

Two-photon double ionization of the H_2 molecule: Cross sections and amplitude analysis

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We perform time-dependent calculations of triply differential cross sections (TDCS) of two-photon double-electron ionization of the aligned H_2 molecule. Our TDCS results for equal energy sharing between photoelectrons agree quite well with a recent time-dependent calculation by Guan *et al.* [X. Guan, K. Bartschat, and B. I. Schneider, *Phys. Rev. A* **77**, 043421 (2008)] who employed a slightly different numerical technique. We supplement these studies by calculating TDCS at an unequal energy sharing and by generating symmetrized ionization amplitudes.

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Two-photon double-electron ionization (TPDI) of the H_2 molecule is a benchmark process that can be used to test the power and efficiency of modern computational techniques employed in atomic collision physics. On the one hand, this process is simple enough to permit a completely *ab initio* treatment. On the other hand, an extremely important role of the few-body correlated dynamics, intrinsic to the TPDI process, makes the problem quite a challenging one. A considerably simpler problem of TPDI of helium had proved itself sufficiently hard to solve with results of various groups painting a somewhat controversial picture [1–3]. For the process of TPDI in the H_2 molecule, an additional degree of complexity is introduced by the two-center nature of the target. It is therefore not surprising that TPDI results on H_2 produced by different computational techniques disagree. Indeed, calculations by Colgan *et al.* [4,5], who employed the time-dependent close-coupling (TDCC) method, and results reported by Morales *et al.* [6], who used an exterior complex scaling (ECS) method, differ considerably. A more recent calculation by Guan *et al.* [7] using yet another numerical technique of solving the time-dependent Schrödinger equation (TDSE) in prolate spheroidal (PS) coordinates gave results sandwiched in between the two sets of data reported by Colgan *et al.* [4] and Morales *et al.* [6].

Guan *et al.* [7] attributed this discrepancy to the fact that the calculations reported in Refs. [4,6,7] employed different laser pulse parameters. Nominally, all three calculations used the same central photon frequency, $\omega = 30$ eV. However, the TDCC calculation [4] was performed for a pulse intensity of 10^{15} W/cm², while the approach used in Ref. [6] was perturbative in nature and a time-independent one. This may indeed make direct comparison of these two calculations not very sensible, since the high intensity of 10^{15} W/cm² used in Ref. [4] may well lie in the nonperturbative regime. The calculation in Ref. [7] was a time-dependent one, but used a weaker laser pulse with an intensity of 10^{14} W/cm². The disagreement of the results of this calculation with those of Ref. [4] may therefore well be, as surmised by Guan *et al.* [7], due to very different field intensities. The difference of the results in Refs. [4,6], on the other hand, could well be attributed to the different pulse shape and duration used in these

calculations. Indeed, the considerable sensitivity of the results to the pulse shape employed in a time-dependent calculation was noted [5]. We have, therefore, two possible parameters—the pulse intensity and the pulse duration—which may explain the observed (and sometimes considerable) differences in the data reported in Refs. [4,6,7].

This multitude of reasons behind the differences in the TPDI results for H_2 prompted us to do an additional set of calculations with the laser parameters overlapping those used in previous works. If we can firmly establish the validity and accuracy of the results for at least some choice of the pulse intensity and pulse duration that will be an important step toward understanding TPDI in the H_2 molecule. One should also take into account the fact that, by their very nature, calculations of photoionization processes in the H_2 molecule are very computationally intensive. Any new piece of information helping to clarify the overall picture comes, therefore, at a considerable cost.

We present below the calculation of TPDI in H_2 using the procedure that we applied before to study the process of single-photon double ionization of H_2 [8]. Since this procedure is time dependent, we cannot consider the limit of the infinitely long laser pulse to which the results obtained in Ref. [6] pertain. We consider below a laser pulse with a duration of eight optical cycles, the carrier frequency $\omega = 30$ eV, and a moderately low field strength of 0.1 a.u. (corresponding to the pulse intensity of 3.5×10^{14} W/cm²). These parameters are not very different from those used in Ref. [7] to which our results should be directly comparable. We consider the evolution of the H_2 molecule in the presence of a linearly polarized electromagnetic (EM) pulse:

$$\mathbf{E}(t) = E_0 f(t) \cos \omega t. \quad (1)$$

The pulse is switched off outside the time interval $(0, T_1)$, where $T_1 = 8T$ and $T = 2\pi/\omega$ is an optical cycle corresponding to the carrier frequency $\omega = 30$ eV. The envelope function $f(t)$ is chosen so that the amplitude of the field is ramped on and off smoothly during one optical cycle T and is constant in between. Note, that this pulse shape is different from the sine-squared pulse used in Ref. [7]. The molecule is assumed to be oriented along the quantization axis $\hat{\mathbf{R}} \parallel \mathbf{z}$. For the cases of the mutual parallel and perpendicular orientations of the field polarization and the molecular axis, which we consider below, the field is polarized along the z axis or the x axis, respectively.

