

Young-type interference patterns in electron emission spectra produced by impact of swift ions on H₂ molecules

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The existence of Young-type interference patterns, recently measured [N. Stolterfoht *et al.*, Phys. Rev. Lett. **87**, 023201 (2001)] in double-differential cross sections for single-electron ionization of H₂ molecules by ion impact, is theoretically supported by calculations obtained using a molecular distorted-wave model introduced here. The importance of adequately describing the target as a molecule is emphasized. The contributions from direct and interference terms are separated.

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Recent measurements of the electron emission spectra in collisions between 60 MeV/u Kr³⁴⁺ ions and H₂ molecules have shown evidence of interference effects [1]. The oscillations observed in the spectra were related to those expected from a Young-type two-slit experiment [2,3] in which the sources of coherent emission are the two nuclei of the molecule. This was demonstrated with a simple model in which the initial bound state of the active electron was represented by a linear combination of atomic hydrogen wave functions separated by an equilibrium internuclear distance ρ [4]. The oscillatory behavior remains even though the calculation of the differential cross sections involves an integration over all the molecular axis orientations.

The appearance of interference effects arising from the two-center geometry of the H₂ molecule has been reported for other processes. During the 1960s, theoretical predictions were done in the cases of electron capture from H₂ by proton impact [5] and in photoionization of H₂⁺ [6]. More recent studies [7,8] have allowed to expand the knowledge about the process leading to experimental verification of the theory [9].

The model calculations presented in Ref. [1] made use of various assumptions which allow to obtain a closed formula for the doubly differential cross section as a function of electron energy E_e and angle θ_e . These approximations are only valid at high enough impact energy, as will be shown here. The aim of this work is to present a theoretical model which includes the molecular character of the target and is free of these approximations. In this way we expect to obtain further evidence to support the existence of the interference effect. For this purpose we extend the continuum-distorted-wave-eikonal-initial-state (CDW-EIS) model for single ionization (see Ref. [10], and references therein) to the case of bare-ion impact on a two-center molecular target. We employ the

straight line version of the impact-parameter approximation and follow the derivation given for single-electron capture [11]. The collision is described from a reference frame fixed on the middle point of the molecular axis and the z quantization axis is chosen in the direction of the incident projectile velocity \vec{v} . Atomic units are used except where otherwise stated.

As we will consider high impact energies, we assume that the relative positions ($\vec{\rho}$) of the nuclei in the molecule remain fixed throughout the collision and differential cross sections are averaged over all possible molecular orientations. As was shown in a recent work for electron-impact ionization of H₂, when the nonionized electron is promoted from the ground state of the target ($^1\Sigma_g^+$) to the ground state of the residual H₂⁺ molecular ion ($^2\Sigma_g^+$), this condition is equivalent to summing the contributions from all final rotational states and to average the contribution of all initial magnetic quantum numbers corresponding to a given initial angular-momentum quantum number [12]. Moreover, using the closure relation for all final vibrational states and assuming that the electronic transition matrix element depends weakly on ρ , differential cross sections can be calculated as a pure electronic transition [13].

In order to further reduce the two-electron problem to a one active electron reaction we consider in the present case an independent electron approximation where the nonionized electron remains frozen in its initial orbital $\varphi_i^P(\vec{x}')$ during the collision. As we are only interested on the cross-section differential on the emitted electron momenta the interactions of the projectile with the nuclei of the molecule and the nonionized electron can be excluded from the theoretical treatment [11]. Thus the initial and final total wave functions are given by

$$\Psi_{i,f}^{+,-} = \chi_{i,f}^{+,-}(\vec{x}, t) \varphi_i^P(\vec{x}'), \quad (1)$$

where \vec{x} (\vec{x}') is the active (passive) electron coordinate. The

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distortion due to the projectile field is only taken into account on the initial and final active electron wave functions χ_i^+ and χ_f^- . The post version of the scattering amplitude can be reduced to the one active electron amplitude,

$$A_{if}^+(\vec{b}, \vec{\rho}) = -i \int_{-\infty}^{+\infty} dt \left\langle \chi_f^- \left| \left(H_a - i \frac{\partial}{\partial t} \right)^\dagger \right| \chi_i^+ \right\rangle, \quad (2)$$

with

$$H_a = -\frac{1}{2} \nabla^2 - \frac{Z_T}{x_1} - \frac{Z_T}{x_2} - \frac{Z_P}{s} + V_{ap}(\vec{x}, \vec{\rho}) \quad (3)$$

