

The Double Ionization of H_2 by Fast Electron Impact: Influence of the Final State Electron–Electron Correlation*

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Abstract—We have determined fully differential cross sections of the $(e, 3e)$ double ionization of H_2 by employing correlated initial- and final-state wave functions. We have constructed for the description of the two slow ejected electrons a symmetrized product of a correlation function and two-center continuum wave functions, which fulfill the correct boundary conditions asymptotically up to the order $O((kr)^{-2})$. We have shown that the introduction of the correlated part of the final-state wave function improves the results on the $(e, 3-1e)$ of H_2 .

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1. INTRODUCTION

The double ionization of atoms and molecules by photons or electrons is of considerable interest in many branches of physics. Such processes are also important for the understanding of electronic structure, the ionization mechanisms, and probing electron–electron correlation in the case of double ionization, which is the main cause of this process [1].

The coincidence detection techniques, measuring the momenta of fragments emerging from atomic collision processes and especially from dissociative ionization experiments of diatomic species by electron impact, are now undergoing a very rapid development [2–5]. This type of coincidence detections have been already performed in collision experiments involving the multiply charged ionic projectiles and the hydrogen molecule [6–9].

The electronic two-center problem, met in the study of inelastic collisions of diatomic targets, is very closely related to the Coulomb three-body problem that one meets in simple ionization experiments by electron impact [10]. In the last few years, many elegant models have been proposed to describe the dynamics of the ejected electron in an $(e, 2e)$ experiment, which presents a unique physical situation together with that of (γ, e) , where an electron can

be found in the field of two fixed nuclei. Joulakian et al. [11] used an approximate 3C-type one parameter wave function inspired by the Pluvillage model [12] constructed by the product of two Coulomb functions. Serov et al. [13] compared a wave packet evolution approach to that using partial waves constructed by the exact solutions in prolate spheroidal coordinates of the two-center Coulomb problem [14].

In the present paper, we study theoretically the double ionization of H_2 by electron impact. We describe the double electronic two-center continuum by a product of two modified two-center Coulomb continuum [15] wave functions. This is particularly interesting, as the double continuum is constituted of two equivalent electrons coming from the electron cloud of the same target, in contrast to $(e, 2e)$ experiments, where only one of the electrons comes from the target. This permits the verification of different theoretical models proposed to describe the electronic continuum and understand the mechanisms of the double ionization. Up to now, $(e, 3e)$ experiments have only been performed on atoms [16]. Recently, $(e, 3-1e)$ experiments on molecular hydrogen at about 600 eV incident energy have been reported [17]. These experiments are double ionization experiments, where only one of the ejected electrons is detected in coincidence with the scattered electron.

2. THEORY

The multiply differential cross section (MDCS) of a general out-of-plane detection of the scattered electron and the two ejected electrons from a diatomic molecule is sixfold, and is given by

$$\sigma^{(6)} = \frac{d^6\sigma}{d\Omega_\rho d\Omega_s d\Omega_1 d\Omega_2 d(k_1^2/2) d(k_2^2/2)} \quad (1)$$

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$$= \frac{k_s k_1 k_2}{k_i} |T_{fi}|^2,$$

where $d\Omega_s$, $d\Omega_1$, $d\Omega_2$, and $d\Omega_\rho$ are, respectively, the elements of the solid angles for the orientations of the scattered and the ejected electrons and the internuclear axis $\boldsymbol{\rho}$. k_i , k_s , k_1 , and k_2 represent, respectively, the moduli of the wave vectors of the incident, scattered, and ejected electrons. In the case of randomly oriented targets, we must pass to the fivefold differential cross section (FDSC) by integrating over all possible and equally probable directions of the molecule in space

$$\sigma^{(5)} = \frac{1}{4\pi} \int d\Omega_\rho \sigma^{(6)}(\boldsymbol{\rho}). \quad (2)$$