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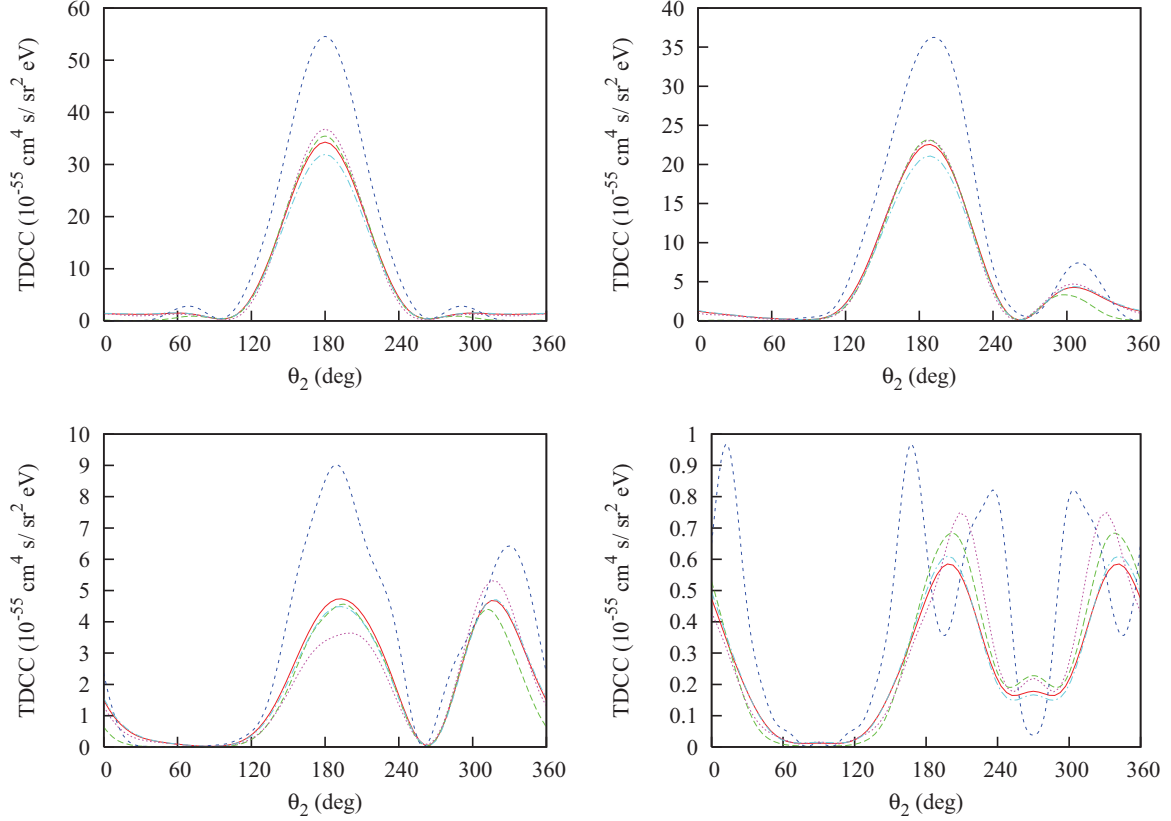


FIG. 1. (Color online) Coplanar TDCS of double ionization of H_2 at a photon energy of 30 eV and equal energy sharing for the perpendicular field-molecule orientation for $\theta_1 = 0^\circ$ (upper left), $\theta_1 = 30^\circ$ (upper right), $\theta_1 = 60^\circ$ (bottom left), and $\theta_1 = 90^\circ$ (bottom right). Shown are the present TDSE results [(red) solid line, length gauge; (cyan) dot-dashed line, velocity gauge]; the TDSE-PS results [7], (green) long dash; the ECS data [6], (blue) short dash; and the TDCC data [4,5], (magenta) dots.

The TDSE, describing the evolution of the H_2 molecule interacting with the field, is solved following closely the computational procedure that we used in Ref. [8] for the process of single-photon double ionization. For the reader's convenience we describe below essential steps of the calculation. We write the TDSE as

$$i\partial\Psi/\partial t = [\hat{H}_{\text{mol}} + \hat{H}_{\text{int}}(t)]\Psi, \quad (2)$$

where $\hat{H}_{\text{mol}}$ is the Hamiltonian of a field-free H_2 molecule and the operator $\hat{H}_{\text{int}}(t)$ describes the molecule interaction. We present below results obtained for both the length and velocity forms of this operator. The Hamiltonian is discretized on a spatial grid $\{r_i\}$ composed of several intervals with a progressively increasing step size. At each grid point the wave function is represented as a partial wave expansion:

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \sum_{l_1, l_2, J} f_{l_1 l_2}^J(r_1, r_2) |l_1(1)l_2(2)J\rangle. \quad (3)$$

Here the notation $|l_1(1)l_2(2)J\rangle$ is used for bipolar harmonics [9], and summation is restricted to l_1 and $l_2 = 0-6$ with the total angular momenta $J = 0-6$.

The initial $^1\Sigma_g$ state was prepared by using imaginary time propagation of the field-free H_2 molecule starting from some trial wave function. This procedure gives the value of -1.167 a.u. for the ground-state energy at the equilibrium internuclear distance $R = 1.401$ a.u. (with the Coulomb

interaction of the nuclei included into the Hamiltonian). With this initial state, the solution of the TDSE is propagated in time over the interval $(0, 8T)$ using the Arnoldi-Lanczos method [10,11].