the active electron Hamiltonian, \vec{b} the impact parameter, Z_T (Z_P) the target (projectile) nuclear charge, \vec{x}_1 (\vec{x}_2) the active electron coordinate with respect to the molecular center 1 (2), and \vec{s} its position with respect to the projectile nucleus. The potential V_{ap} takes into account the influence of the nonionized electron on the active one [11]. The initial and final distorted-wave functions are proposed as $\chi_{i,f}^{+,-} = \varphi_{i,f} \mathcal{L}_{i,f}^{+,-}$, where φ_i (φ_f) is the initial (final) bound (continuum) state of the active electron, and \mathcal{L}_i^+ (\mathcal{L}_f^-) is the distortion factor in the entrance (exit) channel. In CDW-EIS model we have

$$\mathcal{L}_i^+(\vec{s}) = \exp\left(-i \frac{Z_P}{v} \ln(vs + \vec{v} \cdot \vec{s})\right), \quad (4)$$

$$\mathcal{L}_f^-(\vec{s}) = N(\zeta) {}_1F_1(-i\zeta; 1; -ip_s - i\vec{p} \cdot \vec{s}), \quad (5)$$

where \vec{p} is the electron momentum in the projectile frame, $\zeta = Z_P/p$ and, $N(\zeta) = \exp(\pi\zeta/2) \Gamma(1+i\zeta)$. The initial bound wave function is approximated by a variational single-zeta function

$$\varphi_i(\vec{x}, t) \cong [\xi_1(\vec{x}_1) + \xi_2(\vec{x}_2)] e^{-i\epsilon_i t}, \quad (6)$$

with ϵ_i the orbital energy of the active electron and

$$\xi_j(\vec{x}) = N_i(\rho) \left(\frac{Z^3}{\pi}\right)^{1/2} \exp(-Zx), \quad j=1,2, \quad (7)$$

where $N_i(\rho) = 0.5459$ is a normalization factor corresponding to the equilibrium internuclear distance $\rho = 1.4$ and $Z = 1.193$. For the present calculations we take the experimental value for single ionization of H_2 , $\epsilon_i = -0.566$. To represent the final continuum state we use the two-effective center approximation previously employed with success to study electron capture from H_2 by ion impact [11,14] and electron ionization of H_2 by electron impact [13]. In this approximation the molecular continuum is given by

$$\varphi_f^j(\vec{x}, t) = \phi(\vec{x}_j) e^{-i\epsilon_f t + i\beta_j \vec{k} \cdot \vec{\rho}/2}, \quad (8)$$

where $\epsilon_f = k^2/2$ (\vec{k}) is the ejected electron energy (momenta), $\beta_1 = -1$, $\beta_2 = +1$, and

$$\phi(\vec{x}_j) = (2\pi)^{-3/2} e^{i\vec{k} \cdot \vec{x}_j} N(\gamma) {}_1F_1(-i\gamma; 1; -ikx_j - i\vec{k} \cdot \vec{x}_j) \quad (9)$$

according to on which of the functions ξ_j it is projected in the integrand of the transition amplitude (2). In Eq. (9), $\gamma = Z_{eff}/k$ with $Z_{eff} = \sqrt{-2\epsilon_i}$. After a lengthy algebra the transition amplitude reduces to

$$A_{if}^+(\vec{b}, \vec{\rho}) \cong -i \sum_{j=1}^2 e^{-i\beta_j(\vec{k} \cdot \vec{\rho} - q_z \rho_z)/2} A_{if}^{eff,+}(\vec{b}_j), \quad (10)$$

with \vec{b}_j the impact parameter with respect to the molecular center j , $q_z = \Delta\epsilon_{fi}/v$ the z component of the momentum transfer $\vec{q} = -\vec{\eta} - q_z \hat{v}$ and $\Delta\epsilon_{fi} = \epsilon_f - \epsilon_i$. The functions $A_{if}^{eff,+}$ are transition amplitudes corresponding to effective atoms located at the position of each molecular center. The exponential term appearing in Eq. (10) contains the information of the interference between the coherent scattering processes taking place on the molecular centers.

Instead of the scattering amplitude (10) it is more convenient to work with its Fourier transform whose square modulus is given by

$$|R_{if}^+(\vec{\eta}, \vec{\rho})|^2 = 2\{1 + \cos[(\vec{k} + \vec{q}) \cdot \vec{\rho}]\} |R_{if}^{eff,+}(\vec{\eta})|^2, \quad (11)$$

where $R_{if}^{eff,+}$ is the Fourier transform of $A_{if}^{eff,+}$. Averaging over all molecular orientations we obtain the final expression for the doubly differential cross section as a function of electron energy and solid angle ($d\Omega$), which involves the integration over the transverse momentum transfer

$$\begin{aligned} \frac{d^2\sigma}{d\epsilon_f d\Omega} &= 8\pi k \int d\vec{\eta} \left(1 + \frac{\sin(|\vec{k} + \vec{q}|\rho)}{|\vec{k} + \vec{q}|\rho}\right) |R_{if}^{eff,+}(\vec{\eta})|^2 \\ &= S_d(\epsilon_f, \Omega) + S_i(\epsilon_f, \Omega). \end{aligned} \quad (12)$$

We can see that it results in two different terms, S_d and S_i , representing contributions from direct and interference processes, respectively. This approximation is the extension for molecular targets of the method developed in Ref. [15] for multielectronic atoms. Further details will be given in a separate paper.

