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Positron impact ionization of molecular nitrogen

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Abstract

We have carried out distorted wave calculations of positron ionization of molecular nitrogen in order to compare with recent experimental measurements. In this work, the nitrogen molecule was represented by a Gaussian wave function. We find that our CPE model gives the better agreement with the measurements in spite of its simplicity.

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1. Introduction

Positron impact ionization of molecules was recently studied both experimentally and theoretically. Experimental total ionization cross sections were measured for H_2 [1,2], N_2 [3], O_2 [4], CO [5], CO_2 [6] and for organic molecules [7].

The theoretical studies have been limited so far to molecular hydrogen. Distorted wave calculations have used a one-center formalism [8] or two-center molecular wavefunctions [9,10]. The paper by Campeanu et al. [10] used a Gaussian representation of the molecule which can be employed for more complex molecules. In this paper we will use the method of [10] for molecular nitrogen.

2. Theory

The triple differential cross section for the ionization of a homonuclear molecule by positron impact may be written as

$$\frac{d^3\sigma}{d\hat{\mathbf{k}}_\mathbf{f}\, d\hat{\mathbf{k}}_\mathbf{e}\, dE_e} = \sum_r \frac{(2\pi)^4}{E_i} |f_r|^2, \qquad (1)$$

where E_i is the energy of the projectile, E_e the energy of the ejected electron, while $\hat{\mathbf{k}}_\mathbf{e}$ and $\hat{\mathbf{k}}_\mathbf{f}$ stand for the direction of the momenta of the ejected electron and scattered positron, respectively. The summation over r is done over all occupied molecular orbitals. The amplitude can be written as

$$f_r = \langle \phi_f(\mathbf{r_1}) \phi_e(\mathbf{r_2}) | V(r_{12}) | \phi_i(\mathbf{r_1}) \phi_r(\mathbf{r_2}) \rangle, \qquad (2)$$

where ϕ_i and ϕ_f stand for the wavefunction of the incident and scattered positron, respectively, ϕ_e is the wavefunction of the ejected electron, while ϕ_r describes the initial state (orbital) of the active electron. In order for Eq. (1) to be valid, the ejected electron wave function must be orthogonalized to the target wave function. In the above amplitude $\mathbf{r_1}$ is the position vector of the positron, while $\mathbf{r_2}$ stands for the position vectors of the active electron. We are assuming in this model that the electron orbitals in the residual molecular ion are the same as in the target ion during the time of the collision.

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We have used Gaussian wavefunctions for the description of the ground state of the molecule with a basis set of three basis functions. The molecular orbital is a linear combination of Gaussian-type atomic orbitals, as described in [10] for s orbitals. In case of p orbitals we can write

$$\phi^{\mathrm{GF}}_{\mathrm{p}(x;y;z)}(\alpha_q, r) = N_{\mathrm{p}}(x;y;z)\mathrm{e}^{-\alpha_q\gamma_q^2 r^2}, \tag{3}$$

$$N_{\mathrm{p}} = [2^7(\alpha_q\gamma_q^2)^5/\pi^3]^{1/4}, \tag{4}$$

where $(x;y;z)$ symbolizes multiplication with x or y or z. For N_2 we have taken $\gamma_1 = 6.67$ for the $n = 1$ orbitals and $\gamma_2 = 1.95$ for $n = 2$ orbitals as giving the best values for the energy of the molecule.

In the case of σ orbitals the calculations are identical to those presented for H_2, i.e. the molecular orbitals are expanded in a Legendre series.

In the case of the π orbitals the previous method cannot be applied directly because of the φ dependence of the of the wavefunction. The π orbital may be written as

$$\phi_\pi(\mathbf{r_2}) = N_\pi[\Phi_{\mathrm{p}}(\mathbf{r}_{a2}) + \Phi_{\mathrm{p}}(\mathbf{r}_{b2})], \tag{5}$$

where

$$\Phi_{\mathrm{p}}(r_{a2}) = r_{a2}Y_{1m}(\hat{\mathbf{r}}_{\mathbf{a2}})\phi(\mathbf{r}_{a2}). \tag{6}$$

If we transform to the centre of mass coordinates denoted by $\mathbf{r}_2$ we have

$$r_{a2}Y_{1m}(\hat{\mathbf{r}}_{\mathbf{a2}}) = (r_2 - \delta_{m0}R_0/2)Y_{1m}(\hat{\mathbf{r}}_{\mathbf{2}}), \tag{7}$$

where R_0 is the internuclear separation. The orbitals depending on $\mathbf{r}_{b2}$ can be transformed in a similar manner. For the π orbitals with $m = \pm 1$ we have

$$\phi_\pi(\mathbf{r_2}) = N_\pi r_2 Y_{1m}(\hat{\mathbf{r}}_{\mathbf{2}})[\phi(r_{a2}) + \phi(r_{b2})]. \tag{8}$$

Now we can do the Legendre expansion

$$\phi(r_{a2}) + \phi(r_{b2}) = \sum_\lambda C_\lambda(r_2, R_0)P_\lambda(\cos\omega_2). \tag{9}$$

Here, because ω_2 is the angle between the $\mathbf{r_2}$ vector and the $0z$ axis, we can write

$$P_\lambda(\cos\omega_2) = \sqrt{\frac{4\pi}{2\lambda+1}}Y_{\lambda 0}(\hat{\mathbf{r}}_{\mathbf{2}}). \tag{10}$$

