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Nonsequential double ionization of the hydride ion by two-photon absorption

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We apply a recently developed *ab initio* numerical framework to calculate (generalized) total cross sections for the process of nonsequential (direct) two-photon double ionization of the hydride ion (H^-), at photon energies ranging from 7.75 to 10.5 eV. The total cross section is about an order of magnitude larger than the corresponding one obtained for helium, the reason being that the electronic correlation is relatively more important in H^-. Furthermore, we examine single- and triple-differential cross sections at the photon energies 7.75 and 9 eV and find that for the lower photon energy the electron energy distribution attains a maximum when both electrons are emitted with equal energies.

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The invention of high-order harmonic [1,2] and free-electron laser (FEL) [3,4] light sources has inaugurated a new era in atomic physics. With these developments, the role of electron correlations in few-photon multiple ionization processes in atoms [5–9] and molecules [10] can be experimentally investigated. A parallel development of *ab initio* numerical methods, capable of addressing highly correlated electron motion in few-photon [11,12] and multiphoton [13] ionization processes at the most detailed level, has taken place. Double ionization processes have been scrutinized for correlation effects, in particular, those involving only single-photon absorption. More recently, two-electron numerical studies of one-photon double ionization processes have been extended to more complex systems, such as lithium [14,15] and beryllium [15,16].

As a rather complete understanding of the process of one-photon double ionization of helium is emerging [17,18] the related process of nonsequential two-photon double ionization (TPDI) is currently receiving considerable theoretical [12,19–35] and experimental [36–41] attention. Recently, TPDI of H_2 by 30-eV photons was studied theoretically [42–44], and clear discrepancies in the calculated triple-differential cross sections were identified.

Despite the general interest in correlated TPDI processes, focus has been directed toward helium, and less attention has been given to TPDI of the negative hydride ion H^-, a two-electron system held together by electronic correlation. The single bound state of this negative ion, loosely bound at 0.755 13 eV [45], has a correlation energy almost twice its ionization energy, a significantly greater percentage than found for the ground state of helium.

The importance of H^- for stellar opacity was recognized in the early days of quantum mechanics [46] and made it an interesting system for study. Among the issues considered was whether any excited states existed, which if true would have implications for absorption in stellar atmospheres. After some time it became clear such was not the case (see, e.g., Ref. [47]). In more recent times double ionization by single-photon absorption has become a topic of interest [48], and this has continued up to recent times [23,49–53]. In the most recent of these investigations [53], similarities and differences between He and H^- were examined, and although broadly similar trends were observed, some significant differences were found for certain photon energies and electron energy sharings.

In this Rapid Communication, we study the process of two-photon direct (nonsequential) double ionization of the hydride ion (H^-) in the photon energy interval from 7.75 to 10.5 eV, i.e., in a regime where sequential ionization is energetically forbidden. The threshold for nonsequential TPDI of H^- is located at 7.2 eV, whereas the corresponding sequential ionization threshold is at 10.96 eV. In particular, we calculate total, single- and triple-differential cross sections for the direct process and compare, when possible, our results with previously obtained results [54–56].

Our numerical method has been described previously [31,57], but we summarize the key points here for the convenience of the reader. The two-electron wave function, expressed in spherical coordinates, is expanded in a product of B splines and coupled spherical harmonics, and the time evolution of the expansion coefficients is obtained with the implicit Cayley form propagator,

$$\left(\mathbf{S}+\frac{\imath\Delta t}{2}\mathbf{H}\right)\mathbf{c}(t+\Delta t)=\left(\mathbf{S}-\frac{\imath\Delta t}{2}\mathbf{H}\right)\mathbf{c}(t), \qquad (1)$$

where the coefficients $\mathbf{c}$ are defined by

$$\psi(\mathbf{r}_1,\mathbf{r}_2,t)=\sum_{i,j,k}c_{ijk}(t)\frac{B_i(r_1)}{r_1}\frac{B_j(r_2)}{r_2}\mathcal{Y}^{LM}_{l_1,l_2}(\Omega_1,\Omega_2), \qquad (2)$$

$\mathbf{S}$ is the total overlap matrix of the basis, and $\mathbf{H}$ is the discretized, time-dependent Hamiltonian of the H^- system. The sum over all angular quantum numbers $k=\{L,M,l_1,l_2\}$ is in the present case restricted by $M=0$, since only linearly polarized laser interaction along the z axis is considered. Equation (1) contains an implicit part, which is solved by a suitable iterative method. Our calculations have been performed with the Pyprop toolkit [58], using the helium extension package [59], designed to handle propagation and analysis of two-electron systems.