Although all the information about the process can be obtained from the doubly differential cross section (DDCS), this quantity is not appropriate to highlight small effects such as the interference. The DDCS decreases monotonically by several orders of magnitude in the electron energy range of interest. Therefore it is better to plot the ratio $R(k, \theta)$ between the DDCS for ionization of H_2 and two times the DDCS for ionization of H. If there are no effects due to the structure of the molecule we can expect that at the high impact energy considered here, the ratio will give a value close to 1 (it may differ from this value due to the different binding energies of H_2 and H, of the corresponding effective charge Z_{eff} and the normalization of the bound-state wave functions).

In Fig. 1 we plot $R(k, \theta)$ as a function of electron velocity at fixed emission angles of 20° and 30° in comparison with

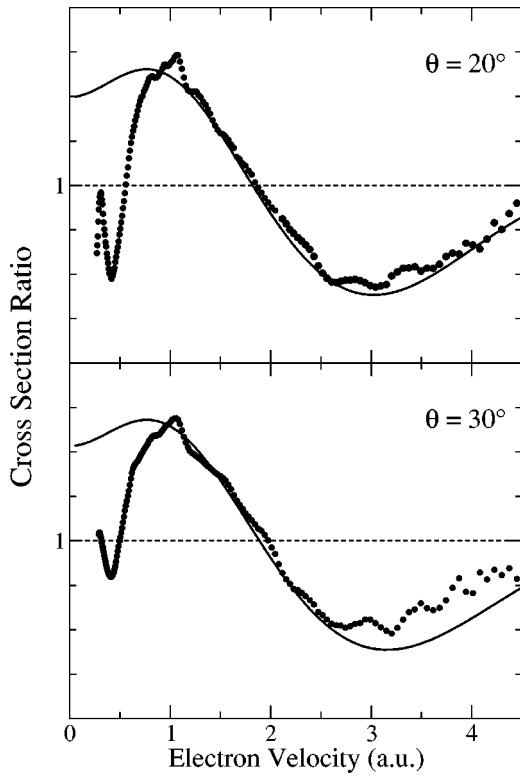


FIG. 1. $R(k, \theta)$ as a function of electron velocity at 20° and 30° emission angles, for 60 MeV/u Kr^{34+} impact. Experiment: (●), from Ref. [1]. Theory: solid line, present CDW-EIS calculation.

the recent experimental data from Ref. [1]. It should be noted that the experimental data have been obtained dividing experimental DDCS for H_2 by theoretical DDCS for two H atoms. In addition, the experimental ratios were normalized in Ref. [1] according to a straight-line fit that takes into account discrepancies between theory and experiment at high electron velocity. This would be unnecessary if experimental results for H target were available. The figure shows that the CDW-EIS model including the molecular structure is in very good agreement with the experimental data. Some disagreement appears at very low electron velocity where, however, the experimental uncertainties are larger. We note that neither our present calculations nor the simplified model presented in Ref. [1] reproduce the structure that appears below 1 a.u. At higher velocities, the theoretical results show, as in the experiment, a distinctive interference pattern. This oscillation can be studied in more detail by looking at the two contributions to the DDCS (12). S_d is independent of the molecular internuclear distance while S_i presents an oscillatory behavior which depends on \vec{q} and \vec{k} and tends to zero at high electron velocity. In Fig. 2 we present the results of each term and the sum for the case of 20° ejection angle. Both terms give similar values at low electron energy. The main difference is that while S_d increases monotonically, S_i shows a damped oscillatory behavior which demonstrates that the interference pattern arises from a coherent emission from both molecular centers.