To find TPDI cross sections we project the solution of the TDSE after the end of the pulse on the set of doubly ionized states constructed from the products of the Coulomb waves describing the electron motion in the field of the central charge with $Z = 2$. To account for possible nonorthogonality of the doubly ionized states and the bound and singly ionized states of the H_2 molecule, we use the recipe proposed in Ref. [12]. According to this recipe, all the radial integrals, which we need to calculate for projecting the solution of the TDSE on the doubly ionized states, should be computed starting from some value $r = R_0$ or, equivalently, the solution of the TDSE after the end of the pulse should be put to zero if either $r_1 < R_0$ or $r_2 < R_0$. Physical justification of this procedure lies in the observation that, if we wait long enough, the wave packet describing doubly ionized states leaves the region where either $r_1 < R_0$ or $r_2 < R_0$. Outside this region, the part of the wave function describing contributions of bound and singly ionized states is heavily dumped due to the bound character of at least one of the electrons. One can expect, therefore, that if we choose R_0 appropriately, then we can avoid contamination of the double ionization process by singly ionized and bound state channels. We illustrated this procedure

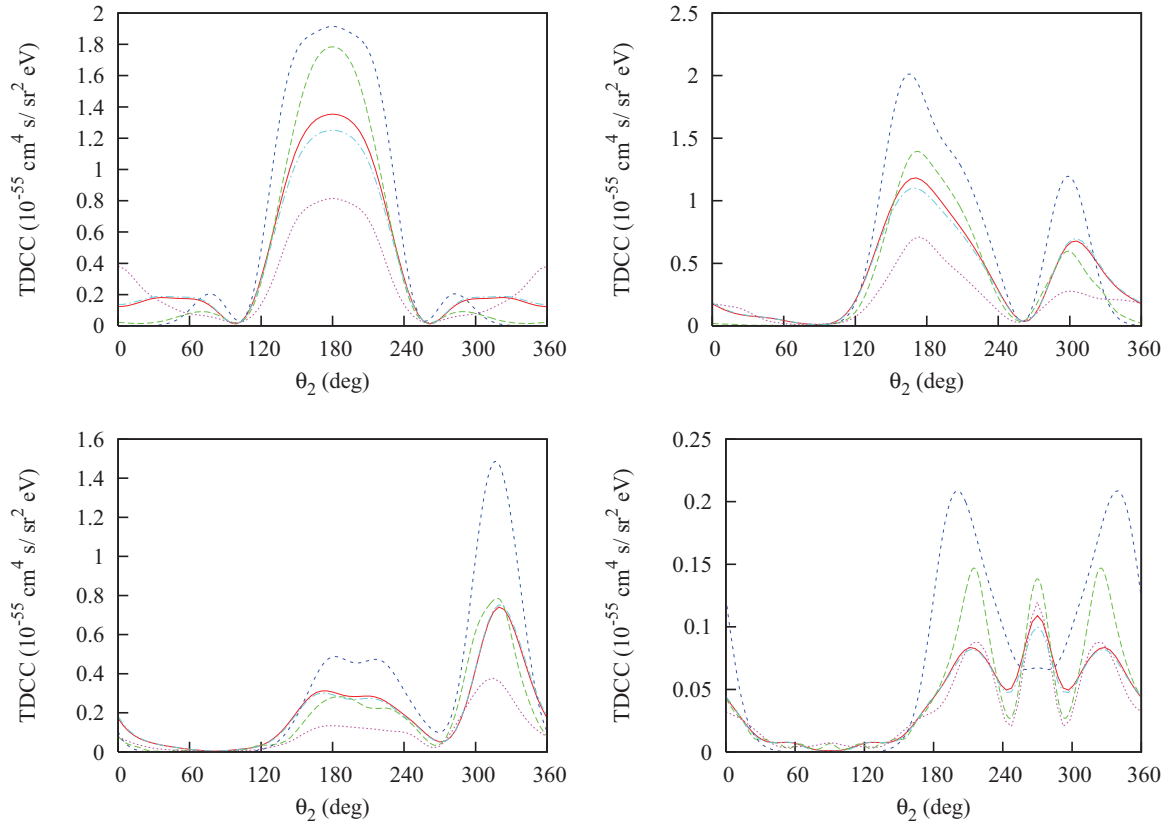


FIG. 2. (Color online) Same as in Fig. 1 for the parallel field orientation.

in detail in Ref. [8], where we checked that computed double ionization probabilities are practically constant for the values of R_0 in a wide interval, provided this interval is chosen so that the doubly ionized wave packet has enough time to leave the region where either $r_1 < R_0$ or $r_2 < R_0$. We performed this check in the present work to ensure that the value $R_0 = 10$ a.u. satisfies this requirement. With this recipe, it is immaterial whether we project the solution of the TDSE on the set of the Coulomb wave functions (as we do presently) or on the set of the continuum wave functions of the H₂⁺ ion (as we did in Ref. [8] and as it has been done in Ref. [7]). Indeed, at distances $r > R_0$, the Hamiltonian of the H₂ molecule differs from that of a single Coulomb center with $Z = 2$ in terms of the order of $(R/R_0)^2$ (R is the internuclear distance), which can be neglected.

