Triple-differential cross section for single ionization of H_2 by electron impact*

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The triple-differential cross section (TDCS) for the (e, 2e) ionization of a hydrogen molecule is calculated using the molecule distorted-wave Born approximation (MDWBA). Distorted waves are obtained by solving momentum-space coupled-channel Lippmann–Schwinger equations, including the ground state and the lowest-lying electronic state of $b^3\Sigma_u$. TDCSs at the incident energy 100 eV in coplanar asymmetric geometry are reported. The present calculations are compared with the available experimental measurements and the theoretical results.

Keywords: (e, 2e) collisions, hydrogen molecule, triple differential cross section

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1. Introduction

Electron-impact ionization of atoms and molecules is one of basic processes in atomic physics. It is important in both fundamental studies and practical applications. The latter includes astrophysics, plasma physics, fusion physics, surface science, and radiation physics.[1] Studies of (e, 2e) reaction on atoms and molecules are powerful tools to understand the structures and the mechanisms of the ionization of atoms and molecules.[2–6] The principle aim in the study of (e, 2e) ionization of atoms and molecules is to determinate the multiply differential cross sections which demand correct descriptions of the continuum state of incident, scattering and eject electrons. In the atomic target, this has been done by using Coulomb and distorted-wave functions. In the case of molecular targets, the description of the continuum electrons in the field of two Coulomb or distorted centers is much more complex because of the properties of molecule multicenters.

As the simplest molecule, a hydrogen molecule is the test target containing most of the complications of molecule targets, which has received a lot of attention.[7–14] A number of experimental studies have been conducted on the electron scattering by hydrogen molecules.[7–10] The triple-differential cross section (TDCS) was measured in a relative scale at intermediate incident energies by Jung *et al.*[7] and the absolute TDCS was obtained at much higher impact energies by Cherid *et al.*[10] In the theoretical aspect, several approximations were developed to describe the dynamics of (e, 2e) ionization on molecule targets by electron-impact: the first order of the Born series (FBA) in which the incident and the scattered electrons were represented by plane waves; and distorted-wave approximation based on the Schwinger variational iterative method. The two-effective-center approximation (TEC)[11] and the two-center continuum approximation (TCC)[12] introduced by Weck *et al.* have been constructed, in which the continuum wave function as a product of a plane wave centered in the molecule mass center and the interaction of the ionized electron with each molecular center was included. Although those varieties of approximations have been performed to calculate the TDCS for electron-impact ionization of H_2, significant disagreements between the experimental and the theoretical results still exist. Recently, Stia *et al.*[13] developed a molecular Brauner–Briggs–Klar (MBBK) model within the framework of the TEC method including the correlated motion of the unbound charged particles in the final channel of the reaction. More recently, Liu *et al.*[14] studied the electron-impact ionization of a hydrogen molecule by using the molecule distorted-wave Born approximation (MDWBA) method, in which the continuum wave functions of electrons are calculated using scattering potentials in momentum space.

Momentum-space methods in electron–molecule scattering theory are attractive from two points of view. First, differential cross sections are expressed in terms of the initial and final electron momenta so that the amplitudes involved in the calculation are directly related to the experiment. Second, the electron–molecule system has a single cen-

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ter in momentum space, whereas a tractable coordinate-space representation is multicentered. Distorted-wave approximations normally represent the interaction of a continuum electron with the remainder of the system by potential scattering. However, in this work, a potential scattering model is not good enough for the continuum electrons. The aim of the present work is to investigate the electron-impact ionization process in the frame of MDWBA, taking account of the coupled potential effects. The incident, scattered and ejected electron distorted-wave functions are obtained by solving the momentum-space coupled-channel Lippmann–Schwinger equations. Target states ($X^1\Sigma_g$ and $b^3\Sigma_u$) are expressed using single configuration self-consistent-field (SCF) orbitals which are linear combinations of primitive Gaussians centered at the atomic sites. Direct and exchange matrix elements are calculated by analytic angular and numerical radial integrations. In the static-exchange-optical calculations of the T-matrix in the second-order term, two channels are included in the present work.

