

Partial-wave formulation in the calculation of atomic and molecular ionization cross sections

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(Received 26 May 1992; revised manuscript received 20 July 1992)

A formulation for calculating the distorted-wave Born approximation cross sections for both atomic and molecular ionizations using the partial-wave-expansion method is introduced. The ionization cross sections of molecular and atomic hydrogen target with positron impact are calculated in the intermediate collision velocity region with this formulation. The results are compared to the updated experiment data. This comparison has offered an alternative evaluation for the present model of ionization process.

PACS number(s): 34.50.Fa, 34.50.Gb, 34.90.+q

Measurements of the positron-impact ionization of molecular hydrogen and atomic hydrogen have been reported by Raith and co-workers [1,2] in recent years. The theoretical treatments with respect of atomic hydrogen target include the quantum-mechanical calculations of Ghosh, Mazumdar, and Basu [3] and of Mukherjee, Singh, and Mazumdar [4], and classical trajectory Monte Carlo approximation (CTMC) of Ohsaki *et al.* [5] and of Wetmore and Olson [6]. But as for the molecular target, to our knowledge, no published theoretical results exist yet.

The distorted-wave Born approximation (DWBA) is largely used to determine the T matrix in ionization collision within the intermediate collision velocity region, such as in the works of Younger [7], who has treated the electron-impact ionization of hydrogenlike and lithiumlike atoms, and the recent works of Basu, Mazumdar, Ghosh [8] and Campeanu, McEachran, and Stauffer [9], who has treated the positron-impact ionization of helium, and of Ghosh, Mazumdar, and Basu [3] and of Mukherjee, Singh, and Mazumdar [4], who have treated the positron-impact ionization of atomic hydrogen.

To calculate the total cross section using DWBA, the partial-wave-expansion method seems to be effective as all the angular integral can be worked out without any difficulty and the dimension of the integration is reduced. But the problem is more difficult when the target is a molecule, as another integration over the orientation of the molecule has to be made, and the partial-wave decomposition of the molecule wave functions seems rather difficult.

Nevertheless, we have found that these difficulties can be handled smoothly while working out the formulation for positron-impact ionization cross section of molecular hydrogen.

To elucidate our partial-wave-expansion method of formulation for the ionization cross sections of molecular target, we first write down the first Born approximation (FBA) or DWBA T -matrix elements:

$$T_{ba} = \langle \Psi_f \Psi_e \Psi_{\text{res}} | V | \Psi_i \Psi_{\text{tar}} \rangle, \quad (1)$$

where Ψ_{tar} and Ψ_{res} correspond to the wave functions of the target particle and the ionized residual particle. Ψ_i, Ψ_f are the wave functions of the projectile particle in the incident and final channels, and Ψ_e is the wave function of the ejected electron. Whether Ψ_i, Ψ_f , and Ψ_e are plane, Coulomb, or other kinds of distorted-wave functions depend on which kind of approximation (FBA, or any kind of DWBA) has been employed.

If we consider a molecular ionization case, we have to deal with the characteristic orientation of the molecule. As in most experiments this orientation is randomly arrayed in the target gas, in calculation of the total cross sections we must average over this randomly arrayed orientation to match experiment data. So the total cross section for molecule ionization should be

$$\sigma(E_i) = \frac{1}{4\pi} \int d\hat{\mathbf{R}} \Delta\sigma(\mathbf{k}_i, \mathbf{R}), \quad (2)$$

where \mathbf{R} denotes the characteristic orientation of the molecule, and $\Delta\sigma(\mathbf{k}_i, \mathbf{R})$ reads

$$\Delta\sigma(\mathbf{k}_i, \mathbf{R}) = \int d\mathbf{k}_f d\mathbf{k}_e d\mathbf{k}_{\text{res}} \delta\left(\frac{1}{2}\mathbf{k}_f^2 + \frac{1}{2}\mathbf{k}_e^2 + \frac{1}{2}\mathbf{k}_{\text{res}}^2 + W - \frac{1}{2}\mathbf{k}_i^2\right) \delta(\mathbf{k}_f + \mathbf{k}_e + \mathbf{k}_{\text{res}} - \mathbf{k}_i) \frac{(2\pi)^4}{k_i} |T_{ba}|^2, \quad (3)$$