The conservation of the energy for fixed internuclear distance ρ gives

$$E_i = E_s + E_1 + E_2 + I, \quad (3)$$

where E_i , E_s , E_1 , and E_2 represent, respectively, the energies of the incident, scattered, and ejected electrons; $I = 51$ eV being the energy necessary to eject two electrons from the target at the equilibrium internuclear distance $\rho = 1.4$ a.u. We define the transition matrix element by the first term of the Born series

$$T_{fi} = \frac{1}{2\pi} \int d\mathbf{r}_0 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \exp(i\mathbf{K}\mathbf{r}_0) \quad (4)$$

$$\times \bar{\chi}_f(\mathbf{r}_1, \mathbf{r}_2) V \varphi_i(\mathbf{r}_1, \mathbf{r}_2).$$

Here, the overline indicates the complex conjugate. \mathbf{r}_0 is the position of the fast incident scattered electron which we will describe as a plane wave. \mathbf{r}_j ($j = 1, 2$) refer to the positions of the bound (ejected) electrons. $\mathbf{K} = \mathbf{k}_i - \mathbf{k}_s$ is the momentum transferred to the target and V represents the Coulomb interaction between the incident electron and the target given by

$$V = -\frac{1}{|\mathbf{r}_0 - \boldsymbol{\rho}/2|} - \frac{1}{|\mathbf{r}_0 + \boldsymbol{\rho}/2|} \quad (5)$$

$$+ \frac{1}{|\mathbf{r}_0 - \mathbf{r}_1|} + \frac{1}{|\mathbf{r}_0 - \mathbf{r}_2|}.$$

Integrating over the position of the fast incident electron \mathbf{r}_0 using the Bethe transformation, we have

$$T_{fi} = \frac{2}{K^2} \int d\mathbf{r}_1 \int d\mathbf{r}_2 \exp(i\mathbf{K}\mathbf{r}_0) \bar{\chi}_f(\mathbf{r}_1, \mathbf{r}_2) \quad (6)$$

$$\times \left(-2 \cos(\mathbf{K}\boldsymbol{\rho}/2) + \exp(i\mathbf{K}\mathbf{r}_1) \right.$$

$$\left. + \exp(i\mathbf{K}\mathbf{r}_2) \right) \varphi_i(\mathbf{r}_1, \mathbf{r}_2).$$

The final-state wave function

$$\chi_f(\mathbf{r}_1, \mathbf{r}_2) \quad (7)$$

$$= \frac{\phi_f(\mathbf{k}_1, \mathbf{r}_1, \mathbf{k}_2, \mathbf{r}_2) + \phi_f(\mathbf{k}_1, \mathbf{r}_2, \mathbf{k}_2, \mathbf{r}_1)}{\sqrt{2}},$$

describes the state of the two equivalent ejected electrons, where

$$\phi_f(\mathbf{k}_1, \mathbf{r}_1, \mathbf{k}_2, \mathbf{r}_2) \quad (8)$$

$$= v(k_{12}) {}_1F_1(i\alpha_{12}, 1, -i(k_{12}r_{12} + \mathbf{k}_{12}\mathbf{r}_{12}))$$

$$\times T(\mathbf{k}_1, \mathbf{r}_1) T(\mathbf{k}_2, \mathbf{r}_2),$$

in which we have introduced, as in the case of atoms [18], the electron–electron correlation. Here $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2$ and

$$v(k_{12}) = \exp\left(-\frac{\pi\alpha_{12}}{2}\right) \Gamma(1 - i\alpha_{12}) \quad (9)$$

represents the Gamow factor with

$$\alpha_{12} = \frac{1}{2k_{12}}, \quad \mathbf{k}_{12} = \frac{1}{2}(\mathbf{k}_1 - \mathbf{k}_2). \quad (10)$$

The final-state wave function satisfies the orthonormality condition in the sense

$$\langle \phi_f(\mathbf{k}_1, \mathbf{r}_1, \mathbf{k}_2, \mathbf{r}_2) | \phi_f(\mathbf{k}'_1, \mathbf{r}_1, \mathbf{k}'_2, \mathbf{r}_2) \rangle \quad (11)$$

$$= \delta(\mathbf{k}_1 - \mathbf{k}'_1) \delta(\mathbf{k}_2 - \mathbf{k}'_2).$$

The two-center continuum (TCC) wave function

$$T(\mathbf{k}_i, \mathbf{r}_j) = \exp(-\pi\alpha_i) (\Gamma(1 - i\alpha_i))^2 \quad (12)$$

