Interference Effects in the Ionization of Diatomic Molecules

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Interference effects in the ionization of H_2 by charged particle and photon impact have been investigated. For the charged particle impact the influence of the residual molecule on the final state has been taken into account in a perturbative way. In case of the photoionization various final-state wavefunctions have been used. The effects of the molecular orientation on the differential photoionization cross sections and the contribution of different partial waves have been also analyzed.

Keywords: Interference; Ionization; Photoionization

I. INTRODUCTION

Interference effects in the photoionization of the hydrogen molecule due to the two-center character of the target have been predicted by Cohen and Fano [1] long time ago. In the last few years special interest was given to the study of these effects. Interference has been evidenced experimentally [2–5] and has been investigated theoretically [6–10] by several groups for the charged particle impact ionization. In case of the photoionization there are mainly theoretical descriptions [11, 12], but interference can be evidenced from the published experimental data [13], as it was shown in [14].

In our previous studies [6, 14], in order to perform the calculations analytically, the final states have been described by plane waves, and the interference pattern was governed only by the two-center character of the initial wavefunction. The experimental study of Stolterfoht *et al.* [4] suggested that the details of the interference effects may be influenced by the two-center character of the final state.

In the present paper we improve the description of our final state by two different methods. In the case of ionization by charged projectiles the interaction of the ejected electron with the nuclei is taken into account as a perturbation. For the photoionization we have tried two different types of two-center continuum wavefunctions.

II. THEORY

A. Ionization by fast charged particles

Concerning the ionization by charged projectiles, the theory for the first-order [6] and second-order [15] theory have been described in detail elsewhere. The cross section is obtained by integrating over the impact parameter and averaging over the angles of the molecular axis the transition probability, which is the sum of the first and second-order amplitudes

$$\sigma = \frac{1}{4\pi}\int d\hat{\mathbf{D}}\int d^2\mathbf{b}|a^{(1)}+a^{(2)}|^2. \tag{1}$$

In the second-order amplitude the first perturbation, $V(Z)$, is the interaction of the electron with the projectile, while the second one, W, is the electron-nuclei interaction. As explained in [16], this amplitude can be written as

$$a^{(2)} = -\frac{\pi}{v_p}\int Kd\hat{\mathbf{K}}\int_{-\infty}^{+\infty} dZ\langle\Psi_{\mathbf{k}}|W|\Psi_{\mathbf{K}}\rangle e^{iqZ}\langle\Psi_{\mathbf{K}}|V(Z)|\Psi_i\rangle. \tag{2}$$

Here Ψ_i stands for the initial and $\Psi_{\mathbf{k}}(\mathbf{r})$ for the final state of the active electron, while v_p is the velocity of the projectile, Z the coordinate of the projectile, $\mathbf{K}$ and $\mathbf{k}$ are the momenta of the electron in the intermediate and final state, respectively, and q the minimum momentum transfer. In spite of the fact that $\Psi_{\mathbf{k}}(\mathbf{r})$ is taken to be a plane wave, the two-center character of the final state is introduced by the perturbation W.

B. Photoionization

In the case of photoionization, the differential cross section for a linearly polarized radiation may be expressed as

$$\sigma = \frac{4\pi^2\alpha}{\omega}|M_{fi}(\omega)|^2, \tag{3}$$

where α is the fine-structure constant, ω the photon angular frequency and M_{fi} the transition matrix element. In the dipole approximation the matrix element may be written as

$$M_{fi}(\omega) = \langle\psi_i|\varepsilon\nabla_r|\psi_f\rangle \tag{4}$$

in velocity form, while in length form we have

$$M_{fi}(\omega) = i\omega\langle\psi_i|\varepsilon\mathbf{r}|\psi_f\rangle, \tag{5}$$

ε being the polarization vector of the electromagnetic wave. The initial state of the active electron is given by a linear combination of two atomic orbitals

$$\psi_i = \frac{1}{\sqrt{2(1+S)}}[\psi_0(\mathbf{r}_a)+\psi_0(\mathbf{r}_b)], \tag{6}$$

where $\psi_0(\mathbf{r}_{a,b})$ are the atomic $1s$ orbitals centered at the nuclei a or b, while S is the overlap integral between these two orbitals.