where $\mathbf{k}_i, \mathbf{k}_f, \mathbf{k}_e$, and \mathbf{k}_{res} denote the momentum of the incident particle, the outgoing projectile particle, the ejected electron, and the residual ion. W is the ionization potential of the molecule. Two delta functions represent the conservation of total momentum and energy, correspondingly. And T_{ba} is given by Eq. (1). If we ignore the recoil of the residual ion we can work out the trivial integrations and Eq. (3) becomes

$$\Delta\sigma(\mathbf{k}_i, \mathbf{R}) = \int d\hat{\mathbf{k}}_f d\hat{\mathbf{k}}_e dk_e [k_e^2(k_i^2 - k_e^2 - 2W)^{1/2}/k_i] (2\pi)^4 |T_{ba}|^2. \quad (4)$$

In Eq. (3) we have already employed the Born-Oppenheimer approximation which assumes that the target molecule does not rotate or vibrate during the collision. And in sequence the direction of \mathbf{R} is frozen during the collision, which makes it possible to take a coordinate system with its z axis along the direction of \mathbf{R} and equivalently make an average over the direction of \mathbf{k}_i to get the total cross section, i.e., Eq. (2) becomes

$$\sigma(E_i) = \frac{1}{4\pi} \int d\hat{\mathbf{k}}_i \Delta\sigma(\mathbf{k}_i, \mathbf{R}) = 4\pi^3 \int d\hat{\mathbf{k}}_i d\hat{\mathbf{k}}_f d\hat{\mathbf{k}}_e dk_e [k_e^2(k_i^2 - k_e^2 - 2W)^{1/2}/k_i] |T_{ba}|^2. \quad (5)$$

If we introduced the standard partial wave expansions of Ψ_i , Ψ_f , and Ψ_e :

$$\Psi(\mathbf{k}, \mathbf{x}) = \sum_{l,m} P_l(k, x) Y_{l,m}^*(\hat{\mathbf{k}}) Y_{l,m}(\hat{\mathbf{x}}). \quad (6)$$

T_{ba} can be expressed as

$$T_{ba}(k_i, k_e, \hat{\mathbf{k}}_i, \hat{\mathbf{k}}_f, \hat{\mathbf{k}}_e) = \sum_{l_i, l_f, l_e} \sum_{m_i, m_f, m_e} Y_{l_i m_i}(\hat{\mathbf{k}}_i) Y_{l_f m_f}(\hat{\mathbf{k}}_f) Y_{l_e m_e}(\hat{\mathbf{k}}_e) \tilde{T}_{ba}, \quad (7)$$

where l_i and l_f are the projectile particle's orbital angular momentum quantum numbers in the incident and final channels, respectively, l_e is the corresponding quantum number of the ejected electron. \tilde{T}_{ba} is excluded of the angular part of \mathbf{k}_i , \mathbf{k}_f , and \mathbf{k}_e . Performing the angular integrations using the orthogonal relationship of the spherical harmonic functions Eq. (5) becomes

$$\sigma(E_i) = 4\pi^3 \sum_{l_i, l_f, l_e} \sum_{m_i, m_f, m_e} \int_0^{k_{\max}} dk_e [k_e^2(k_i^2 - k_e^2 - 2W)^{1/2}/k_i] |\tilde{T}_{ba}|^2 \quad (8)$$

thus the total cross section can be expressed as

$$\sigma(E_i) = \sum_{l_i, l_f, l_e} \sigma_p(l_i, l_f, l_e), \quad (9)$$

where σ_p denotes the corresponding partial-wave contributions to the total cross section.

This formulation is also valid for the atomic target case as we can take an average over an imaginary orientation of the atom to obtain Eq. (2), the similarly transfer to Eq. (5). Again we return to the summation form given by Eq. (9).