We are presently interested in the doubly ionized component of the wave packet, which we extract by projection onto $Z=1$ Coulomb waves, after propagating the wave packet for some additional optical cycles after the action of the pulse. The

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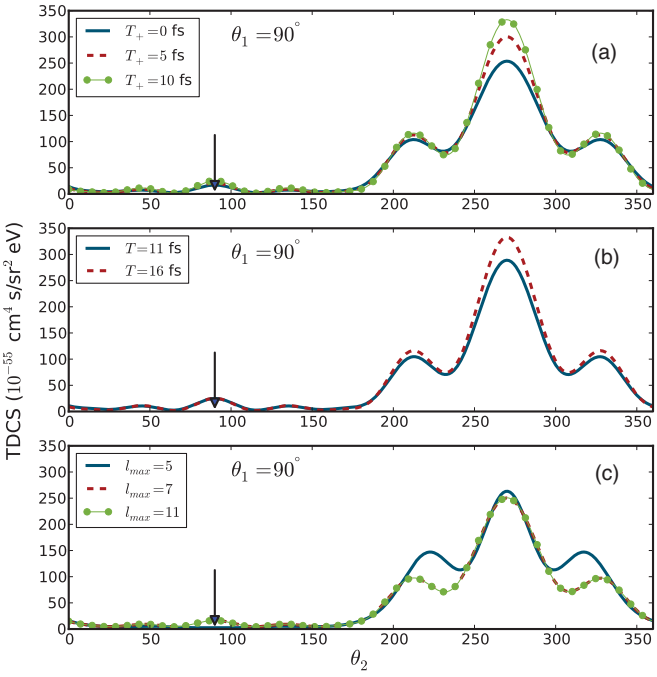

FIG. 1. (Color online) Convergence of the TDCS with respect to (a) additional propagation time T_+ added after the pulse, for $l_{\max} = 7$ and 16 fs pulse duration; (b) pulse duration T, for $l_{\max} = 7$ and $T_+ = 10$ fs; and (c) size of the angular momentum expansion, $(L,l_1,l_2) = (2,5,5)$, $(2,7,7)$, and $(3,11,11)$, for $T = 11$ fs and $T_+ = 5$ fs. In all cases, the TDCS is obtained at $\hbar\omega = 7.75$ eV, with a $\sin^2$ pulse, $E_1 = 0.6$ eV, and with one of the electrons emitted at $\theta_1 = 90^\circ$ (indicated by the arrows in the figures) with respect to the polarization axis.

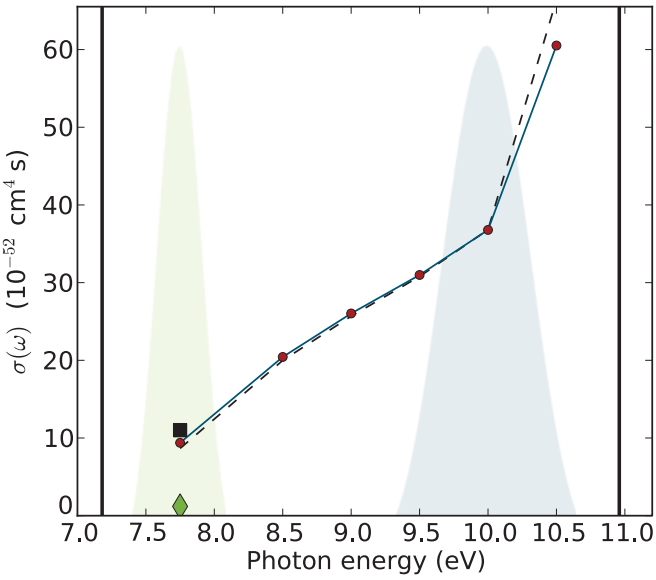

FIG. 2. (Color online) Total cross section for the nonsequential two-photon double ionization of H^-, extracted 5 fs after the pulse. Solid blue line with red dots: Present result. Green diamond: Result of Mercouris *et al.* [56]. Black square: Result of Kornberg [55]. Dashed black line: Cross section extracted right after the pulse. The vertical lines define the two-photon direct double ionization region. Shaded areas indicate the spectral widths of the 20-cycle (right) and 30-cycle (left) pulses.

latter is a well-known procedure intended to improve the quality of the Coulomb wave approximation [60,61] and has been used, for instance, in obtaining the two-photon double ionization of helium [28] and H_2 [44]. Useful quantities, including cross sections resolved in both energy and angles, may be calculated from the projected wave packet (see, e.g., Ref. [28]).