The differential probability of TPDI can now be found as

$$P(\mathbf{k}_1, \mathbf{k}_2) = |\langle \Psi_{\mathbf{k}_1, \mathbf{k}_2}^- | \Psi(T_1) \rangle|^2, \quad (4)$$

where $\Psi(T_1)$ is the solution of the TDSE at the moment of time $T_1 = 8T$ corresponding to the end of the xuv pulse, and radial integrals are calculated as described above. The differential probability (4) is converted into the fully resolved triply differential cross section (TDCS) in a usual way. First, we take into account the fact that for the pulse of a finite duration (1) the energy is conserved only approximately, whereas the TDCS is defined on the energy shell which corresponds to the strict energy conservation. We can use a procedure [13] to

account for this fact. The corresponding formula reads

$$P^{\text{av}}(E_1, \hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2) = \frac{1}{E \sin 2\alpha} \int_0^\pi P(u \hat{\mathbf{k}}_1 \sin \alpha, u \hat{\mathbf{k}}_2 \cos \alpha) u du, \quad (5)$$

where E is the excess energy and parameter α determines energy sharing, so that energies of escaping electrons are $E_1 = E \sin^2 \alpha$ and $E_2 = E \cos^2 \alpha$, and $\hat{\mathbf{k}}_i = \mathbf{k}_i/k$. Thus obtained differential probability P^{av} is connected to the TDCS (in units of cm⁴ s/sr²/eV) as

$$\frac{d^3\sigma}{dE_1 d\hat{\mathbf{k}}_1 d\hat{\mathbf{k}}_2} = C \frac{P^{\text{av}}(E_1, \hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2)}{T_1^{\text{eff}}}, \quad (6)$$

where $C = 2^5 \pi^2 a_0^4 \tau \omega^2 c^{-2} E_0^{-4} / \text{Ry}$, $c \approx 137$ is the speed of light in atomic units, $a_0 = 0.529 \times 10^{-8}$ cm is the Bohr radius, $\tau = 2.418 \times 10^{-17}$ s is the atomic unit of time, E_0 is the peak strength of the laser field expressed in atomic units, and Ry ≈ 13.6 eV is the Rydberg constant. The Effective pulse duration T_1^{eff} introduced in Eq. (6) takes into account the particular pulse shape $f(t)$ used in Eq. (1) and is determined by the equation $\int_0^{T_1} f^4(t) \cos^4 \omega t dt = \frac{3}{8} T_1^{\text{eff}}$ [14,15]. For a pure monochromatic pulse with $f(t) = 1$ we would have $\int_0^{T_1} \cos^4 \omega t dt = \frac{3}{8} T_1$, which explains the appearance of the factor 3/8 in the formula for T_1^{eff} . For the pulse shape and length that we consider T_1^{eff} differs from the total pulse duration T_1 by about 20%.

In the present work, we calculate the TDCS for the cases of perpendicular and parallel field-molecule orientations.