In computation of the total cross sections it is of great importance to see how many angular quantum numbers have to be carried out to obtain an accurate result. We have observed the fact that $\sigma_p(l_i, l_f, l_e)$ converges fairly rapidly with l_e but converges rather slowly with l_i and l_f , so that we should make an extrapolation for l_i and l_f . In order to make the extrapolation reasonable, we arrange $\sigma_p(l_i, l_f, l_e)$ in several patterns of sequence. If we consider only the ground-state-ground-state scattering (initial state of the target and final state of the residual ion are in ground states), due to the conservation of parity and angular momentum, for $l_e = 0$ there is only one sequence, namely, $\sigma_p(n, n, 0)$, where $n = 0, 1, 2, \dots$; $l_e = 1$ yields two patterns of sequence, $\sigma_p(n+1, n, 1)$ and $\sigma_p(n, n+1, 1)$; $l_e = 2$ yields three patterns, $\sigma_p(n+2, n, 2)$, $\sigma_p(n+1, n+1, 2)$, and $\sigma_p(n, n+2, 2)$; etc. For each sequence we found that σ_p shows a smooth function of n , and at large n , σ_p decreases exponentially. So after sufficient values of each sequence have been obtained we can make an accurate extrapolation. This method leads to a convenient scheme to calculate an ionization cross section to higher impact energy, saving a great deal of computer time.

Figure 1 shows the behavior of $\sigma_p(n+1, n, 1)$ varying with n . Here σ_p is the partial-wave contributions to the total cross sections of the e^+ -H₂ ionization process. Three curves correspond to three different impact energies. All curves fall down exponentially at large n . The higher the impact energy, the slower σ_p converges. From this we should see that the cross sections become difficult to calculate at higher impact energy.

Using Eq. (8) and the above-mentioned scheme, we have obtained e^+ -H₂ ionization cross-section data.

In the approximation of the T -matrix elements, we

adopt the DWBA with the same model proposed by Campeanu, McEachran, Stauffer [9], which has proved successful in calculation of e^+ -He ionization cross sections by producing results that agreed well with the experiment data of Ref. [10]. In this model, Ψ_i is taken as a plane wave, Ψ_f and Ψ_e are Coulomb waves satisfying the Schrödinger equations:

$$\left(\frac{1}{2}\hat{p}^2 + V_x\right)\Psi_x = E_x\Psi_x, \quad (10)$$

where $x = f$ or e . For $E_e < E_f$,

$$V_e = -1/r, \quad V_f = 0, \quad (11)$$

but for $E_e > E_f$ it is assumed that the slower-moving positron screens the residual ion so that the ejected electron moves in the field of the double positive charge and in sequence,

$$V_e = -2/r, \quad V_f = 1/r. \quad (12)$$

Some details of the calculation of the e^+ -H₂ ionization cross section have been given in the appendix. Our results compared with the experiment data of Refs. [1,11] are shown in Fig. 2.

Using the same distorted-wave model used in the

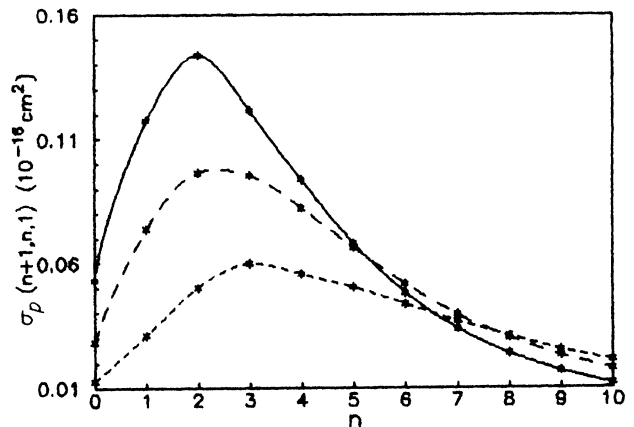


FIG. 1. Partial-wave contributions $\sigma_p(l_i, l_f, l_e)$ of the e^+ -H₂ total ionization cross section, with impact energy of 75 eV (solid line), 100 eV (long-dashed line), and 150 eV (short-dashed line).

