Intermediate-energy electron-impact dissociative ionization-excitation of molecular hydrogen

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Abstract

We have implemented three variants of the exterior complex scaling procedure in prolate spheroidal coordinates (PS-ECS) to study the dissociative electron impact ionization-excitation of hydrogen molecule, where the emerging electrons and one of the protons are detected in coincidence for the first time in a recent experiment. In the first variant, designated PSECS-1B, the two target electrons are treated \textit{ab initio} while the interaction of the incident-scattered electron is taken into account using the first term of the Born series. In the second, PSECS-2BCD, the second Born term is introduced in the dipole approximation. In the third approach, designated PSECS-SW, applied to the ionization-excitation to the $2p\sigma_u$ level of $\text{H}_2^+$, the multi-configurational single active electron approximation is used for the target, while the interaction of the incident electron with the target is described \textit{ab initio}. Our results agree partially with those of a recent experiment which is in progress.

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I. INTRODUCTION

Simple (e,2e) ionization of atomic and molecular targets designates complete inelastic electron-target collision experiments, where the scattered and the ejected electrons from the target are detected in coincidence [1–4]. In the case of diatomic molecules, one has to separate the vibrational and rotational movements (see for that [5]) and average over all possible directions of the molecule in the laboratory frame with respect to which the emerging electrons are detected.

In recent years many molecular orientation resolved experiments have been undertaken [6–9]. The main experimental methods to realize these experiments are either by aligning the target before the collision by exciting its rotational movement by a polarized laser [10] or/and by detecting the emerging dissociated nuclei in coincidence with the ejected and scattered electrons. Recently, the latter approach was applied to the simple ionization excitation of molecular hydrogen H$_2$ to the 2$s\sigma_g$, 2$p\pi_u$ and 2$p\sigma_u$ levels of H$_2^+$ [7]. Compared to the situation in the COLTRIMS method [8], the emerging proton is detected here with higher energy resolution, which represents a quite important step in this domain. We will designate this type of coincidence experiments (e,2e+p) simple ionization.

We have in the past determined the multiply differential cross section of this process [11–13] for high (5 keV) incident energy values and have observed interesting interference phenomena and have shown like in [8, 9] that the proton emerges preferentially in the direction of the momentum vector transferred to the system by the incident electron [13, 14].

In the present paper, we apply the exterior complex scaling method in prolate spheroidal coordinates (PS-ECS) [15] to the determination of the multi-fold differential cross section MDCS of the ionization-excitation of H$_2$ to the 2$s\sigma_g$, 2$p\pi_u$ and 2$p\sigma_u$ levels of H$_2^+$. Our aim is to gain physical insight in this complex problem, to interpret the results of [7] and give theoretical guidance to near future experiments.

II. THEORY

We apply three variants based on the exterior complex scaling method in prolate-spheroidal coordinates (PS-ECS) developed in [15]. This method is based on the solution of
the six-dimensional driven Schrödinger equation in prolate spheroidal (elliptic) coordinates

\[ (\hat{H} - E)\psi^+(+) = -\hat{\mu}\psi_i \]  

(1)

where \( \psi^+(+) \) is the perturbed part of the wave function, with the boundary conditions of an out-going wave provided by the exterior complex scaling (ECS). The \( E \) is the final energy of the electrons are described by \( \psi^+(+) \).

The scattering of the electron on two-electron molecule results in a nine-dimensional Schrödinger equation, which we reduce first to a six-dimensional equation. The nuclei are considered to be fixed during the process. The choice of the appropriate approach to do this reduction depends on the mechanism that we want to study. In the ionization-excitation, one of the target electrons can be ejected by the incident electron, while the second can be excited by a second impact (sequential mechanism). We can also have the excitation of the second electron by the inter-electron correlations in the initial target state (kick-off mechanism), or in the final target state (final-state scattering).

For a relatively fast incident electron, the natural approach is the application of the Born series for the incident and scattered electron. In this approach, \( \psi^+(+)(r_1, r_2) \) represents the wave function of the (unknown) final state of the target electrons with coordinates \( r_1 \) and \( r_2 \) in Eq.(1). The \( \hat{H} \) is the Hamiltonian of the target. The \( E \) designates the final energy of the target electrons and \( \psi_i(r_1, r_2) \) the wave function of the initial state of the target. The excitation operator \( \hat{\mu} \) takes into account the effect of the incident electron.

