Tracing fingerprints of Young type interferences in angular distributions of ejected electrons from molecular targets

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Abstract. Interference patterns in angular distributions of ionized electrons due to coherent emission from the proximities of the nuclei of molecular hydrogen are investigated in (e,2e) collisions. It is shown that double and single differential cross sections show signatures of this effect.

1. Introduction

More than four decades ago, Samson and Cairns [1] obtained experimental photoabsorption total cross sections for N2 and O2 molecules, showing that some undulations appeared for photon energies in the 30-60 eV range. Studying the case of H2+ targets, Cohen and Fano [2] attributed the possible origin of this behavior, observed for larger molecules, to interference patterns due to coherent electron emission from the proximities of each one of the molecular centers. These studies were followed by other ones on photoionization not only of H2+ but also of H2 molecules [3-4].

Hanssen et al. [5] (see also Joulakian et al. [6]) investigated theoretically the effect for impact of electron beams, associating the predicted interference patterns in the ionization spectra with the famous two-slit Young experiment [7-10], with the two molecular centers of a H2+ molecular ion playing the rôle of the slits. As the considered spectrum corresponds to electrons ejected from the target we have in fact only the second-half of the Young experiment and so the effect is known as the “Young type interferences” one.

The effect was measured only thirty five years later, for electron ionization of H2 by 60 MeV/n Kr34+ ions in double differential cross sections (DDCS) as a function of the energy of the emitted electron at fixed values of the solid angle subtended by it [11]. In fact, to enhance the visibility of Young type structures experimental DDCS were divided by theoretical ones corresponding to effective hydrogen atoms. In further work the effect was shown to appear in the ratio of experimental DDCS of H2 molecules and H atoms [12]. The experimental evidence of coherent electron emission from molecules spurred a lot of theoretical and experimental research with ion beams, but also with photons and electrons beams (for a short review see reference [13]). In particular, an intense activity related to this subject was recently developed for photoionization of H2 and N2 molecules [14-18].
We focus our present interest on the case of electron beam impact on H$_2$ targets. Electrons as projectiles are fundamentally different from equivelocity ions because they are negatively charged and light mass particles with larger de Broglie wave-lengths. Interference patterns were measured for the first time in DDCS for 2.4 keV e$^-$ + D$_2$ collisions [19]. Further evidence of the existence of this effect was also given in triple differential cross sections (TDCS) for H$_2$ targets in coplanar asymmetric kinematics [20]. TDCS were presented as a function of the polar angle of the emitted electron at fixed emission energy and for a given projectile scattering angle in the forward direction. In fact, the ratio between experimental TDCS for hydrogen molecules [21] and theoretical TDCS for an effective atomic hydrogen showed an oscillatory behaviour associated with two-center coherent electron emission. Further works were also developed but investigating the comportment of the ratio of experimental TDCS for H$_2$ and He. In the first of them a supression of the recoil peak with respect to the binary encounter one was shown to appear as a consequence of partial destructive interference [22]. In a second one, for the same collision system and also within a coplanar asymmetric geometry, it was shown that depending of the kinematical conditions describing the emitted and scattered electrons in the exit channel, constructive patterns can be also observed, as an increase of the recoil peak with respect to the binary one instead of the supression measured for destructive interference [23]. In all these investigations the theoretical analysis of the data to support the constructive and destructive interference hypothesis was based on an interference factor previously predicted by Stia et al. [24]. It should be also mentioned that in general the experimental TDCS were well described using distorted wave [24-25] and convergent close-coupling [26] models. 

Very recently, the Young-type interference effect was measured in DDCS for impact of 8 keV-e$^-$ on hydrogen molecules as a function of the momentum coordinates of the ejected electron [27-28]. In particular, ionized electron angular distributions were determined. Moreover, fingerprints of the effect were found also in angular- and energy-dependent single differential cross sections (SDCS). The scope of the present work is to give a theoretical analysis of the angular spectrum giving some physical explanations of the experimental data behaviour.