In our calculation the interference pattern depends on the electron emission angle [see Eq. (12)]. However, at the high

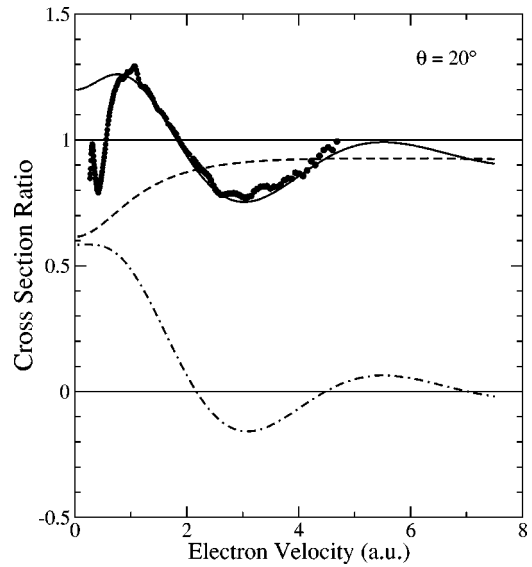


FIG. 2. Same as Fig. 1 at 20° emission angle. Experiment: (●), from Ref. [1]. Theory: solid line, present CDW-EIS calculation; dashed-line, contribution from S_d ; dot-dashed line, contribution from S_i .

impact energy considered here the variation is quite small. To test this behavior in more detail we have performed calculations of $R(k, \theta)$ for H^+ impact at a much lower impact energy (1 MeV). The results at two emission angles, 45° and 150° , are shown in Fig. 3. Surprisingly the interference persists and is strong as in the Kr^{34+} case. However, a major

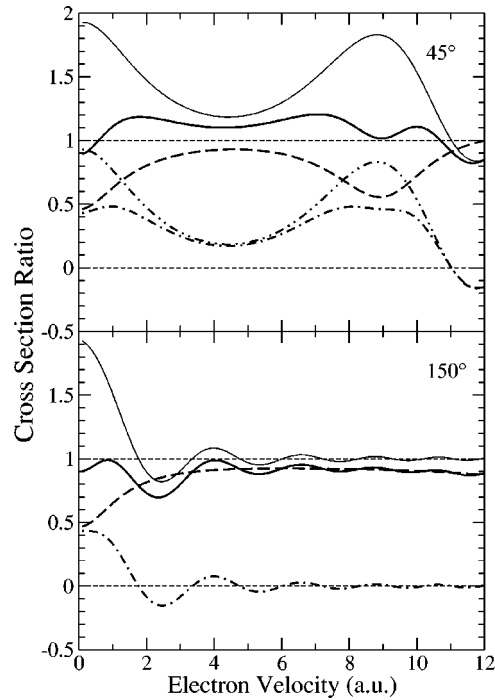


FIG. 3. Same as Fig. 1 at 45° and 150° emission angles for 1 MeV H^+ impact. Theory: solid line, present CDW-EIS calculation; dashed line, contribution from S_d ; dot-dashed line, contribution from S_i ; thin-solid line, $(S_d+S_i)/S_d$; double-dot-dashed line, S_i/S_d .

difference is that the interference pattern is strongly dependent on the emission angle. It is striking that, in the electron velocity range considered, $R(k, \theta)$ shows a large plateau in the forward direction and, on the contrary, a clear interference pattern in the backward direction. Therefore, the former must be analyzed with more care.

At the impact energy considered here, the electron energy range includes the range in which the binary encounter peak appears. As H and H₂ have different Compton profiles, the ratio $R(k, \theta)$ may have additional structures due to this effect. This can be seen when we plot the ratios corresponding to the contributions from S_d (dashed line) and S_i (dot-dashed line). While at 150° the contribution from S_d shows a smooth monotonic behavior, as already observed for the Kr³⁴⁺ case, for 45° it presents a dip at the position of the binary encounter peak. We have checked that the same feature appears at all emission angles below 90°. Therefore the differences in the target bound wave functions give rise to additional structures which have a very different origin than the oscillations from the interference pattern. To make a further check on this result we have plotted the ratio $(S_d + S_i)/S_d$ (thin solid line), which corresponds to a replacement of the H target with an effective one with the same Compton profile used for H₂. As the first term is equal to 1 at all electron energies, in this way we eliminate any structure other than the interferences. As predicted the structure in the position of the binary peak disappears and the ratio shows an

interference pattern which arises exclusively from the interference term S_i . This pattern is almost identical to the one obtained from the ratio between S_i and the results for the two H atoms (dashed line) as seen from the comparison with the ratio S_i/S_d (double-dot dashed line). Therefore the interference pattern is present in $R(k, \theta)$ although partially masked by the additional structures which appear from the contribution of S_d . This is confirmed by the fact that for 150° the interference pattern does not change at all.

In summary we have shown that theoretical calculations using the CDW-EIS model with a proper account for the molecular structure of H₂ can reproduce Young-type interference patterns observed in recent experiments. Contributions from direct and interference terms can be separated, showing their relative importance as a function of the emitted electron velocity. Our theoretical calculations show that Young-type interference patterns also appear at smaller impact energy where there is a strong dependence on the emission angle. In this case the results must be analyzed with care as the different Compton profiles of H and H₂ can give rise to oscillations in the ratio of doubly differential cross sections at the position of the binary encounter peak.

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