For the description of the final state we have used two types of two-center wavefunctions. The first is the 2C wavefunction

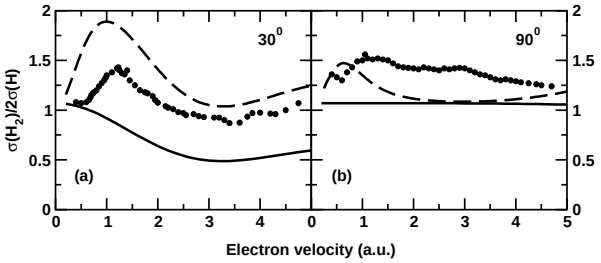

FIG. 1: $\sigma(H_2)/2\sigma(2H)$ ionization cross section ratio for 68 MeV/u Kr^{33+} projectiles as a function of the ejected electron velocity. Our first-order (solid lines) and second-order (dashed lines) results are compared with the experimental data of Stolterfoht *et al.* [3].

used also by Walter and Briggs [11], a plane wave multiplied by two Coulomb distortion factors centered at the nuclei

$$\psi_{2C} = (2\pi)^{-\frac{3}{2}} e^{i\mathbf{kr}} e^{-\pi\gamma} e^{ik(r_a+r_b)} \Gamma^2(1+i\gamma) \times H(1+i\gamma,1;-i(kr_a-\mathbf{r}_a\mathbf{k}) H(1+i\gamma,1;-i(kr_b-\mathbf{r}_b\mathbf{k}). \quad (7)$$

$H(a,b;x)$ is the confluent hypergeometric function and $\gamma = \rho/k$ is the Sommerfeld parameter. The second wavefunction is a linear combination of two Coulomb functions:

$$\psi_{2CA} = \frac{1}{2}(2\pi)^{-\frac{3}{2}} e^{-\frac{\pi\gamma}{2}} \Gamma(1+i\gamma) \times \left[e^{ikr_a} H(1+i\gamma,1;-i(kr_a-\mathbf{kr}_a)) + e^{ikr_b} H(1+i\gamma,1;-i(kr_b-\mathbf{kr}_b)) \right] \quad (8)$$

This wavefunction does not reproduce correctly the asymptotic behavior, but it describes well the two-center character of the final state in the vicinity of the nuclei.

The calculations have been performed also directly, but with an expansion of the initial and final wavefunctions in terms of spherical harmonics.

III. RESULTS

A. Ionization by fast charged particles

For the charged particle impact, calculations were performed for 68 MeV/u Kr^{33+} projectiles. Fig. 1 represents the $\sigma(H_2)/2\sigma(H)$ differential cross section ratio as a function of the ejected electron velocity for 30^0 and 90^0 electron ejection angles. As one may observe, the improvement of the description of the electron-nuclei interaction in the final state leads to a better agreement with the experimental data. Specifically, the decrease of the cross section ratio as the velocity tends to 0 is reproduced by only this second-order calculation.

B. Photoionization

In the case of photoionization, the calculations were made for fixed orientations of the molecular axis, and a fixed direction of the ejected electron momentum. In these conditions direct comparison with the results of Fojón *et al.* [12] is not possible, but interesting conclusions can be formulated. Fig. 2 shows the differential photoionization cross section as a function of the electron momentum, when the electron is ejected in the direction of the polarization vector, and the axis of the H_2 molecule has the same direction. This geometry was chosen because in this case interference effects are maximal. Comparing to our previous results (obtained with plane waves, in velocity and length gauge), the results seem to be improved. First, we obtain the first minimum in the cross section at higher value of the momentum than for the plane waves, in accordance with the findings of Fojón *et al.* [12]. Second, if the momentum of the ejected electron approaches

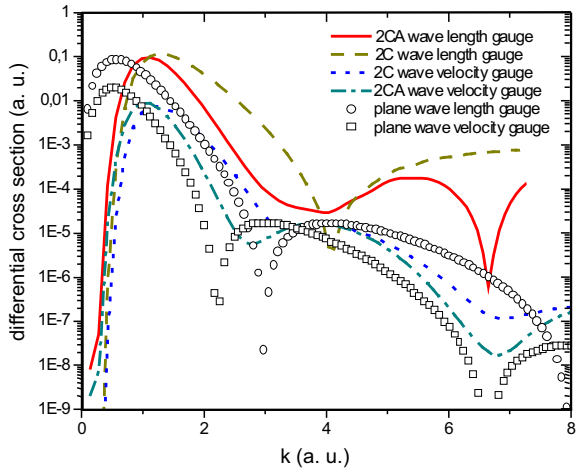

FIG. 2: Differential cross section for the photoionization of H_2 as a function of the ejected electron momentum, with electron ejection and the molecular axis in the direction of the polarization vector, obtained with different final-state wavefunctions.