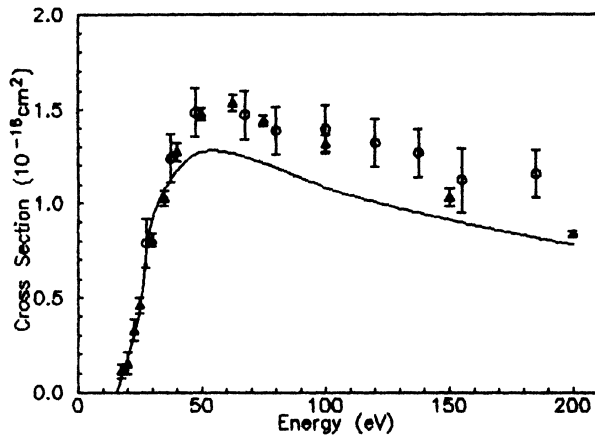


FIG. 2. Positron-impact-ionization cross sections of molecular hydrogen. Present result: solid line; experiment data of Ref. [1]: circles; experiment data of Ref. [11]: triangles.

$e^+ + \text{H}_2$ ionization and the same partial-wave-expansion formulation, we have also calculated the $e^+ + \text{H}$ ionization cross sections. This work has already been done by Mukherjee, Singh, and Mazumdar [4] within impact energy below 70 eV. The only reason that they have not calculated to the higher-energy region is that at higher energy the convergence of σ_p in Eq. (9) with respect to l_i or l_f is rather slow, as shown in Fig. 1, which raises the computation difficulty. Our extrapolation scheme has shown some advantage in this circumstance for saving computer time. So after having generated the results of Ref. [4] as a test of our program, we supplemented the $e^+ + \text{H}$ ionization cross-section data up to the impact energy of 150 eV. Our results are shown in Fig. 3.

Now we have three sets of positron-impact ionization cross sections of atomic helium, atomic hydrogen, and molecular hydrogen calculated by the same model of DWBA. The comparison with experimental data shows that the results of the $e^+ + \text{He}$ and $e^+ + \text{H}_2$ ionization is more satisfying than that of the $e^+ + \text{H}$ case. (The comparison of experimental and theoretical results of $e^+ + \text{He}$ ionization cross sections can be found in Ref. [10].)

Suppose the experimental data are correct (though this is actually dubious). The results may be due to the polarization effect caused by positron impact. When a relatively slow positron approaches the atomic hydrogen target, it attracts the electron cloud around the atomic hydrogen, which will enhance the collision cross sections because the positron heads on an electron cloud with higher density. To create the same enhancement in the two-electron targets of He and H_2 , the positron must pull two electrons, each to the same polarized level as it pulls the one electron in the atomic hydrogen target. But this is impossible, as the two electrons repel each other. So the polarization effect is weakened in two-electron targets. As the present model has not included this effect, this may be an explanation of the fact that the employment of the present DWBA model on the $e^+ + \text{He}$ and $e^+ + \text{H}_2$ ionization is more accurate than the $e^+ + \text{H}$ case. Moreover, the polarization effect may explain the recent discovery of the enhancement of the positron-impact ionization cross sections to the electron-impact ionization cross sections [12].

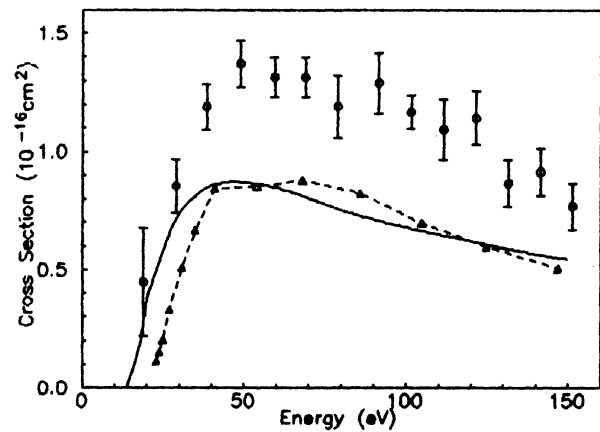


FIG. 3. Positron-impact-ionization cross sections of atomic hydrogen. Present result: solid line; CTMC result of Ref. [6]: dashed line; experiment data of Ref. [1]: circles.

In conclusion, positron-impact ionizations of simple atoms and molecules have offered an ideal testing ground for basic ionization theory. With a partial-wave-expansion formulation established for the calculation of molecular ionization cross sections, we have calculated the total cross sections of positron-impact ionization of molecular hydrogen, and the corresponding data for atomic hydrogen are also produced. The results have given some hints for the weaknesses of the present DWBA model.