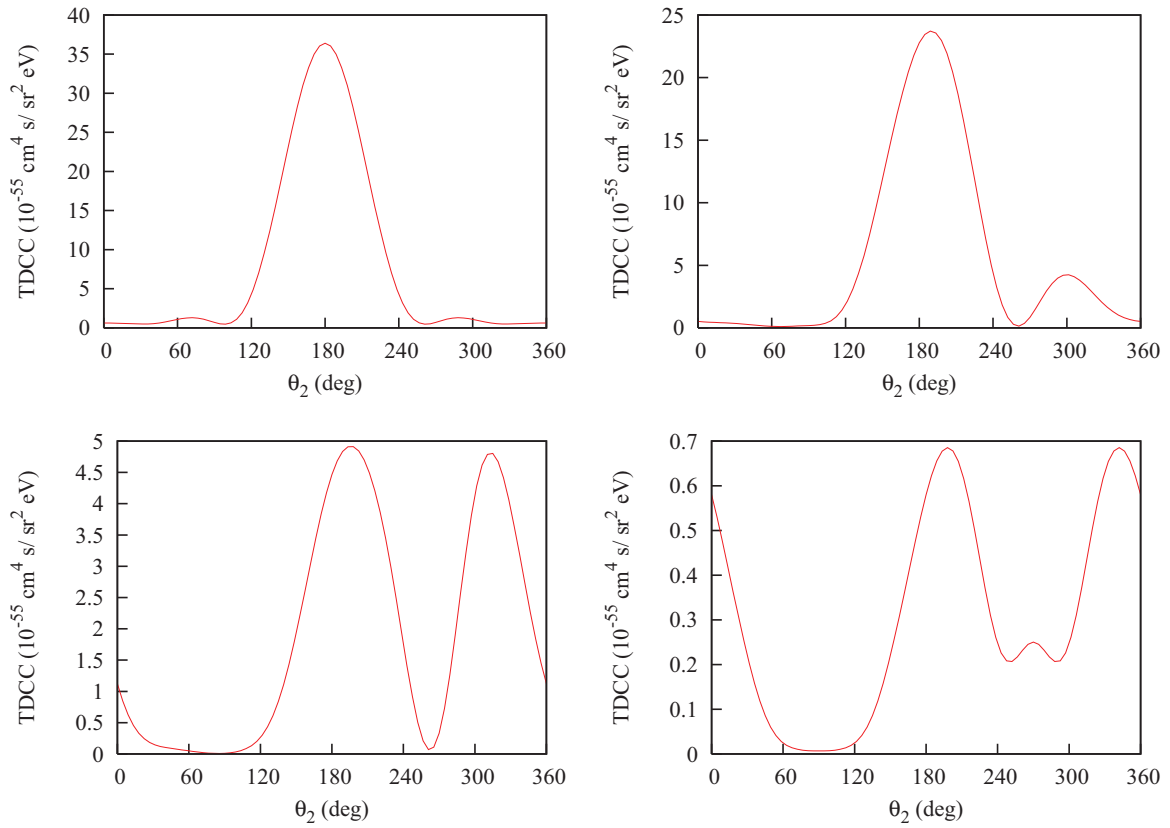


FIG. 3. (Color online) Coplanar TDCCS of double ionization of H_2 at a photon energy of 30 eV and unequal energy sharing for the perpendicular field-molecule orientation for $\theta_1 = 0^\circ$ (upper left), $\theta_1 = 30^\circ$ (upper right), $\theta_1 = 60^\circ$ (bottom left), and $\theta_1 = 90^\circ$ (bottom right) and for the energy of the fixed electron, $E_1 = 5.4$ eV.

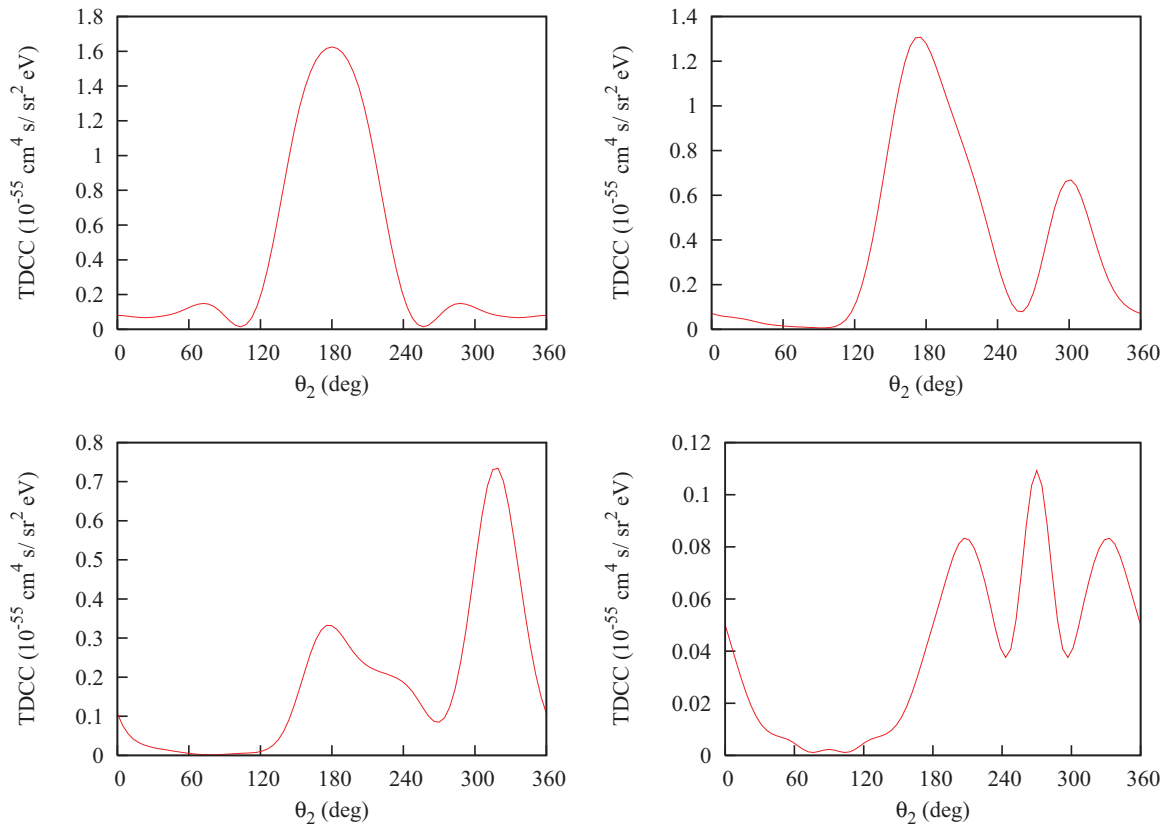


FIG. 4. (Color online) The same as in Fig. 3 for the parallel field-molecule orientation.