In our calculations we have used a radial box extending to either 300 or 400 a.u., covered by 221 or 293 B splines, respectively. Close to the origin the density of B splines was increased to better resolve the ground state. Electrons ejected through the direct process have very low excess energy (<8 eV), and thus only a small number of splines are required. We have also checked that our results remained consistent when the density of B splines was increased. For the ground state (with $l_{\max} = 9$) we find an energy of $-0.527\,726$ a.u., in good agreement with the nonrelativistic value $-0.527\,751$ a.u. calculated by Pekeris [45].

The total cross section is fairly well converged using a smaller box ($r_{\max} = 200$ a.u.) and 10-cycle pulses, except for close to the thresholds, but we employ 20- or 30-cycle pulses for the results presented here to improve the energy resolution and to check for convergence. The intensity of our pulse was set to 10^{11} W/cm^2, which is weak enough that three-photon processes are negligible, but it still produces a good two-photon signal. Increasing the intensity by a factor of 10 resulted in a change in total cross section of about 0.1%.

Convergence of the triple-differential cross section (TDCS) at $\theta_1 = 90^\circ$ with respect to temporal propagation of the wave packet after the pulse as well as pulse duration is shown in Figs. 1(a) and 1(b). Although apparently the TDCS at 7.75-eV photon energy is not fully converged, even with the longest pulse (16 fs) considered and with 10 fs additional propagation time added after the pulse, and in particular at the angle $\theta_2 = 270^\circ$, the overall shape of the TDCS object is obtained. Applying both a sine-squared and a Gaussian pulse, we have also checked that the results are insensitive to the actual pulse shape. Furthermore, calculations in the length and velocity gauges yielded the same result.

Since we are using a multipole expansion to numerically represent the electron-electron interaction, and since H^- is strongly correlated, careful checks of convergence in $l_{\max}$ must be made. In Fig. 1(c) we show the TDCS for different values of (L,l_1,l_2). Although (2,5,5) correctly yields the overall shapes, $l_{\max} = 7$ is required for quantitatively convergent results on the scales in the figure, similar to what is the case for helium [28] and one-photon double ionization of H^- [53]. The total and single-differential cross sections are less sensitive, $l_{\max} = 5$ being sufficient for convergence in those cases.

Figure 2 shows the calculated total cross section for the process of nonsequential two-photon double ionization of H^- in the photon energy interval 7.75 to 10.5 eV. Also shown are the results of Kornberg [55] and Mercouris *et al.* [56] for the photon energy 7.75 eV. Kornberg obtained the value 11×10^{-52} cm^4 s for the cross section, whereas the value of Mercouris *et al.* is an order of magnitude lower, i.e., 1.2×10^{-52} cm^4 s. In comparison, our value is 9.8×10^{-52} cm^4 s, which in turn is about 10% lower than the value of Kornberg.

The general shape of the cross section is qualitatively similar to the corresponding one in helium (see, e.g.,