The authors would like to thank Professor Yaoyang Liu, Professor Zifang Zhou, and Dr. Jiyun Kuang for motivating conversations. The financial support of the GRAAMD (Chinese Research for Atomic and Molecular Data) is gratefully acknowledged.

APPENDIX

In the calculation of the $e^+ + \text{H}_2$ ionization cross sections, we have adopted the ground-state wave function of H_2 due to Wang [13]:

$$\Psi_{\text{H}_2}(\mathbf{r}_1, \mathbf{r}_2) = c_i [\phi_a(\mathbf{r}_1)\phi_b(\mathbf{r}_2) + \phi_a(\mathbf{r}_2)\phi_b(\mathbf{r}_1)], \quad (\text{A1})$$

where

$$\phi_a(\mathbf{r}) = \lambda^{3/2}/\sqrt{\pi e^{-\lambda|\mathbf{r}-\mathbf{R}|}}, \quad (\text{A2})$$

$$\phi_b(\mathbf{r}) = \lambda^{3/2}/\sqrt{\pi e^{-\lambda|\mathbf{r}+\mathbf{R}|}}, \quad (\text{A3})$$

c_i is a normalization constant, $|\mathbf{R}|$ is half the internuclear separation of H_2 , and λ takes the value of 1.166.

The ionization amplitude is given by

$$T_{ba} = \langle \Psi_f(\mathbf{x}) \Psi_{\text{ion}+e}(\mathbf{r}_1, \mathbf{r}_2) | V_i | \Psi_i(\mathbf{x}) \Psi_{\text{H}_2}(\mathbf{r}_1, \mathbf{r}_2) \rangle, \quad (\text{A4})$$

where

$$V_i = \frac{1}{|\mathbf{x}-\mathbf{R}|} + \frac{1}{|\mathbf{x}+\mathbf{R}|} - \frac{1}{|\mathbf{x}-\mathbf{r}_1|} - \frac{1}{|\mathbf{x}-\mathbf{r}_2|}. \quad (\text{A5})$$

$\Psi_{\text{ion}+e}$ represents the symmetrized final-state wave function of the ion H_2^+ and the ejected electron, orthogonalized to the initial state Ψ_{H_2} :

$$\Psi_{\text{ion}+e} = \Psi'_{\text{ion}+e} - \langle \Psi_{\text{H}_2} | \Psi'_{\text{ion}+e} \rangle \Psi_{\text{H}_2}, \quad (\text{A6})$$

where

$$\Psi'_{\text{ion}+e} = c_f \{ \Psi_{\text{ion}}(\mathbf{r}_1) \Psi_e(\mathbf{r}_2) + \Psi_{\text{ion}}(\mathbf{r}_2) \Psi_e(\mathbf{r}_1) \}. \quad (\text{A7})$$

This modification should make the behavior of the final channel state in the area near nuclei more rational.

After performing the partial-wave analysis using the formula

$$E^{-\lambda|\mathbf{r}_1-\mathbf{r}_2|} = \sum_{l,m} F_l(\lambda r_1, \lambda r_2) Y_{l,m}^*(\hat{\mathbf{r}}_1) Y_{l,m}(\hat{\mathbf{r}}_2) \quad (\text{A8})$$

with

$$F_l(\lambda r_1, r_2) = -4\pi[(r_>/r_<)^{1/2} K_{l+1/2}(\lambda r_>) I_{l+1/2}(\lambda r_<) + (r_</r_>)^{1/2} K_{l+1/2}(\lambda r_>) I'_{l+1/2}(\lambda r_<)] \quad (\text{A9})$$

and

$$e^{-\lambda|\mathbf{r}_1-\mathbf{r}_2|/|\mathbf{r}_1-\mathbf{r}_2|} = \sum_{l,m} \bar{F}_l(\lambda r_1, \lambda r_2) Y_{l,m}^*(\hat{\mathbf{r}}_1) Y_{l,m}(\hat{\mathbf{r}}_2) \quad (\text{A10})$$

with

$$\bar{F}_l(\lambda r_1, \lambda r_2) = 4\pi K_{l+1/2}(\lambda r_>) I_{l+1/2}(\lambda r_<)/(r_1 r_2)^{1/2} \quad (\text{A11})$$

and performing some analytical integrations the \tilde{T}_{ba} in Eq. (8) turns out to be