The first order Born term of the excitation operator has the form \[ \hat{\mu}_{1B} = -\frac{1}{2\pi} \langle k_s | V | k_i \rangle = \frac{2}{K^2} \left[ e^{iK \cdot r_2} + e^{iK \cdot r_1} - e^{iK \cdot R/2} - e^{-iK \cdot R/2} \right] \]  

(2)

where \( R \) is internuclear distance, and \( R = Rn_R \) is the vector with the orientation coinciding with the molecular axis orientation. Next, \( V \) is the potential energy term of the interaction of the molecule with the incident electron, \( k_i \) is its momentum, \( k_s \) represents the momentum of the scattered electron, and \( K \) the momentum transfer. Here we assume \( |k\rangle = \exp(iK \cdot r_0) \), \( r_0 \) being the position vector of the fast incident/scattered electron. With this choice, the kick-off and the final-state scattering processes are taken into account, but not the sequential mechanism. This will be done by considering the second Born effects in a relatively simplified manner \[16\]. In fact, we add to the excitation operator \( \hat{\mu} \) the corrected double dipole
approximation of the second Born term \[16\]

\[
\hat{\mu}_{2BC}(r_1, r_2) = \sum_{M_1, M_2 = -1}^{1} \mathcal{M}^{M_1 M_2}(x_{1M_1} + x_{2M_1})(x_{1M_2} + x_{2M_2}). \tag{3}
\]

Here \(\mathcal{M}^{M_1 M_2}\) is the second-order cross derivative term of the Born series for the scattered electron in the closure approximation, \(x_{1M}\) and \(x_{2M}\) are components of the vectors \(r_1\) and \(r_2\), respectively. The two dipole factors act on both target electrons.

The Born series approach is not well adapted to our problem, where we have an intermediate electron impact energy of 178 eV, and the plane wave description of the incident-scattered electron is not quite appropriate. So we implement an alternative approach, in which the interaction of the incident electron with the target is described precisely, but the interaction between the target electrons in the final state is omitted. The initial state of \(H_2^+\) can be expanded over products of bound states of \(H_2^+\)

\[
\Phi_0(r_1, r_2) = \sum_{n_1 l_1 m, n_2 l_2} c_{n_1 l_1 n_2 l_2 m} \varphi_{n_1 l_1 m}(r_1) \varphi_{n_2 l_2 - m}(r_2), \tag{4}
\]

where \(n, l, m\) is the set of spheroidal quantum numbers, which specify the bound states \(\varphi_{nlm}(r)\) of \(H_2^+\), \(c_{n_1 l_1 n_2 l_2 m}\) are the expansion coefficients. Each of the terms in the sum (4) can be considered as an electron configuration. Under the assumption, that the ejection of one of the electrons does not change the other electron state, and the final ion state is \(\varphi_{n_2 l_2 - m}\), the initial state of the electron to be ejected may be described by the function

\[
\varphi_0(r_1) = \langle \varphi_{n_2 l_2 - m}(r_2) | \Phi_0(r_1, r_2) \rangle. \tag{5}
\]

Then, following (4), the initial state of the electron to be ejected may be expressed as

\[
\varphi_0(r_1) = \sum_{n, l} c_{nl n_2 l_2 m} \varphi_{nlm}(r_1). \tag{6}
\]

Each term of this expansion can be associated with the unperturbed wave function, describing a system “bound electron + incoming fast electron”

\[
\psi_i(r_0, r_1) = \frac{1}{\sqrt{2}} \left[ \exp(i k_i \cdot r_0) \varphi_{nlm}(r_1) + (-1)^S \exp(i k_i \cdot r_1) \varphi_{nlm}(r_0) \right], \tag{7}
\]

where \(S\) is the total spin of the incident and target electrons possessing the value \(S = 0\) with probability \(1/4\), or the value \(S = 1\) with probability \(3/4\). This function may be substituted in right-hand-side of Eq.(1) allowing to obtain the wave function \(\psi^{(+)}(r_0, r_1)\) of the scattered
and the ejected electrons. For this aim in Eq. (1) \( E \) should be set equal to the total energy of the incident and the target electron, and the operator \( \hat{\mu} \) equal to the potential of the interaction between the incident and the target electrons

\[
\hat{\mu} = \frac{1}{|\mathbf{r}_0 - \mathbf{r}_1|}.
\] (8)