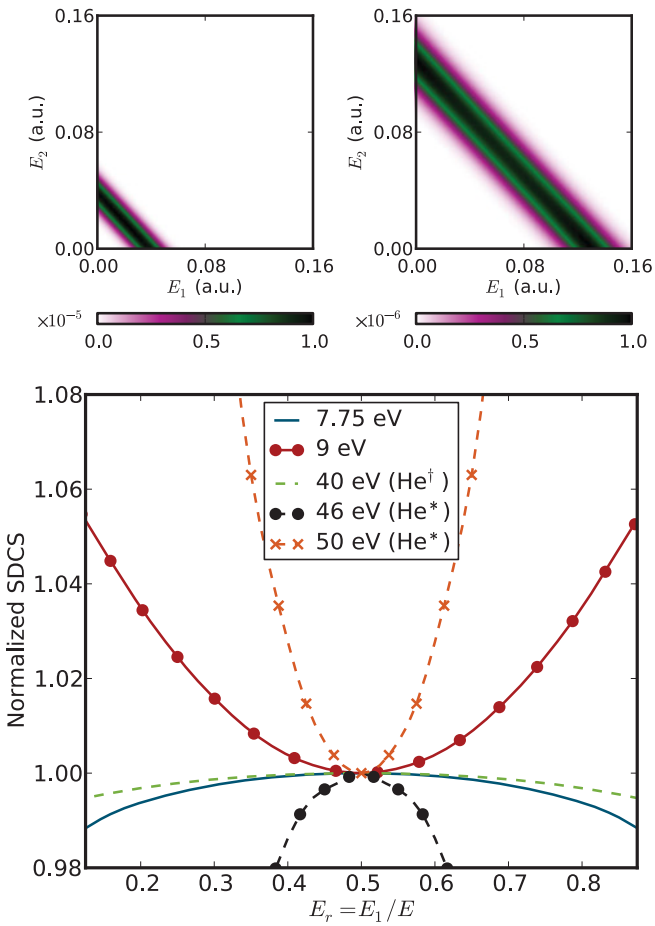

FIG. 3. (Color online) Upper panels: Energy distribution (given in a.u.) after two-photon double ionization for the two photon energies 7.75 (left panel) and 9 eV (right panel), respectively. The pulse duration was 30 (20) cycles (sine-squared envelope) for 7.75 (9) eV, and the wave function was propagated 10 (5) fs after the pulse. Lower panel: Corresponding SDCS for the photon energies 7.75 (lower solid [blue] curve) and 9 eV (upper solid [red] curve with dots). †Pyprop result for helium for 40-eV photons (dashed [green] line). $^*J$ matrix results (with correlation) from Foumouo *et al.* [30]. The total cross sections for the three cases are 9.8, 26.0, and 0.1 $\times 10^{-52}$ cm^4 s for 7.75, 9, and 40 eV (He), respectively.

Refs. [28,31,33]), but about an order of magnitude larger. We also observe a fairly sharp increase of the cross section near the upper threshold, also similar to the helium case [24,26,28,33], but longer pulses would be required to properly resolve the cross section in this region (the shaded areas in Fig. 2 indicate the spectra of the pulses used in the present work).

Figure 3 (lower panel) shows the single-differential cross section (SDCS) at the photon energies 7.75 and 9 eV. Interestingly, at the lower photon energy, the energy distribution exhibits a maximum (negative concavity) when both electrons are emitted with similar energies, while at the higher photon energy the distribution is U shaped. The SDCSs are obtained from the respective energy distributions of the two electrons, shown in the upper panels of Fig. 3. Recently, it was predicted that the anti-U shape could also appear in neon at lower photon energies [32], and it has furthermore been suggested that the energy spectrum of helium has a negative concavity near the

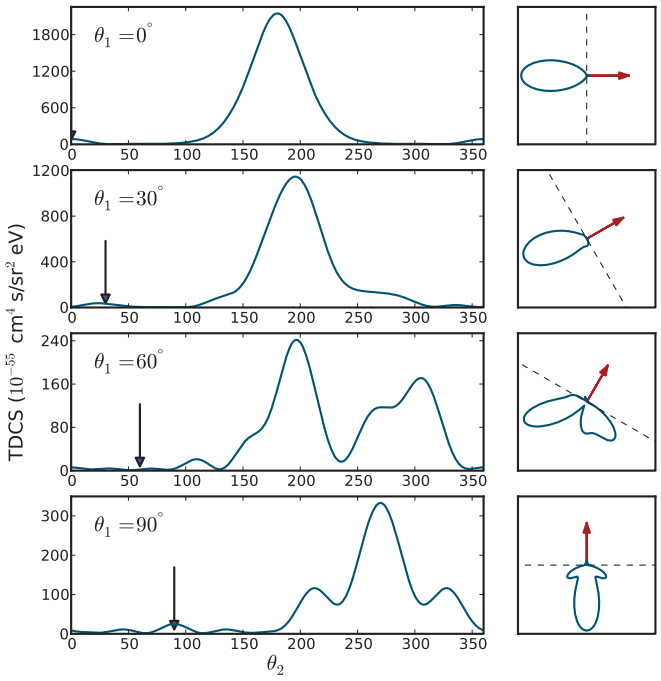

FIG. 4. (Color online) TDCS at $\hbar\omega = 7.75$ eV, obtained with $E_1 = 0.6$ eV, a 16-fs sin^2 pulse (30 cycles), and $l_{\max} = 7$. The TDCS was obtained with one of the electrons emitted at an angle θ_1 (indicated by the arrows in the figures) with respect to the polarization axis and was extracted some 10 fs after the end of the pulse.