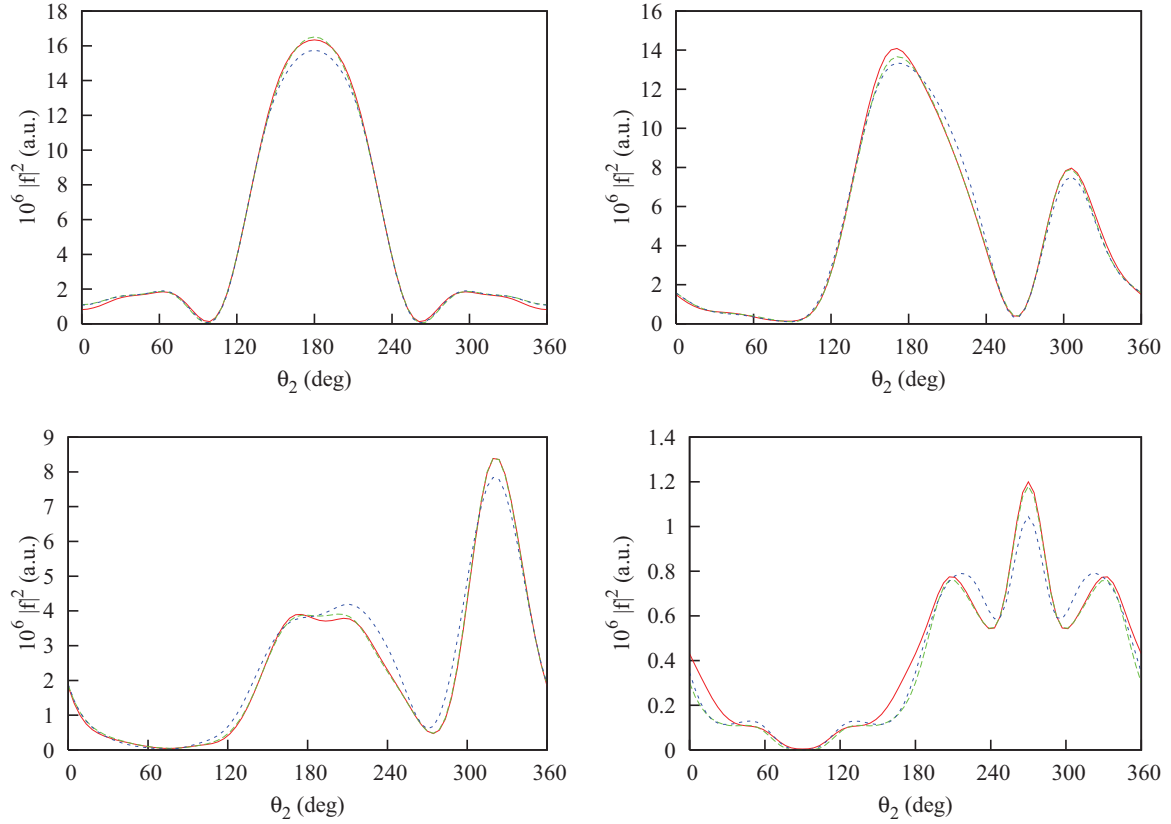


FIG. 5. (Color online) Double ionization probability of H₂ at photon energy of 30 eV and equal energy sharing for the parallel field-molecule orientation for $\theta_1 = 0^\circ$ (upper left), $\theta_1 = 30^\circ$ (upper right), $\theta_1 = 60^\circ$ (bottom left), and $\theta_1 = 90^\circ$ (bottom right). Shown are: probability computed using "true" (as explained in the text) amplitude (red) solid line, "reduced" amplitude (green) dashed line, results of the fitting procedure (blue) dots.

The TDCS results for the case of equal energy sharing are shown in Figs. 1 and 2, respectively. Here the coplanar kinematics is chosen in which both the photoelectrons are detected in the common plane of the polarization and internuclear axes. The escape angle of one of the photoelectrons relative to the polarization axis of light θ_1 is fixed, while the angle of the second photoelectron θ_2 varies in the full 2π range. These TDCS can be compared directly with the data [4,5], the results [6], and the recent calculation [7].

For the perpendicular field-molecule orientation (Fig. 1), the present TDSE results using both the length and velocity forms of the interaction Hamiltonian, the TDSE-PS results [7], and the TDCC data [4,5] all agree very well. The ECS calculation [6] is noticeably different in magnitude for all θ_1 values. For $\theta_1 = 90^\circ$ it is also different in shape. It is suggested in Ref. [7], that the ECS calculation [6] is time independent and uses effectively a pulse of an infinite length with a zero bandwidth. On the other hand, the other calculations are explicitly time dependent and employ a driving laser pulse with a nonzero bandwidth, which may induce doubly excited resonant states of H₂ to serve as intermediate states in the TPDI process. However, a very close agreement of these time-dependent calculations using different pulse parameters and forms of interaction Hamiltonian put such an explanation in doubt.