In the present paper, we report the TDCS for electron-impact ionization of H_2 in coplanar asymmetric geometry at the incident energy of 100 eV, and compare it with the available experimental measurements and theoretical results.

2. Theoretical details

The TDCS for electron-impact ionization is given in atomic units by

$$\frac{\mathrm{d}^3\sigma}{\mathrm{d}\Omega_\mathrm{f}\mathrm{d}\Omega_\mathrm{s}\mathrm{d}E_\mathrm{f}} = (2\pi)^4\frac{k_\mathrm{f}k_\mathrm{s}}{k_0}\sum_\mathrm{av}|\langle \boldsymbol{k}_\mathrm{f}\boldsymbol{k}_\mathrm{s}|T|\psi\boldsymbol{k}_0\rangle|^2, \tag{1}$$

where the notation $\sum_\mathrm{av}$ represents a sum over the final and average over initial magnetic and spin degeneracies, and symmetric normalization has been adopted. The momentum of the incident electron is $\boldsymbol{k}_0$, $\boldsymbol{k}_\mathrm{f}$, and $\boldsymbol{k}_\mathrm{s}$ are the momenta of the fast and slow outgoing electrons, ψ is the active bound orbital of the ground-state target.

In the distorted-wave Born approximation,[15] the amplitude is given as

$$\langle \boldsymbol{k}_\mathrm{f}\boldsymbol{k}_\mathrm{s}|T|\psi\boldsymbol{k}_0\rangle = \langle \chi^{(-)}(\boldsymbol{k}_\mathrm{f})\chi^{(-)}(\boldsymbol{k}_\mathrm{s})|V|\psi\chi^{(+)}(\boldsymbol{k}_0)\rangle, \tag{2}$$

where V is the Coulomb potential, and $|\chi_0^{(\pm)}(\boldsymbol{k})\rangle$ is the distorted-wave with outgoing and incoming boundary conditions.

Within the Born–Oppenheimer (BO) approximation, the electronic states are separated from the vibration-rotation states. We treat the rotation of the molecules as an adiabatic process and average over the orientations of the molecule. For the ground state of target vibrations, we retain the assumption that vibrations and rotations are uncoupled.

We make the partial wave expansion for the V-matrix element as

$$\begin{aligned}
&\langle \chi^{(-)}(\boldsymbol{k}_\mathrm{f})\chi^{(-)}(\boldsymbol{k}_\mathrm{s})|V|\psi\chi^{(+)}(\boldsymbol{k}_0)\rangle \\
&= \sum_{L'M'L''M''LM}\langle \hat{\boldsymbol{k}}_\mathrm{f}|L'M'\rangle\langle \hat{\boldsymbol{k}}_\mathrm{s}|L''M''\rangle \\
&\quad\times\langle \chi^{(-)}(\boldsymbol{k}_\mathrm{f})L'M'\chi^{(-)}(\boldsymbol{k}_\mathrm{s})L''M''\|V\|\psi\chi^{(+)}(\boldsymbol{k}_0)LM\rangle \\
&\quad\times\langle LM|\hat{\boldsymbol{k}}_0\rangle.
\end{aligned} \tag{3}$$

Then, we transform the initial and final momenta into momenta in the laboratory frame. The transformation is effected by the rotation matrices

$$\begin{aligned}
&\langle \chi^{(-)}(\boldsymbol{k}_\mathrm{f})\chi^{(-)}(\boldsymbol{k}_\mathrm{s})|V|\psi\chi^{(+)}(\boldsymbol{k}_0)\rangle \\
&= \sum_{LML'M'L''M''\mu''\mu'}\sqrt{\frac{2L+1}{4\pi}}D^{L'*}_{M'\mu'}D^{L''*}_{M''\mu''}Y_{L'\mu'}(\hat{k}^z_\mathrm{f})Y_{L''\mu''}(\hat{k}^z_\mathrm{s}) \\
&\quad\times\langle \chi^{(-)}(k_\mathrm{f})L'M',\chi^{(-)}(k_\mathrm{s})L''M''\|V\|\psi\chi^{(+)}(k_0)LM\rangle \\
&\quad\times Y_{L0}(\hat{k}^z_0)D^L_{M0}.
\end{aligned} \tag{4}$$

We choose the positive z direction as the incoming direction of momentum k_0.