$$\times \frac{\exp(i\mathbf{k}_i\mathbf{r}_j)}{(2\pi)^{3/2}} {}_1F_1(i\alpha_i, 1, -i(k_i r_{ja} + \mathbf{k}_i\mathbf{r}_{ja}))$$

$$\times {}_1F_1(i\alpha_i, 1, -i(k_i r_{jb} + \mathbf{k}_i\mathbf{r}_{jb}))$$

is borrowed from [19]. It describes the ejected electron in the field of two Coulomb centers with

$$\alpha_i = -\frac{Z_i}{k_i}, \quad \mathbf{r}_{ja} = \mathbf{r}_j + \boldsymbol{\rho}/2, \quad (13)$$

$$\mathbf{r}_{jb} = \mathbf{r}_j - \boldsymbol{\rho}/2, \quad i, j = 1, 2,$$

and $Z_i = 1$. Finally, $\varphi_i(\mathbf{r}_1, \mathbf{r}_2)$ represents the space part of the initial-state wave function.

Taking into account the symmetry of the final and initial functions $\varphi_i(\mathbf{r}_1, \mathbf{r}_2) = \varphi_i(\mathbf{r}_2, \mathbf{r}_1)$ with respect to exchange of \mathbf{r}_1 and \mathbf{r}_2 , we can reduce the expression of the transition matrix element to the following six-dimensional integral:

$$T_{fi} = \frac{2\sqrt{2}}{K^2} \int d\mathbf{r}_1 \int d\mathbf{r}_2 \bar{\phi}_f(\mathbf{r}_1, \mathbf{r}_2) \quad (14)$$

$$\times \left(-2 \cos(\mathbf{K}\boldsymbol{\rho}/2) + \exp(i\mathbf{K}\mathbf{r}_1) \right.$$

$$\left. + \exp(i\mathbf{K}\mathbf{r}_2) \right) \varphi_i(\mathbf{r}_1, \mathbf{r}_2).$$

The space coordinates of the wave functions are defined in the molecular frame of reference, whose origin is fixed on the center of mass of the molecule and whose z -axis is parallel to the internuclear vector $\boldsymbol{\rho}$ of constant modulus.

We consider the wave function $\varphi_i(\mathbf{r}_1, \mathbf{r}_2)$ for the initial electronic Σ_g fundamental state of the target

The values of the variational parameters

C_1	0.436611001245	C_2	-0.720456518414	C_3	0.195567664329
α_1	0.781942647364	α_5	0.332387136618	α_7	0.837471876213
α_2	0.449576921040	α_6	0.905250662680	α_8	0.380442714559
α_3	0.110605279800				
α_4	1.267607929845				
γ_1	0.015459390969	γ_2	0.259398892559	γ_3	0.227120883164

H₂ obtained by a variational fourteen-parameter calculation

$$\varphi_i(\mathbf{r}_1, \mathbf{r}_2) = C_1\psi_1 + C_2\psi_2 + C_3\psi_3, \quad (15)$$

with

$$\begin{aligned} \psi_1 &= (1 + P_{12})(1 + P_{ab}) \quad (16) \\ &\times \exp(-\alpha_1 r_{1a} - \alpha_2 r_{1b} - \alpha_3 r_{2a} - \alpha_4 r_{2b} - \gamma_1 r_{12}), \\ \psi_2 &= (1 + P_{12}) \\ &\times \exp(-\alpha_5 r_{1a} - \alpha_6 r_{1b} - \alpha_6 r_{2a} - \alpha_5 r_{2b} - \gamma_2 r_{12}), \\ \psi_3 &= (1 + P_{12}) \\ &\times \exp(-\alpha_7 r_{1a} - \alpha_7 r_{1b} - \alpha_8 r_{2a} - \alpha_8 r_{2b} - \gamma_3 r_{12}). \end{aligned}$$