Further, for the π orbital we obtain

$$\phi_\pi(\mathbf{r_2}) = N_\pi \sum_\lambda \sqrt{\frac{4\pi}{2\lambda+1}}c_\lambda(r_2, R_0)Y_{\lambda 0}(\hat{\mathbf{r}}_{\mathbf{2}})Y_{1m}(\hat{\mathbf{r}}_{\mathbf{2}}), \tag{11}$$

with

$$c_\lambda(r_2, R_0) = r_2 C_\lambda(r_2, R_0). \tag{12}$$

The π orbitals with $m = 0$ can be handled in a similar way. The product of two spherical harmonics may be expressed as

$$Y_{\lambda 0}(\hat{\mathbf{r}}_{\mathbf{2}})Y_{1m}(\hat{\mathbf{r}}_{\mathbf{2}}) = \sum_{l_b m_b}\sqrt{\frac{(2\lambda+1)3}{4\pi(2l_b+1)}}C^{l_b 0}_{\lambda 010}C^{l_b m_b}_{\lambda 01m}Y_{l_b m_b}(\hat{\mathbf{r}}_{\mathbf{2}}), \tag{13}$$

where the $C^{l_b 0}_{\lambda 010}$ are the usual vector coupling coefficients. Transforming from the molecular frame into the lab frame (denoted by primes) we get

$$Y_{l_b m}(\hat{\mathbf{r}}'_{\mathbf{2}}) = \sum_\mu Y_{l_b\mu}(\hat{\mathbf{r}}_{\mathbf{2}})D^{l_b}_{\mu m}(\alpha, \beta, \gamma), \tag{14}$$

where the $D^{l_b}_{\mu m}(\alpha, \beta, \gamma)$ are the matrix elements of the rotation operator which rotates the molecular frame through the Euler angles (α, β, γ) into the laboratory frame. Finally, the π orbital may be expressed as

$$\phi_\pi(\mathbf{r_2}) = N_\pi \sum_\lambda c_\lambda(r_2, R_0)\sum_{l_b}\sqrt{\frac{3}{2l_b+1}}C^{l_b 0}_{\lambda 010}C^{l_b m_b}_{\lambda 01m} \times \sum_\mu Y_{l_b\mu}(\hat{\mathbf{r}}'_{\mathbf{2}})D^{l_b}_{\mu m}(\alpha, \beta, \gamma). \tag{15}$$

Since the molecules have an arbitrary orientation in the laboratory frame, we average the cross section over the Euler angles, using

$$\int_0^{2\pi}\int_0^{\pi}\int_0^{2\pi} D^{*l'_b}_{\mu' m'}(\alpha, \beta, \gamma)D^{l_b}_{\mu m}(\alpha, \beta, \gamma)\mathrm{d}\alpha\sin\beta\mathrm{d}\beta\mathrm{d}\gamma = \frac{8\pi^2}{2l_b+1}\delta_{\mu\mu'}\delta_{mm'}\delta_{l_b l'_b}. \tag{16}$$

In the calculation of the total cross section, we must also average over the angles of the outgoing electron and positron as well as the energy of the ejected electron as described in [11] for the atomic case.

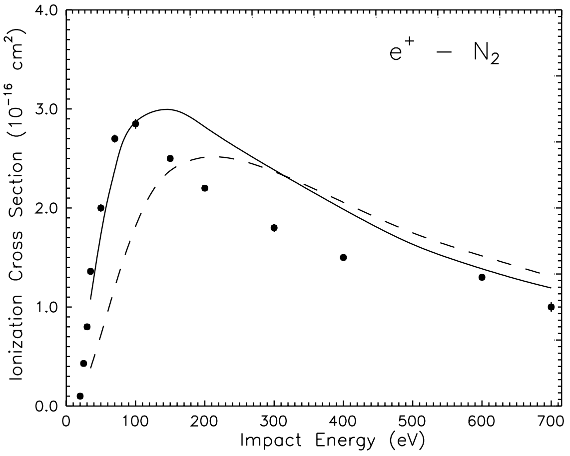

Fig. 1. Positron impact ionization of N_2. The experimental points are from Blume et al. [5]. Our CPE and CPE4 model results are represented by the continuous and dashed curves, respectively.

In the expansion on l_b for gerade orbitals only the terms with even l_b are nonzero, while for ungerade orbitals only odd l_b contributes. In our present calculations we have taken into account only the terms with $l_b \leqslant 3$. The terms with $l_b > 3$ contribute less then 0.5%, to the total cross sections and have been neglected.

Here we have used the models CPE and CPE4 previously applied to positron ionization of H_2 in [10]. In these models the free particles are represented by Coulomb waves with various effective charges as defined in [10].

3. Results and discussion

In Fig. 1 we present the integrated ionization cross sections corresponding to models CPE and CPE4 together with the experimental data of Bluhme et al. [3]. Our simpler model CPE is in better agreement with the experimental results in the region of the peak of the cross section and converges to the experiment as the impact energy increases. It is interesting to note that for the ionization of noble gases and molecular hydrogen model the more elaborate CPE4 model was in better agreement with the experiment than CPE [12].

4. Conclusions

Our calculations show that the use of distorted-wave models with Gaussian wave function for the molecular target can produce data which are in reasonable agreement with the experiment even for more complex molecules. We intend to try the same approach for other complex molecules.

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