lower threshold [30]. The results of Foumouo *et al.* [30] at 46- and 50-eV photon energies, obtained by including final state correlations, are shown in Fig. 3 for comparison. The dashed

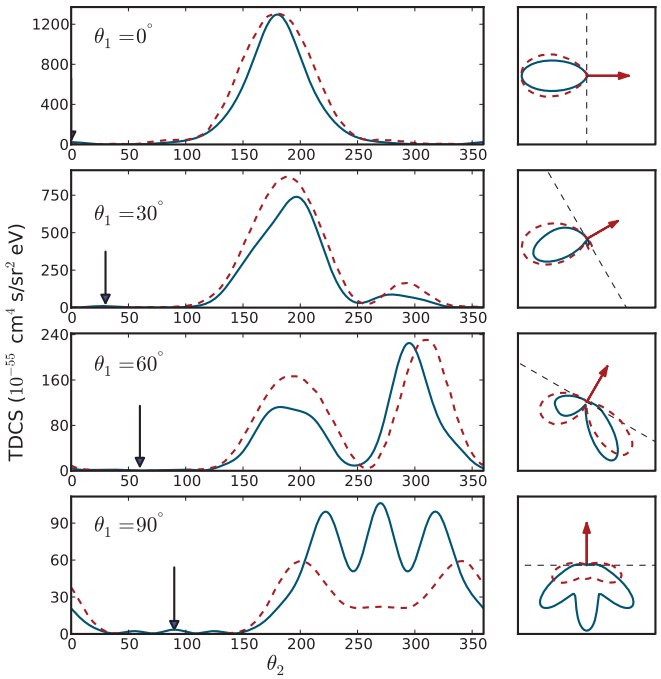

FIG. 5. (Color online) Same as Fig. 4, but for $\hbar\omega = 9$ eV, $E_1 = 1.9$ eV, and a 10-fs pulse (20 cycles). Dashed (red) line: Results for He from Ref. [28] at 42-eV photon energy, 4-fs pulse, and $E_1 = 2.5$ eV, scaled by a factor of 100 in all panels.

line in the figure shows the corresponding SDCS obtained for helium at the photon energy 40 eV, close to the lower threshold in helium (39.4 eV), and indeed a slightly negative concavity is observed. The SDCS result for helium (40 eV) was derived from a pulse 7 fs in duration and extracted by projecting onto $Z = 2$ Coulomb waves. For details about the calculation, see Ref. [31].

Figures 4 and 5 show the (conditional) TDCS for the photon energies 7.75 and 9 eV (see Eq. (15) of [28]). The corresponding TDCS of helium obtained at 42 eV [28,62,63] is shown in Fig. 5. The TDCSs of H^- differ somewhat from those of He, but nevertheless they share many common features. It is worth noting that the excess energy in the helium case is somewhat higher than for the H^- one. The figures clearly show that there is a strong backward-forward asymmetry in the distributions at all angles. This clear back-to-back emission pattern has already been pointed out in helium [22,28,30,64–66], but it turns out that the effect is even more pronounced in H^-. This is particularly the case at 90°, where the emission pattern bends away from the dashed center line to a greater extent than what is found for helium. We anticipate that the reason for this is that the electronic correlation is relatively more important in H^- than in He, which is also the case for a similar effect in the TDCS of one-photon double ionization of H^- [53].

Comparing the lowermost panels in Figs. 4 and 5, respectively, corresponding to $\theta_1 = 90°$, striking differences in the TDCSs are seen. For the lower photon energy (7.75 eV) the second electron is most likely emitted in the backward direction, while for the higher photon energy (9 eV) it has similar probabilities of being ejected at $\theta_2 \simeq 220°$, 270°, and 320°. As such, the angular lobe pointing in the backward direction becomes relatively less important with increasing photon energy, being most pronounced in the immediate vicinity of the threshold for nonsequential TPDI. The strong sensitivity of the backward lobe to the photon energy provides some evidence for highly correlated electron dynamics in the proximity of the threshold [30].

In conclusion, we have obtained single- and triple-differential cross sections as well as total cross sections for the process of nonsequential double ionization of the hydride ion (H^-) by two-photon impact. Due partly to the strong correlation in this system, the total cross section is about an order of magnitude larger than the helium counterpart. Furthermore, a strong backward-forward asymmetry in the angular distributions of the ejected electrons is observed at all angles. Finally, we find that the electrons are preferably emitted with similar energies when the photon energy is low.

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