In turn, from the scattering wave function \( \psi^{(+)}(\mathbf{r}_0, \mathbf{r}_1) \) one may extract the scattering amplitude \([17]\). The latter is in fact the amplitude of the impact ionization of \( \text{H}_2^+ \) initially being in \( \varphi_{nlm} \) state. Since the electron spin directions are unchanged during the scattering, the total cross section \( \sigma_{\text{H}_2^+}(nlm) \) of \( \text{H}_2^+[nlm](e,2e) \) might be combined from the corresponding partial cross sections for both values of \( S \) with the respective weight factors.

Finally, reminding the Eq. (6) we arrive to the expression of the \( \text{H}_2^+ \) impact ionization-excitation cross section via the sum of the cross sections \( \sigma_{\text{H}_2^+}(n_1l_1m_2) \) of the impact ionization of the \( \text{H}_2^+ \) in the initial \( \varphi_{nlm} \) states:

\[
\sigma_{n_2l_2m_2} = \sum_{n_1l_1} |c_{n_1l_1n_2l_2m_2}|^2 \sigma_{\text{H}_2^+}(n_1l_1m_2).
\] (9)

In what follows, we will refer to this approach as PSECS-SW (SW for scattered wave). In this approximation the inter-electron correlations are taken into account only in the \( \text{H}_2 \) initial state, while neglecting the interaction between the molecular electrons during the impact. It means, that we consider only the kick-off process, while the final-state scattering and the sequential mechanism are omitted. On the other hand, the distortion of the incoming wave due to the interaction with the nuclei and the post-collisional interaction of the ejected electron with the scattered electron is taken into account.

The comparison of the results of PSECS-1B, PSECS-2B and PSECS-SW on the ionization-excitation of \( \text{H}_2 \) to the \( 2p\sigma_u^+ \) level of \( \text{H}_2^+ \) with the experimental data should reveal the dominant effects in each experimental situation.

### III. RESULTS

In this part we will compare our results to the experimental ones published in \([7]\). Some supplementary experimental results of the same authors are also available on the site of the Many Particle Spectroscopy Conference held in Berlin in 2012 \([18]\). Perpendicular and parallel alignments of the internuclear axis to the incidence direction and to the momentum
transfer directions are considered. In all these cases the variation of the MDCS of (e,2e) simple ionization and of H$_2$ with formation of the residual H$_2^+$ in $2s\sigma_g$, $2p\pi_u$, $2p\sigma_u$ states are given in terms of the scattering angle $\theta_s$.

Now, the most common feature of all (e,2e) simple ionization experiments is that the binary peek, which shows the preferential ejection direction, is oriented around that of the momentum transfer. In the experimental conditions here, this region is found around $35^\circ \leq \theta_e \leq 65^\circ$ measured with respect to the incidence direction (all electrons are detected in the incidence plane, the $\theta_s$ and $\theta_e$ are measured with respect to the direction of $k_i$). Following [7], we have thus integrated the MDCS over the ejection angle $\theta_e$ of the slow electron.

$$
\frac{d^3\sigma}{dE_e d\phi_e d\Omega_s} = \int_{\theta_a}^{\theta_b} \frac{d^3\sigma}{dE_e d\Omega_e d\Omega_s} d\theta_e.
$$