The distorted-waves are approximately written as

$$\begin{aligned}
|\chi^{(+)}(\boldsymbol{k}_0)\rangle &= |\boldsymbol{k}_0\rangle + \frac{T}{E_0^{(+)}-H}|\boldsymbol{k}_0\rangle, \\
|\chi^{(-)}(\boldsymbol{k}_\mathrm{f})\rangle &= |\boldsymbol{k}_\mathrm{f}\rangle + \frac{T}{E_\mathrm{f}^{(-)}-H}|\boldsymbol{k}_\mathrm{f}\rangle, \\
|\chi^{(-)}(\boldsymbol{k}_\mathrm{s})\rangle &= |\boldsymbol{k}_\mathrm{s}\rangle + \frac{T}{E_\mathrm{s}^{(-)}-H}|\boldsymbol{k}_\mathrm{s}\rangle.
\end{aligned} \tag{5}$$

Substituting Eq. (5) into Eq. (4), the reduced V-matrix element, which neglects the second- and third-order terms, is given by

$$\begin{aligned}
&\langle \chi^{(-)}(k_\mathrm{f})L'M',\chi^{(-)}(k_\mathrm{s})L''M''\|V\|\psi\chi^{(+)}(k_0)LM\rangle \\
&\simeq \langle k_\mathrm{f}L'M',k_\mathrm{s}L''M''\|V\|\psi k_0LM\rangle \\
&\quad+\langle k_\mathrm{f}L'M',k_\mathrm{s}L''M''\|\frac{VT}{E_0-H}\|\psi k_0LM\rangle \\
&\quad+\langle k_\mathrm{f}L'M',k_\mathrm{s}L''M''\|\frac{TV}{E_\mathrm{f}-H}\|\psi k_0LM\rangle \\
&\quad+\langle k_\mathrm{f}L'M',k_\mathrm{s}L''M''\|\frac{TV}{E_\mathrm{s}-H}\|\psi k_0LM\rangle,
\end{aligned} \tag{6}$$

where

$$\begin{aligned}
&\langle k_\mathrm{f}L'M',k_\mathrm{s}L''M''\|\frac{VT}{E-H}\|\psi k_0LM\rangle \\
&= \sum_{jL'''M'''}\int \mathrm{d}qq^2\langle k_\mathrm{f}L'M'k_\mathrm{s}L''M''\|V\|\psi_jL'''M'''q\rangle \\
&\quad\times\frac{1}{E^+-\frac{1}{2}q^2-\varepsilon_j}\langle qL'''M'''\psi_j\|T\|\psi k_0LM\rangle.
\end{aligned} \tag{7}$$

Here, E_0, E_f, and E_s are the energy of the incident, fast- and slow-outgoing electrons, respectively, ε_j is the energy of the corresponding molecular orbital ψ_j.

The T-matrix element for transition from the initial channel $|0\boldsymbol{k}\rangle$ to the final channel $|i\boldsymbol{k}'\rangle$ is calculated by solving the coupled-channel Lippmann–Schwinger equations[16]

$$\langle \boldsymbol{k}'i|T|0\boldsymbol{k}\rangle = \langle \boldsymbol{k}'i|V^{(Q)}|0\boldsymbol{k}\rangle + \sum_n \int \mathrm{d}^3\boldsymbol{q}\langle \boldsymbol{k}'i|V^{(Q)}|n\boldsymbol{q}\rangle \times \frac{1}{E^{(+)} - q^2/2 - \varepsilon_n}\langle \boldsymbol{q}n|T|0\boldsymbol{k}\rangle, \quad (8)$$

where $\boldsymbol{k}$ and $\boldsymbol{k}'$ are the incoming and outgoing momenta of the projectile electron, respectively, $|n\rangle$ denotes the n-th target state of intermediate channels, $V^{(Q)}$ represents the static-exchange potential plus the *ab initio* equivalent-local optical polarization potential in electron–molecule collisions, which was given in Ref. [16]. The electronic states of the molecule are given in terms of the electronic Hamiltonian H_{T}