The case of the parallel field orientation presented in Fig. 2 is more difficult to analyze. In this case, the TDCS is an order of magnitude smaller than that for the perpendicular orientation. It is also known to converge much more slowly with the number of partial waves in expansion (3). Nevertheless, we find that our results agree quite well with the TDSE-PS results [7] for all the fixed angles except for $\theta_1 = 90^\circ$, where TDCS becomes extremely small. The agreement of our results with the TDCC calculation [4,6] is not so good. We performed our calculations for a field intensity of 3.5×10^{14} W/cm², which is intermediate between the intensity of 10^{14} W/cm² used in Ref. [7] and the much higher intensity of 10^{15} W/cm² used in the calculations in Refs. [4,6]. We tend to agree with the authors of Ref. [7] that the latter intensity may indeed be in a nonperturbative regime, which may account for the observed difference of the present results and the results of Ref. [7] on the one hand, and the data obtained in Refs. [4,6] on the other. Indeed, we may notice from the Fig. 2 that our TDCS are generally slightly lower than the TDCS obtained in Ref. [7], with the cross sections given in Refs. [4,6] being smaller yet. If we interpret this trend as a general manifestation of the field intensity effects, we obtain an explanation for the observed difference in the results.

In Figs. 3 and 4, we show the TDCS obtained for an unequal energy sharing ($E_1 = 5.4$ eV, $E_2 = 3.4$ eV) for the

same geometries as in Figs. 1 and 2. These TDCS are quite similar to their equal energy sharing counterparts. This similarity is, most likely, due to the fact that the underlying symmetric amplitudes of TPDI are much larger than the corresponding antisymmetric amplitudes. More on the amplitude analysis of the TPDI process can be found in our earlier work [16].

For the case of the parallel field-molecule orientation we also performed analysis of the amplitudes of the double photoionization process similar to the one we presented in Ref. [8] for the single-electron photoionization of a hydrogen molecule. In the case of atomic TPDI, the ionization amplitudes can be expressed in terms of the invariant combinations of the different vectors characterizing the process as follows [17]:

$$f(\mathbf{k}_1, \mathbf{k}_2) = f_0(\mathbf{e} \cdot \mathbf{e}) + f_1(\mathbf{e} \cdot \hat{\mathbf{k}}_1)^2 + f_2(\mathbf{e} \cdot \hat{\mathbf{k}}_2)^2 + f_s[(\mathbf{e} \cdot \hat{\mathbf{k}}_1)(\mathbf{e} \cdot \hat{\mathbf{k}}_2)], \quad (7)$$

where $\mathbf{e}$ is the EM field polarization vector. The functions f_0 , f_1 , f_2 , and f_s in Eq. (7) depend only on the electron energies and the angle $\theta_{12} = \arccos \hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2$ between the electron momenta.

In the case of molecular TPDI, Eq. (7) should be, in principle, modified, since we have an additional vector in the problem, the internuclear vector $\mathbf{R}_N$, to form invariant scalar products quadratic in $\mathbf{e}$. We shall see, however, that, at least for the case of the coplanar geometry and parallel field-molecule orientation, these purely molecular effects can be safely neglected.

In the case of the equal energy sharing between the two electrons which we only consider here, Eq. (7) can be rewritten as

$$f(\mathbf{k}_1, \mathbf{k}_2) = A(\theta_{12})[\cos^2 \theta_1 + \cos^2 \theta_2] + B(\theta_{12}) \cos \theta_1 \cos \theta_2 + C(\theta_{12}), \quad (8)$$

where θ_1 and θ_2 are angles between the photoelectron momenta and the polarization vector. For a simpler case of the one-photon double ionization, the only symmetric amplitude can easily be extracted from the TDCS data simply dividing the TDCS by a corresponding kinematic factor as we did in Ref. [8]. In the present case of TPDI, where several symmetric amplitudes are to be determined, we have to employ a slightly more complicated procedure.

We can expand the amplitude function $A(\theta_{12})$ in a series of Legendre polynomials:

$$A(\theta_{12}) = \sum_0^{L_{\max}} A_l P_l(\theta_{12}). \quad (9)$$

Analogous expansions can be written for the amplitude functions B and C . To be consistent we choose $L_{\max} = 6$ in Eq. (9) which is the highest angular momentum retained in Eq. (3) representing the two-electron wave function. We obtain thus a set of the coefficients A_l , B_l , and C_l which we treat as fitting parameters in the following minimization procedure.

Using solution of the TDSE, we compute ionization amplitudes for a set of different electron momenta pairs $\mathbf{k}_1^i, \mathbf{k}_2^i$. Typically, we use a set of about $M = 60$ different pairs $\mathbf{k}_1^i, \mathbf{k}_2^i$. For each pair $\mathbf{k}_1^i, \mathbf{k}_2^i$, we actually compute two amplitudes,