All our PSECS-1B/2B calculations were performed for the total electronic energy value $E = 23.3$ eV which represents the sum of the energy values of the ejected electron and the binding energy of the residual ion $E = E_e + E_{ion}$. The probability amplitudes of the ionization-excitation with the formation of the residual ion in the $2s\sigma_g$, $2p\pi_u$ and $2p\sigma_u$ states were extracted from a single wave function $\psi^+(r_1, r_2)$. Due to the fixed total energy $E$, in our calculations the ejected electron energies $E_e = E - E_{ion}$ were different for the different final ion states: 40 eV (that correspond to its value in the experiment [7, 18]) in the case of $2p\sigma_u$ final ion state, 35.8 eV in the case of $2p\pi_u$ and 34 eV in the case of $2s\sigma_g$ state. Since the difference between the ionization energy values of H$_2$ to the $2s\sigma_g$ and $2p\pi_u$ level of H$_2^+$ is smaller than the energy resolution of the experiment [7, 18], we have summed the corresponding MDCS for these two levels. So, the average ejected electron energy was appeared to be equal to 35 eV for the $2s\sigma_g + 2p\pi_u$ case.

On figures 1 and 2 we compare our results for ionization-excitation H$_2$ with formation the residual H$_2^+$ in $2s\sigma_g$ and $2p\pi_u$ states to the experimental ones for the above mentioned orientations of the molecule. In spite of the fact, that the simple plane wave description of the incident and scattered electrons is used in PSECS-1B, the results show the same structure as the experimental points. We observe also, that the introduction of the second Born terms in PSECS-2B does not bring any change in the structure of the graphs. This can be explained by the fact that kick-off and final-state scattering are more important than the sequential mechanism in these situations. One can also observe on figure 2b, which corresponds to the
situation where the molecule is aligned with the direction of the momentum transfer, the absence of the maximum around $\theta_s = 25^\circ$, in contrast to the four preceding cases shown on Figures 1 and 2.

We next consider the ionization-excitation of $\text{H}_2$ with formation of the residual $\text{H}_2^+$ in $2p\sigma_u$ level. On Figures 3 and 4 the variations of the MDCS for $2p\sigma_u$-case for the different orientations described above are given. Here the results concerning the case of perpendicular alignment to the incident plane designated by $\mathbf{R} \parallel Oy$ are not presented as they are exactly zero in all theoretical methods (due to symmetry of final state) and are negligibly small in the experimental results.

The main observation that we can make from these figures is that the results obtained by PSECS-1B and PSECS-2B do not reproduce the structures of the experimental points. To go beyond plane wave description for the incident and scattered waves we apply the PSECS-SW approach described above. As mentioned there, this approach takes into account the distortion undergone by the incident and the scattered waves due to their interaction with the nuclei and due to the post-collisional interaction with the ejected electron. It has meanwhile the disadvantage of being unable to describe the final-state scattering and the sequential mechanism. The dominating term in the projection in equation (5) of the initial state on $2p\sigma_u$ orbital is of the same $2p\sigma_u$ type, such that

$$\langle \varphi_{2p\sigma_u} | \Phi_0(r_1, r_2) \rangle \approx -0.0874 \varphi_{2p\sigma_u}(r_1).$$

So the main idea here is to calculate the MDCS of the electron impact ionization of $\text{H}_2^+$ (in the $\text{H}_2$ equilibrium internuclear distance $R = 1.4$ a.u.) for the $2p\sigma_u$ level, and multiply it by the coefficient $|c_{2p\sigma_u, 2p\sigma_u}|^2 = 0.00764$. Since the ionization energy of $\text{H}_2^+ (2p\sigma_u)$ is less than that of the ionization-excitation energy of $\text{H}_2$, we lowered the impact energy to $E_i = 160$ eV to keep the same energy values of the ejected and scattered electrons in the ionization of $\text{H}_2$ ($E_e = 40$ eV and $E_s = 100$ eV). We should mention here, that the SW method was not applied in the preceding cases for $2s\sigma_g$ and $2p\pi_u$, because of the necessity for a large number of terms in the sum on right-hand-side of Eq.(9) in the case of the final ion state $2s\sigma_g$.