$$(\varepsilon_n - H_{\mathrm{T}})|n\rangle = 0. \quad (9)$$

The molecular orbital ψ_j is represented by a linear combination of atomic orbitals (LCAO) centered at the nuclear sites R_n as

$$\langle r|\psi_j\rangle = \sum_{gnqk} c_{gnqkj} \mathrm{e}^{-\alpha_{gnqk}|\boldsymbol{r}+\boldsymbol{R}_n|^2} \sum_{lm} \gamma_{pnlm} r^l Y_{lm}(\hat{r}), \quad (10)$$

where q labels the symmetry of the atomic orbitals (e.g., S, P_x, P_y, P_z, etc.), k labels the atomic orbital, which is a linear combination of primitive Gaussians, g labels the primitive Gaussians, j labels the molecular orbitals, and n labels the nuclear sites. The Gaussians in Eq. (10) are expanded in spherical Bessel functions of imaginary argument $i_\sigma(x)$ as[17]

$$\mathrm{e}^{-\alpha_i|\boldsymbol{r}+\boldsymbol{R}_i|^2} = \mathrm{e}^{-\alpha_i(r+R_i)^2} \sum_{\sigma\chi} (-1)^\sigma (4\pi) i_\sigma(2\alpha_i r R_i) Y^*_{\sigma\chi}(\hat{r}) Y_{\sigma\chi}(\hat{\boldsymbol{R}}_i). \quad (11)$$

The reduced V-matrix element of the direct term and exchange term are given in Ref. [14].

In the present calculation, the truncation parameters used for the expansion of the continuum orbits, as well as the matrix elements, are $L'_{\max} = 8$, $L''_{\max} = 10$, $L_{\max} = 6$, and $M_{\max} = 6$. The calculation results have already been converged with these cutoff parameters.

3. Results and discussion

Triple-differential cross sections of electron-H_2 collision as a function of the ejection angle θ_{s} at the impact energy $E_{\mathrm{i}} = 100$ eV and the emission energy $E_{\mathrm{s}} = 4.5$ eV are displayed in Figs. 1 and 2. The angular variable defines the direction of the slow outgoing electron, relative to the direction of the incident electron, whereas the directions of the fast outgoing electron are fixed at $\theta_{\mathrm{f}} = 15^\circ$ and 7°, respectively. Present results are compared with experimental data[7] and other theoretical results (TEC[11] and those from MBBK[13]). Since experimental data of Jung *et al.*[7] were obtained in a relative scale, we extracted experimental points from the experimental curves[7] and normalized them to present results at the maximum value corresponding to the binary encounter peak, in order to make the comparison possible.

Bray *et al.*[18] emphasized the importance of the multichannel effect when studying the electron-impact ionization of atoms. Moreover, Monzani *et al.*[19] believed that the neglect of the multichannel effect is attributed to the major discrepancies between their results and the experimental data. To investigate the multichannel effect, we also present the corresponding single-channel results of molecule distorted-wave Born approximation (SC-MDWBA).[14]