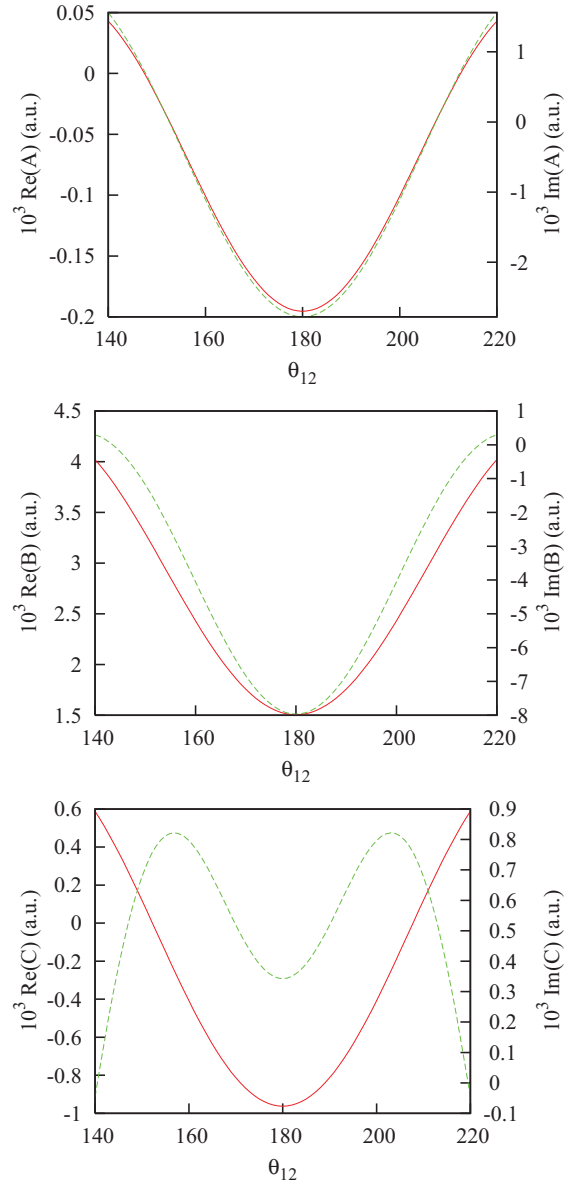


FIG. 6. (Color online) Real [(red) solid line] and imaginary [(green) dashed line] parts of the coefficients A , B , and C as functions of angle θ_{12} between the electron momenta near $\theta_{12} = 180^\circ$.

a “true” amplitude $f(\mathbf{k}_1^i, \mathbf{k}_2^i)$ and a “reduced” amplitude $f_r(\mathbf{k}_1^i, \mathbf{k}_2^i)$ which we obtain by retaining the terms with $|l_1 - l_2| \leq 2$ in the partial wave expansion (3). Knowledge of f_r is important since it tells us if Eq. (8) can be used to describe molecular TPDI in the first place. We remind the reader that, strictly speaking, this equation applies only for atomic TPDI. The tensorial structure of Eqs. (8) and (9) tells us that in the partial wave expansion of the corresponding amplitudes only the terms with $|l_1 - l_2| \leq 2$ can be present. If, therefore, we find that condition $|f_r - f| \ll |f|$ is violated, parametrization based on Eqs. (8) and (9) cannot be used. We checked that the condition $|f_r - f| < 0.05|f|$ holds for each pair $\mathbf{k}_1^i, \mathbf{k}_2^i$ that we use.

By feeding the fitting parameters (A_l, B_l, C_l) into Eqs. (8) and (9), we can compute the trial value $f_t(\mathbf{k}_1^i, \mathbf{k}_2^i)$ of the

ionization amplitude. We define a functional,

$$S(A_l, B_l, C_l) = \sum_{i=1}^M |f_r(\mathbf{k}_1^i, \mathbf{k}_2^i) - f_l(\mathbf{k}_1^i, \mathbf{k}_2^i)|^2, \quad (10)$$

which can be easily minimized with respect to the set of the fitting parameters (A_l, B_l, C_l) reducing in a standard way the minimization of the bilinear functional S defined in Eq. (10) to the solution of a linear system of equations for the coefficients (A_l, B_l, C_l).

This procedure is illustrated in Fig. 5, where we show the ionization probabilities for the same geometries as in Fig. 2 computed using the true amplitude, the reduced amplitude, and the amplitude provided by the minimization procedure that we described above.

We see that the results of the three calculations are quite close to each other, thereby confirming the validity and consistency of the whole procedure. The probabilities, as functions of the angle θ_2 , have angular shapes very similar to the cross sections displayed in Fig. 2. This is to be expected since the TDCS as defined by Eq. (5) is a suitably averaged probability distribution. Thus the obtained amplitude functions A , B , and C are shown in Fig. 6.

The amplitudes A and B have a Gaussian form in agreement with the Wannier-type theories of the double photoionization [18–21]. The amplitude C is clearly non-Gaussian. We can possibly explain this fact by noting that this amplitude is the smallest of all the amplitudes used in the parametrization (7). It may be that the accuracy of the description of the double

ionization based on the quadratic expansion of the electron potential near the saddle point, which ultimately leads to the Gaussian amplitudes [19,20], is not sufficient in this case.

To summarize, we performed time-dependent calculations of the TPDI process in the aligned H₂ molecule. We found that under the equal energy sharing conditions our results, both in the length and velocity gauges, agree quite well with each other and with the TDSE-PS results [7]. This gives certain confidence in the validity and accuracy of our results, which, for a problem as complicated as molecular TPDI, is gratifying. We also calculated TDCS for an unequal energy sharing for both parallel and perpendicular geometries and found them similar to the equivalent set of TDCS for the equal energy sharing.

We performed analysis of the photoionization amplitudes using invariant parametrization based on Eq. (8) for the case of equal energy sharing and the parallel field-molecule orientation. We found that all the amplitudes in Eq. (8), with the exception of the amplitude C , have simple Gaussian form in agreement with the Wannier-type theories of the double photoionization process.

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