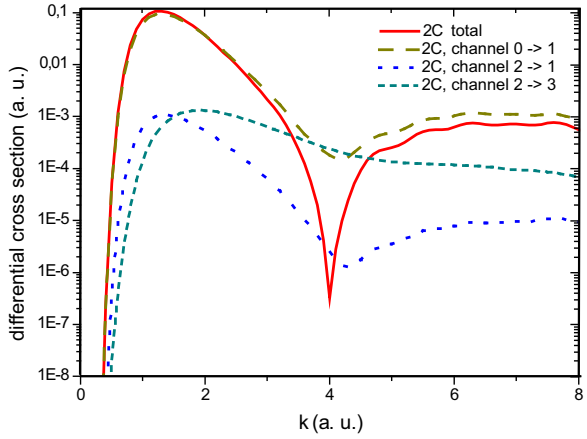

FIG. 3: Partial-wave contributions to the differential cross section for the photoionization of H_2 as a function of the ejected electron momentum, with electron ejection and the molecular axis in the direction of the polarization vector, obtained with the 2C wavefunction

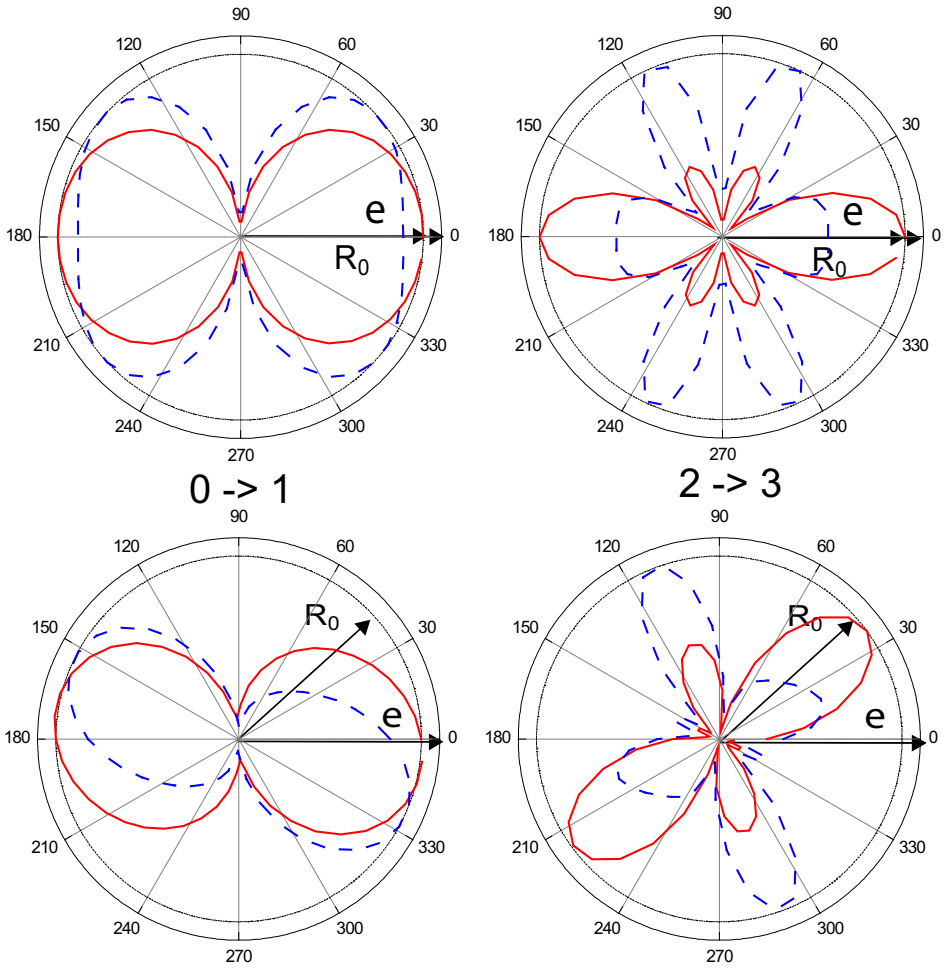

FIG. 4: Polar representation of the normalized differential photoionization cross section of the H_2 for 82.4 eV photon energy as a function of the ejection direction for the two main partial wave channels and for two different orientations of the R_0 axis (solid line – 2C, dashed line – 2CA wavefunctions).

zero, the cross section decreases much more rapidly than for plane waves, leading to a decreasing molecule-to-atom ratio, in accordance with the experiment [13] and the calculations of Fojón *et al.* [12]. Our finding suggests that not only electron correlation (as stated by Fojón *et al.* [12]) but also the two-center character of the wavefunction lead to this effect.