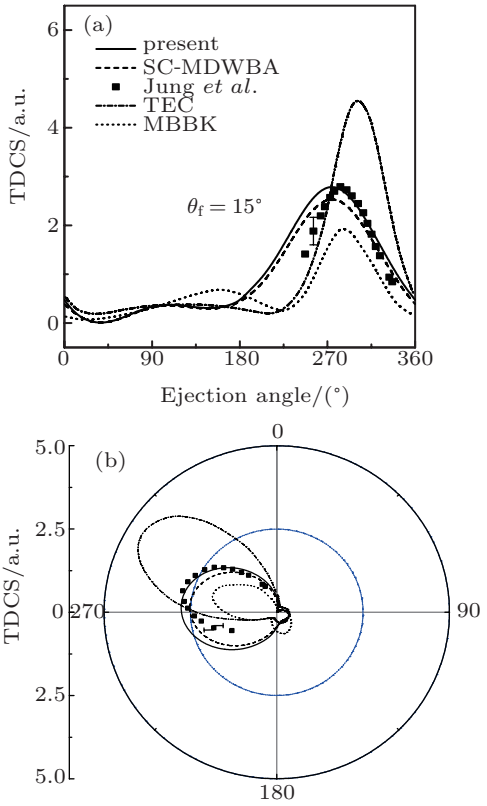


Fig. 1. TDCS as a function of the ejection angle θ_{s} presents in cartesian coordinates (a) and spherical coordinates (b). Here, the impact energy $E_{\mathrm{i}} = 100$ eV, the emission energy $E_{\mathrm{s}} = 4.5$ eV, and scattering angles $\theta_{\mathrm{f}} = 15^\circ$. Present results, SC-MDWBA results,[14] TEC results,[11] MBBK results,[13] and experimental results[7] are represented by the solid line, dashed line, dash-dotted line, dotted line, and solid squares, respectively.

In general, the present results are in good agreement with the experimental data. As shown in the figures, the curve of present results displays a broader binary-peak than those of the experimental data[7] and other theoretical results.[11,13] The present predictions for the binary-peak position show a slight shift towards smaller angles with respect to experimental data,[7] while the results of TEC[11] and MBBK[13] shift to larger angles. We believe that neglecting the interaction of the outgoing electrons may be attributed to this discrepancy. The present results overestimate the experimental data at an-

gles smaller than the binary peak. The values of the present TDCS are larger than that of the MBBK[13] at the binary-peak region, while the situation is reversed at the recoil peak angular domain. The TEC[7] model produces the largest binary peak, while it gives a similar behavior with present results at the recoil-peak range. Comparing with SC-MDWBA results[14], the increase in the binary region of the present results shows the multichannel effects of including the lowest-lying excitation state of $b^3\Sigma_u$, when solving coupled-channel Lippmann–Schwinger integral equations. As limited channels are considered in the present status, modification of the TDCS is not remarkable. The exact role played by other excited states will be taken into account in the future.

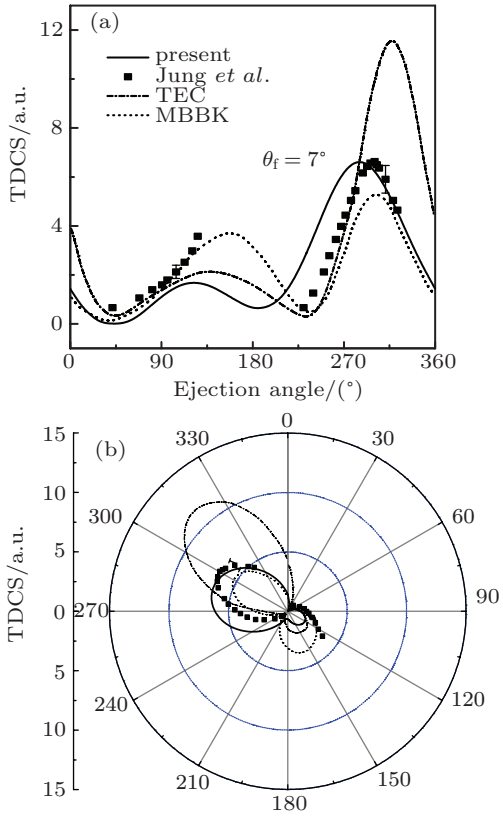


Fig. 2. TDCS as a function of the ejection angle θ_s presented in Cartesian coordinates (a) and spherical coordinates (b). Here, the impact energy $E_i = 100$ eV, the emission energy $E_s = 4.5$ eV, and scattering angles $\theta_f = 7^o$. Present results, TEC results,[11] MBBK results,[13] and experimental results[7] are represented by the solid line, dash-dotted line, dotted line, and solid squares, respectively.

4. Conclusions

In summary, the present work reports a MC-MDWBA calculation for the electron-impact ionization of H_2 at incident energy of 100 eV. The inclusion of the channel-coupling effect in solving the T-matrix in the present model has a reasonable influence on the results of coplanar asymmetric geometry for electron-H_2 scattering ionization study. The discrepancy between the present results and the experiment data is probably due to the neglect of interactions between higher lying channels and the post-collision interactions of the two outgoing electrons in the present model. In their present status, MC-MDWBA calculations are restricted to two states for coupled states expansions. More coupled channels will be incorporated in the following work. We hope that improvements will remove the remaining disagreements.

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