Here, P_{12} and P_{ab} are the operators which interchange electrons ($1 \longleftrightarrow 2$) and the two nuclei ($a \longleftrightarrow b$), respectively, and α_i , γ_j are the positive variational parameters. The matrix elements \mathbf{A} and \mathbf{B} of the generalized eigenvalue problem with fixed parameters α_i , γ_j

$$\mathbf{A}\mathbf{C} = \mathbf{E}\mathbf{B}\mathbf{C}, \quad \mathbf{C} = (C_1, C_2, C_3)^T, \quad (17)$$

consist of six-dimensional integrals. They are reduced to the three-dimensional integrals by means of the Fourier transform. The ground-state energy E is minimized by the variation of the parameters α_i , γ_j , using the comprehensive modified Newton algorithm (see the table). This gives a ground-state energy of $-1.174\,136$ a.u., while the upper bound of the ground-state energy is equal to $-1.174\,475\,663$ a.u. (used 1920 Hylleraas–CI functions [20]).

The required transition matrix element (14) will be obtained by six-dimensional integrals.

As a first step, we neglect the correlation in the final-state wave function $\phi_f(\mathbf{r}_1, \mathbf{r}_2)$ represented by the confluent hypergeometric function ${}_1F_1(i\alpha_{12}, 1, -i(k_{12}r_{12} + \mathbf{k}_{12}\mathbf{r}_{12}))$, and choose the discrete wave function $\varphi_i(\mathbf{r}_1, \mathbf{r}_2)$ by a variational three-parameter calculation borrowed from [21]

$$\varphi_i(\mathbf{r}_1, \mathbf{r}_2) = N(\rho)(\phi(1)\psi(2) + \psi(1)\phi(2)), \quad (18)$$

with

$$\phi(j) = x_a(j) + \epsilon x_b(j), \quad (19)$$

$$\begin{aligned} \psi(j) &= \epsilon x_a(j) + x_b(j), \\ x_a(j) &= \exp(-\beta\xi_j - \gamma\eta_j), \\ x_b(j) &= \exp(-\beta\xi_j + \gamma\eta_j), \end{aligned}$$

where $\xi_j = (r_{ja} + r_{jb})/\rho$, $\eta_j = (r_{ja} - r_{jb})/\rho$; $\beta = 0.835$, $\gamma = 0.775$, and $\epsilon = 0.137$ are variational parameters. Here, the norm $N = 0.255$ and the ground-state energy is equal to -1.149 a.u.

In this case, after using the Fourier transform for one of the centers for each electron, the transition matrix element (14) will be given by the products of two three-dimensional integrals. Here, the integrand functions consist of the defined elementary functions.

3. RESULTS

We have chosen in our calculations the same experimental conditions, where the scattered electron of 500 eV is detected at an angle $\theta_s = 1.5^\circ$ with respect to the incident electron beam. The ejected electrons of much lower energy values are detected in coincidence with the scattered electron. The energy of the incident electron is chosen by the energy conservation equation (3). In Fig. 1, we show the variation of the FDCS of the ionization of H₂ in terms of the ejection angle. In Fig. 1a we show the results obtained by the wave function including the correlation part and in Fig. 1b the results without the correlation part. We observe that the introduction of the correlations shows some structures which are also observed in the case of double photoionization [22].

Once we have done, the necessary observations concerning the behavior of the multiply differential cross sections (MDCS) with our two-center approach, we have tried to reproduce the results of the $(e, 3-1e)$ experiments realized by the Orsay group [17]. $(e, 3-1e)$ designates double ionization experiments in which the scattered and only one of the ejected electrons are detected in coincidence. The energy of the non-observed electron is deduced from the energy conservation equation (3). This presents a practical advantage for the experimental realization, as it needs the coincidence detection of two electrons