The PSECS-SW results are shown on Figures 3 and 4. The corresponding PSECS-SW curves have the same order of magnitude, but different structures compared to those of the PSECS-1B and 2B curves. The dependence of PSECS-SW results on $\theta_s$ seems to be closer
FIG. 1: (Color online) The variation of the MDCS of \((e,2e)\) simple ionization of \(H_2\) with formation the residual \(H^+_2\) in \(2s\sigma_g\) and \(2p\pi_u\) states, integrated over the ejection angle of slow electron \(\theta_e\) between \(35^\circ \leq \theta_e \leq 65^\circ\), in terms of the scattering angle \(\theta_s\). The energy of the incident electron \(E_i = 179\text{eV}\), and that of the ejected electron \(E_e = 35\text{ eV}\). Results of PSECS-2B are presented by the solid line, those of PSECS-1B by the dashed line and the experimental data \cite{7} by dots. The orientation of the internuclear axis: a) \(\mathbf{R} \parallel Ox\), b) \(\mathbf{R} \parallel Oy\), c) \(\mathbf{R} \parallel Oz\). Oz \parallel \mathbf{k}_i\).
FIG. 2: (Color online) The same as on Fig. 1 but for $Oz \parallel K$. In a) $R \parallel Ox$; b) $R \parallel Oz$. The case $R \parallel Oy$ is already presented in the Fig. 1(b).

to the experimental.

In what follows we will try to discuss the physical reasons of the large discrepancy that exists between the PSECS-1B/2B and the PSECS-SW results in the case of $2p\sigma_u$. It is easy to see, that the most significant difference of PSECS-1B/2B from the experimental data in both cases $2s\sigma_g + 2p\pi_u$ and $2p\sigma_u$ is observed for $R \parallel K$ (Figures 2b and 4b), while for $R \perp K$ (Figures 2a and 4a) the results of PSECS-1B/2B are close to the experimental ones. In the case of $2p\sigma_u$, the large difference from the experimental results is also observed for the orientations parallel and perpendicular to the incidence direction $R \parallel k_i$ and $R \perp k_i$. This difference is smaller than that of the case $R \parallel K$ and higher than that of $R \perp K$. So, the angular domain of the molecular orientation near that of the momentum transfer, where PSECS-1B/2B are failing, is much wider for $2p\sigma_u$ than for $2s\sigma_g + 2p\pi_u$.

This observations can be explained using the known two-center interference model [19, 20].
FIG. 3: (Color online) The same as on Fig. 1 but with formation of residual ion in the 2pσ_u level. Here the ejection energy $E_e = 40$ eV. Results of PSECS-2B (solid line), PSECS-1B (dashed line), PSECS-SW (dotted line), experimental data [7] (dots). a) $\mathbf{R} \parallel Ox$; b) $\mathbf{R} \parallel Oz$. The experimental results concerning the case $\mathbf{R} \parallel Oy$ are not shown here since they are relatively small with respect to the other cases.

of the impact ionization of H_2. Although this model in its simplest form is unable to give correct quantitative fitting of TDCS [9], it is useful for qualitative analysis of its structure [21]. Let us consider for the case of impact ionization-excitation of H_2 with formation of H_2^+ in 2pσ_u state, the first Born matrix element

$$f_{1B} = -\frac{2}{K^2} \langle k_e \ 2p\sigma_u | \exp(iK \cdot r_1) + \exp(iK \cdot r_2) | 0 \rangle.$$  \hspace{1cm} (11)

Here $|0\rangle \equiv \Phi_0(r_1, r_2)$ represents the ground state of H_2 and

$$|k_e \ 2p\sigma_u\rangle = \frac{1}{\sqrt{2}} [\varphi_{k_e}(r_1)\varphi_{2p\sigma_u}(r_2) + \varphi_{2p\sigma_u}(r_1)\varphi_{k_e}(r_2)].$$  \hspace{1cm} (12)
represents the final state, where \( \varphi_{2p\sigma_u}(r) \) is a wave function of \( 2p\sigma_u \) of \( \text{H}_2^+ \) ion, and \( \varphi_{k_e}(r) \) is a wave function of the continuum state of \( \text{H}_2^+ \) ion. From the expansion (1) for \( \Phi_0 \) the projection \( \langle k_e|0 \rangle = 0 \), because \( \varphi_{k_e}(r) \) is orthogonal to all bound states of \( \text{H}_2^+ \), and \( \langle 2p\sigma_u|0 \rangle \approx c_{2p\sigma_u,2p\sigma_u}\varphi_{2p\sigma_u}(r) \) as mentioned earlier. So we can write

\[
f_{1B} \simeq -\frac{2^{3/2}}{K^2} c_{2p\sigma_u,2p\sigma_u} \langle k_e| \exp(iK \cdot r)|2p\sigma_u \rangle.
\] (13)