$$\begin{aligned} \tilde{T}_{ba} = & 2C_i C_f I_0 \left[\delta_{m_e 0} \sum_{l_i} \begin{bmatrix} l_i & l_f & l_1 \\ -m_i & m_f & 0 \end{bmatrix} I_1 I_2 \right. \\ & \left. + \sum_{l_i, l_2} \begin{bmatrix} l_i & l_f & l_1 \\ m_i & -m_f & m_e \end{bmatrix} \begin{bmatrix} l_e & l_1 & l_2 \\ -m_e & m_e & 0 \end{bmatrix} I_3 \right], \quad (\text{A12}) \end{aligned}$$

where

$$\begin{bmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{bmatrix} = \int Y_{l_1 m_1}(\hat{\mathbf{r}}) Y_{l_2 m_2}(\hat{\mathbf{r}}) Y_{l_3 m_3}(\hat{\mathbf{r}}) d\hat{\mathbf{r}}, \quad (\text{A13})$$

$$I_0 = \int \phi_a(\mathbf{r}) \Psi_{\text{ion}}(\mathbf{r}) d^3 \mathbf{r}, \quad (\text{A14})$$

$$I_1 = \int_0^\infty r^2 dr F_{l_e}(\lambda r, \lambda R) P_{l_e}(k_e, r), \quad (\text{A15})$$

$$I_2 = \int_0^\infty x^2 dx P_{l_i}^*(k_i x) Q(x) P_{l_f}(k_f, x), \quad (\text{A16})$$

$$\begin{aligned} I_3 = & \int_0^\infty x_2^2 dx_2 \int_0^\infty x_1^2 dx_1 P_{l_i}^*(k_i, x_1) [2\delta_{l_2}^{\text{even}} F_{l_2}(\lambda x_2, \lambda R)] \\ & \times \frac{4\pi}{2l_1+1} (x_1^{l_1} / x_2^{l_1+1}) P_{l_e}(k_e, x_2) \\ & \times P_{l_f}(k_f, x_1), \quad (\text{A17}) \end{aligned}$$

where P_{l_i} , P_{l_e} , P_{l_f} , and $2\delta_{l_2}^{\text{even}} F_{l_2}(\lambda x_2, \lambda R)$ denote the partial waves of Ψ_i , Ψ_f , Ψ_e , and Ψ_{H_2} correspondingly. $\delta_l^{\text{even}} = 1$ when l is an even number and 0 when l is an odd number. And $Q(x)$ is a lengthy expression containing F_l and \bar{F}_l .

[1] G. Spicher, B. Olsson, W. Raith, G. Sinapius, and W. Sperber, Phys. Rev. Lett. **64**, 1019 (1990).
 [2] D. Fromme, G. Kruse, W. Raith, and G. Sinapius, J. Phys. B **21**, L261 (1988).
 [3] A. S. Ghosh, P. S. Mazumdar, and M. Basu, Can. J. Phys. **63**, 621 (1985).
 [4] K. K. Mukherjee, N. R. Singh, and P. S. Mazumdar, J. Phys. B **22**, 99 (1989).
 [5] A. Ohsaki, T. Watanabe, K. Nakanishi, and K. Iguchi, Phys. Rev. A **32**, 2640 (1985).
 [6] A. E. Wetmore and R. E. Olson, Phys. Rev. A **34**, 2822 (1986).

[7] S. M. Younger, Phys. Rev. A **22**, 111 (1980).
 [8] M. Basu, P. S. Mazumdar, and A. S. Ghosh, J. Phys. B **18**, 369 (1985).
 [9] R. I. Campeanu, R. P. McEachran, and A. D. Stauffer, J. Phys. B **20**, 1635 (1987).
 [10] D. Fromme, G. Kruse, W. Raith, and G. Sinapius, Phys. Rev. Lett. **57**, 3031 (1986).
 [11] H. Knudsen, L. B. Nielsen, M. Charlton, and M. R. Poulsen, J. Phys. B **23**, 3955 (1990).
 [12] D. R. Schultz, R. E. Olson, and C. O. Reinhold, J. Phys. B **24**, 521 (1991).
 [13] S. C. Wang, Phys. Rev. **31**, 579 (1928).