The partial-wave analysis (Fig. 3) of the photoionization shows, as expected, that the $l = 0 \rightarrow l = 1$ channel is dominant for most of the momenta values, but also this channel shows more pronounced interference effects than the $l = 2 \rightarrow l = 3$ channel. It is interesting that the interference between the different channels leads to a more pronounced minimum than the $l = 0 \rightarrow l = 1$ channel alone.

The polar graphs on Fig. 4 show the angular distribution of the ejected electron for the two main channels, for 82.4 eV photon energy and two different molecular orientations. For the molecular axis in the direction of the polarization vector, the p and the f character of the final states may be easily recognized. Molecular orientation has a less influence for the $l = 0 \rightarrow l = 1$ channel, but for the $l = 2 \rightarrow l = 3$ channel we observe a tendency of ejecting the electron in the molecular axis direction, mainly for the 2C wavefunction.

IV. CONCLUSIONS

In conclusion, taking into account the two-center character of the final-state wavefunction in the ionization of H_2 is important in the study of interference effects. In the case of charged particle impact the electron–residual ion interaction has been taken into account as a perturbation, and the behavior of the cross section ratio at low ejected velocities has been improved. In the case of photoionization, two different types of final wavefunctions have been tried. At this stage no direct comparison with the experiment is possible, but the 2C function seems to be more realistic.

[1] H.D. Cohen and U. Fano, Phys. Rev. **150**, 30 (1966).
[2] N. Stolterfoht, B. Sulik, V. Hoffmann, B. Skogvall, J.Y. Chesnel, J. Ragnama, F. Frémont, D. Hennecart, A. Cassimi, X. Husson, A.L. Landers, J. A. Tanis, M. E. Galassi, and R. D. Rivarola, Phys. Rev. Lett. **87**, 023201 (2001).
[3] N. Stolterfoht, B. Sulik, L. Gulyás, B. Skogvall, J.Y. Chesnel, F. Frémont, D. Hennecart, A. Cassimi, L. Adoui, S. Hossain, and J.A. Tanis, Phys. Rev. A **67**, 030702 (2003).
[4] N. Stolterfoht, B. Sulik, B. Skogvall, J.Y. Chesnel, F. Frémont, D. Hennecart, A. Cassimi, L. Adoui, S. Hossain, and J.A. Tanis, Phys. Rev. A **69**, 012701 (2004).
[5] S. Hossain, A.S. Alnaser, A.L. Landers, D.J. Pole, H. Knutson, A. Robinson, B. Stamper, N. Stolterfoht, and J.A. Tanis, Nucl. Instr. and Meth. in Phys. Res. B **205**, 484 (2003).
[6] L. Nagy, L. Kocbach, K. Póra, and J. P. Hansen, J. Phys. B. **35**, L453 (2002).
[7] M. E. Gallasi, R. D. Rivarola, P. D. Fainstein, and N. Stolterfoht, Phys. Rev. A **66**, 052705 (2002).
[8] G. Laurent, P. D. Fainstein, M. E. Galassi, R. D. Rivarola, L. Adoui, and A. Cassimi, J. Phys. B **35**, L495 (2002).
[9] C.R. Stia, O. A. Fojón, P. F. Weck, J. Hanssen, and R.D. Rivarola, J Phys. B: At. Mol. Phys. **36**, L257 (2003).
[10] L. Sarkadi, J. Phys. B **36**, 2153 (2003).
[11] M. Walter and J. Briggs, J. Phys. B: At. Mol. Phys. 32, 2487 (1999).
[12] O.A. Fojón, J. Fernandez, A. Palacios, R. D. Rivarola, and F. Martin, J. Phys. B **37**, 3035 (2004).
[13] J.A.R. Samson and G.N. Haddad, J. Opt. Soc. Am. B **11**, 277 (1994).
[14] L. Nagy, S. Borbély, and K. Póra, Phys. Lett. A **327**, 281 (2004).
[15] K. Póra and L. Nagy, Nucl. Instrum. Methods B **233**, 293 (2005).
[16] L. Nagy, Nucl. Instrum. Methods B **124**, 271 (1997).