If we assume, that the two centers of the molecule are so far, that \( \text{H}_2^+ \) state can be presented by the combination of two single-center atomic functions

\[
\varphi_{2p\sigma_u}(r) \simeq \frac{1}{\sqrt{2}} [\varphi_{1s}(r - R/2) - \varphi_{1s}(r + R/2)],
\]

and use a plane wave description for the final state \( \varphi_{k_e}(r) \simeq (2\pi)^{-3/2} \exp(ik_e \cdot r) \), we can write

\[
f_{1B} \simeq 2c_{2p\sigma_u,2p\sigma_u} f_{1B1s} \sin[(K - k_e) \cdot R/2].
\] (14)
Here

\[ f_{1B1s} = -\frac{2}{K^2} \langle \mathbf{k}_e | \exp(i\mathbf{K} \cdot \mathbf{r}) | 1s \rangle. \]  

(15)

is the first-Born amplitude of the impact ionization of a single hydrogen atom in the ground state. Eq. (14) contains an interference factor resulting from the two-center nature of the target. Note that here the interference factor is \( \sin[(\mathbf{K} - \mathbf{k}_e) \cdot \mathbf{R}/2] \), and not \( \cos[(\mathbf{K} - \mathbf{k}_e) \cdot \mathbf{R}/2] \) as in \([20]\), because we consider the ungerade final state. The interference peak of the cross section in the case \( \mathbf{R} \parallel \mathbf{K} \) is obtained for \( KR = \pi \), while for \( \mathbf{R} \perp \mathbf{K} \) the interference factor does not depend on \( K \) and thus not on \( \theta_s \). The positions of the maxima of the PSECS-1B and PSECS-2B curves for \( \mathbf{R} \parallel \mathbf{K} \) on Fig. 4(b) confirms this result. For PSECS-SW the interference peak is observed for lower values of the scattering angle.

Hence, the large discrepancy of PSECS-1B/2B results from the experimental ones for the molecular orientation \( \mathbf{R} \parallel \mathbf{K} \) in the \( 2p\sigma_u \) case appears to be a consequence of the strong sensibility of the positions of the interference maxima to the deviation of the description of the incident/scattered electron from the plane wave. This is caused by the interaction of the electron with the molecule before and after the collision. In \( 2s\sigma_g \) and \( 2p\pi_u \) states, the overlap between single-center functions is larger than that in \( 2p\sigma_u \), and the two-center interference is less evident. This can be the reason why the plane wave approximation for the incident/scattered electron gives better results for these levels.

IV. CONCLUSION

We have calculated the multifold differential cross section of electron impact dissociative ionization of \( \text{H}_2 \) by three different variants of the exterior complex scaling procedure in prolate spheroidal coordinates (PSECS). Our results are compared to those of a recent experiment that detects in coincidence, for the first time, the emerging electrons and one of the protons. In the first variant, designated PSECS-1B, the two target electrons are treated \textit{ab initio} while the interaction of the incident-scattered electron is taken into account using the first term of the Born series. In the second one, PSECS-2BCD, the second Born term is introduced in the dipole approximation. In the third approach, designated PSECS-SW, applied to the ionization-excitation to the \( 2p\sigma_u \) level of \( \text{H}^+_2 \), the multi-configurational single active electron approximation is used for the target, while the interaction of the incident
electron with target is described ab initio. Our results obtained by the PSECS-1B and 2B agree quite well with experimental results concerning the ionization excitation to the $2s\sigma_g$ and $2p\pi_u$ levels of $H_2^+$ and do not agree with those concerning the $2p\sigma_u$ level. This can be explained by the fact, that the the scattering angle values corresponding to the maxima are very sensitive to the interaction of the incident electron with the molecule, in situations where the maxima is caused by the two-center interference. The PSECS-SW approach reproduces quite well the structure of the experimental curve in this case.

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