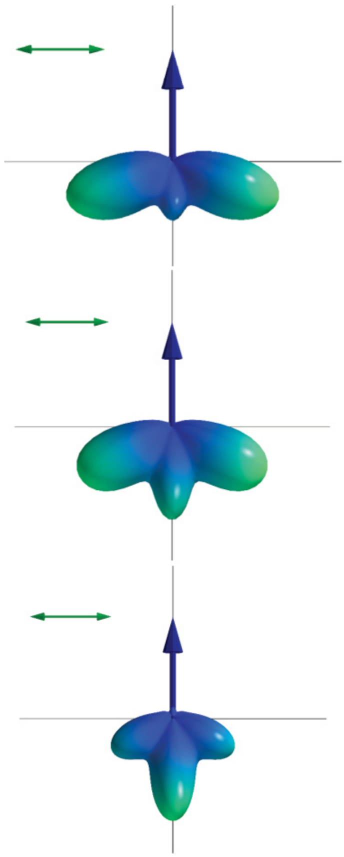

FIG. 1. (Color online) Conditional angular probability distribution for the secondary electron, given that the primary electron is emitted perpendicular to the (linear) laser polarization direction. In the distribution we sum over all possible excess energies of both electrons. The (green) left-right arrow indicates the laser polarization direction, and the (blue) arrow pointing upward defines the direction of emission of the primary electron. The laser field has a central frequency corresponding to 40 eV photons and is taken to be sinusoidal with a sine-square temporal profile and peak intensity 10^{13} W/cm^2. Upper panel: Results for 0.5 fs (total) pulse duration. Total double ionization probability 1.4×10^{-8}. Intermediate panel: Results for 1.0 fs (total) pulse duration. Total double ionization probability 1.4×10^{-8}. Lower panel: Results for 4.0 fs (total) pulse duration. Total double ionization probability 3.5×10^{-8}.

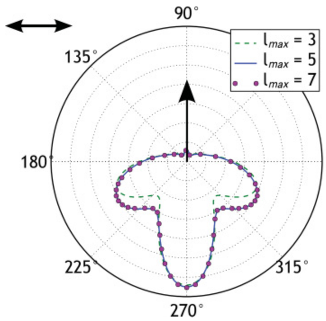

FIG. 2. (Color online) Convergence of the angular probability distribution shown in the lower panel in Fig. 1 versus l_{max}. The figure shows results for $l_{\text{max}} = 3$, 5, and 7. $L_{\text{max}} = 2$ in all cases.

in previous studies in He [5,11,13,29–31], H^- [25], and H_2 [26–28,32], and we will only comment briefly on this feature here. Assuming for the moment that the electrons absorb one photon each in the two-photon double ionization event, this would, to a zeroth-order approximation, give rise to a p-lobe structure (oriented along the laser polarization direction) in their respective angular distributions. If now for some reason one of the electrons happens to be emitted perpendicular to the laser polarization direction, it is rather clear that the p lobe of the other electron would be bent down in the opposite direction, simply because of the Coulomb repulsion between them. Within this simple classical picture, the bending should also become less and less pronounced with increasing photon energy (i.e., for higher and higher excess energies of the electrons), a feature that is consistent with *ab initio* findings.

In a recent work [15], it was suggested that the most likely two-photon two-electron ejection route is comprised of the absorption of one photon by each electron, giving rise to the left and right lobes in the angular distributions. Thus, a possible explanation for the third lobe in the distributions could be a competing mechanism, in which the primary electron absorbs two photons and then knocks out the secondary electron in an $(e,2e)$-like process. It is known that a similar knockout mechanism plays a major role in the closely related one-photon double ionization scenario [33–36]. As such, this process would represent the direct two-photon counterpart to the one-photon double ionization. While it is difficult to test this hypothesis directly, it is reasonable to assume that the ionization resulting from this mechanism will be roughly proportional to the probability for two-photon single ionization (TPSI). This value can be extracted from the wave function. To assure that one of the electrons absorbs both photons, we select the portion of TPSI in which the bound electron is left in the ground state of the He^+ ion. Figure 3 shows this TPSI cross section and the two-photon double ionization (TPDI) cross section as a function of the photon energy. The relative importance of TPSI at low energies supports a scenario where one electron absorbs two photons (corresponding to TPSI) and subsequently knocks out the second electron in a half-collision-like process [33,34], resulting in TPDI. Such a process could then be an important channel of ionization at the lowest photon energies and be responsible for the third lobe in the angular distributions in Fig. 1.

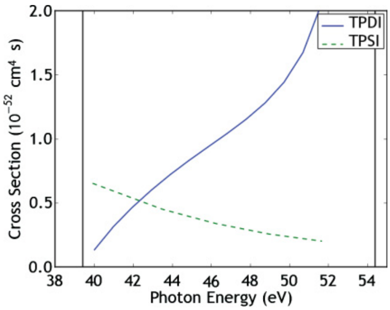

FIG. 3. (Color online) Generalized cross section for direct (nonsequential) two-photon single (TPSI) and double (TPDI) ionization of helium. In the TPSI process, the residual He^+ ion is assumed to be left in the ground state (i.e., the figure displays the partial cross section for this particular ionization scenario). The TPDI cross sections are extracted from Ref. [14]. The vertical lines define the two-photon direct double ionization region.

Recently, a similar three-lobe shape of the angular distribution in H^- was reported [25]. It turns out that the third lobe is even more distinct in H^- in that it survives over a larger interval of photon energies. For helium it is suppressed already at 42 eV (i.e., only 2–3 eV above threshold [11]). We anticipate that this difference can be attributed to the assumption that knockout mechanisms are relatively more important in highly correlated systems.

In concluding this report, we would like to note the close resemblance between the angular distributions obtained in the present work and the corresponding distributions reported in a completely different strong-field nondipole (single) ionization scenario in atomic hydrogen [37,38]. Despite stemming from completely different ionization processes, these studies have one thing in common in that the three-lobe structure was attributed to two underlying (classical) ionization mechanisms. In the present work, these two competing double ionization scenarios are (in a semiclassical picture), respectively, one single-photon absorption by each electron [15] and absorption of two photons by the primary electron followed by a knockout of the second electron in an $(e,2e)$-like process. We presume that the second process is the dominating one at lower photon energies, whereas the first takes over at higher photon energies. This assumption is also in agreement with the findings in a recent work [15].

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