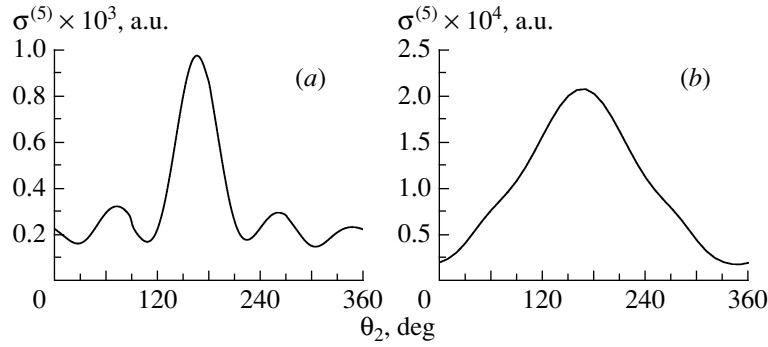


Fig. 1. The variation of the fivefold differential cross section for H_2 with the ejection direction. The direction of the second electron is taken parallel to the momentum transfer \mathbf{K} at $E_i = 612$ eV, $E_s = 500$ eV, $E_1 = 10$ eV, $E_2 = 51$ eV, and $\theta_s = 1.5^\circ$. (a) With correlation. (b) Without correlation.

instead of three and thus larger MDCS. Theoretically this can be determined by integrating our results over the solid angles Ω_2 of the second ejected electron:

$$\sigma^{(4)} = \int d\Omega_2 \sigma^{(5)}. \quad (20)$$

The energy conservation insures that the coincidences observed correspond well to a double ionization event. In Fig. 2 we compare our results obtained by the full correlated function given by the solid curve to those obtained by the application of the uncorrelated TCC product [19] (dotted curve), and by the application of simplified second Born method [23]

(dash-dotted curve). We have scaled these theoretical results to our result at the maximum around 345° . The experimental results [17], on the other hand, were scaled to our results around 300° . We observe that the theoretical approaches reproduce curves, which are symmetrical around 345° , the direction of the momentum transfer $\mathbf{K} = \mathbf{k}_i - \mathbf{k}_s$, which is an axis of symmetry in the first Born theory. The experimental peak is found around 320° , and for the moment, neither theory can reproduce this result. We observe nevertheless that our correlated results in continuous black line show an improvement, when compared specially with the one corresponding to the uncorrelated TCC (dotted curve). It passes very near the experimental values in the regions found between 250° and 300° and in the region between 90° and 120° .

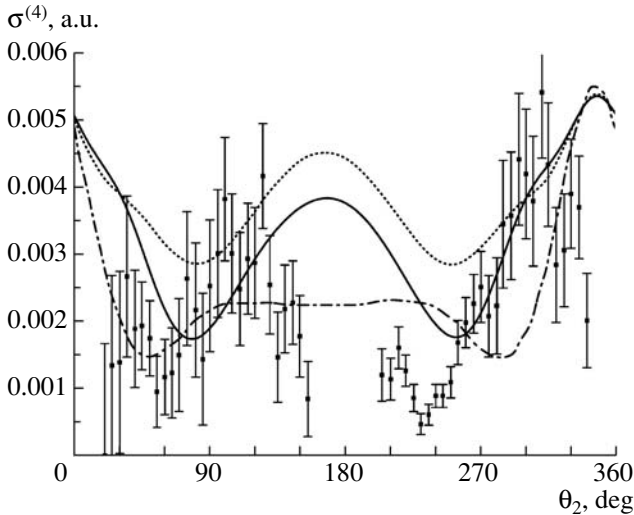


Fig. 2. The variation of the fourfold differential cross section of the $(e, 3-1e)$ ionization H_2 for $E_i = 612$ eV, $E_s = 500$ eV, $E_1 = 10$ eV, $E_2 = 51$ eV, $\theta_s = 1.5^\circ$. Present work, solid curve. Full squares with error bars correspond to experimental results of [17] scaled to the theory for the best visual fit at the binary lobe (around 300°). Dotted [19] and dash-dotted [23] curves are scaled to the solid curve at 345° .

4. CONCLUSIONS

We have determined the fully differential cross section of the double ionization of H_2 by applying a product of two modified two-center continuum functions. Our results show that, as expected, the two ejected electrons tend to emerge in opposite directions. The comparison with the results of $(e, 3-1e)$ experiments show that the use of the first Born term in the perturbation gives the good structure, but it cannot give the optimal direction of the experiment. Here, the difficulty of describing the two-center continuum is added to the main difficulty of describing the double electronic continuum and to the difficulty of describing second-order effects. This demands a larger-scale computational effort that we intend to realize next.

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