Atomic units will be used in the following except where otherwise stated.

2. Theory

Let us study the ionization process produced by the impact of a fast electron beam on a hydrogen molecule,

\[ e^- + H_2 \left( ^1 \Sigma_g^+ \right) \rightarrow 2e^- + H_2^+ \left( ^3 \Sigma_g^+ \right), \quad (1) \]

Highly asymmetric kinematics is considered, so that exchange effects can be neglected. Furthermore, the internuclear molecular axis is considered to remain as frozen during the collision, so that vibration and rotation of the target are not taken into account. This approximation, supported by the fact that the collision time is much shorter than the vibrational and rotational ones, has been previously studied in detail [29] and applied with success [30-32]. Moreover, the electronic reaction is assumed to be produced at the fixed equilibrium internuclear distance $\rho_0$ of the initial H$_2$ state.

The process is described from the center of mass of the target and the initial wavefunction is represented as

\[ \psi_i(\vec{r}_1, \vec{r}_2, \vec{R}) = \frac{\exp(i\vec{k}_i \cdot \vec{R})}{(2\pi)^{\frac{3}{2}}} \phi_i(\rho_0, \vec{r}_1, \vec{r}_2) \quad (2) \]

where $\vec{k}_i$ is the reduced mass of the projectile electron and $\vec{R}$ its position, $\vec{r}_j$ ($j=1,2$) being the position of the $j$th electron and $\phi_i$ the Heitler-London type wavefunction given by

\[ \phi_i(\rho_0, \vec{r}_1, \vec{r}_2) = N_i(\rho_0) \left( e^{-\alpha_0 r_1} e^{-\alpha_0 r_2} + e^{-\alpha_0 r_1} e^{-\alpha_0 r_2} \right), \quad (3) \]
with \( N_i(\rho_i) \) a normalization factor, \( \rho_i = 1.406 \), \( \alpha = 1.166 \) and, \( \vec{r}_{ju}(\vec{r}_{jb}) \) the position of the \( j \)-th electron with respect to the molecular nucleus \( a \ (b) \).

A two-effective center approximation is adopted to describe the final wavefunction [29]. In such approximation the single ionization process is assumed to be produced in the proximities of each one of the molecular centers, while the non-ionized electron completely screens the charge of the nucleus from which ionization is not produced. Without loss of generality, let us take the electron 1 as the one ionized. Thus, if it is ionized from the proximities of the center \( a \ (b) \), the electron 2 is considered to completely screen the center \( b \ (a) \). Thus, the final wavefunction in the exit channel is chosen as

\[
\psi_f(\vec{r}_1, \vec{r}_2) = \frac{\exp \left( i \vec{k} \cdot \vec{R} \right)}{2\pi^{3/2} (\rho_0)^{1/2}} \phi_f(\rho_0, \vec{r}_2) C(\vec{k}_e, \vec{r}_i, \gamma); \quad i = a, b \tag{4}
\]

where \( \vec{k}_s \) and \( \vec{k}_e \) are the linear momenta of the scattered and ejected electrons respectively, \( \gamma = -Z_T/\gamma_e \) with \( Z_T \) the nuclear charge associated with each one of the molecular centers and \( \phi_f \) the wavefunction corresponding to the fundamental bound state of the residual target given by

\[
\phi_f(\rho_0, \vec{r}_2) = N_f(\rho_0)(e^{-\beta_e r_e} + e^{-\beta_e r_e}) \tag{5}
\]

In Equation (5), \( N_f(\rho_0) \) is a normalization factor, the parameter \( \beta = 1.3918 \) and \( C(\vec{k}_e, \vec{r}_i, \gamma) \) is the Coulomb wavefunction describing the electron in a continuum state of the \( i \)-molecular center,

\[
C(\vec{k}_e, \vec{r}_i, \gamma) = \frac{\exp \left( i \vec{k} \cdot \vec{r}_i \right)}{2\pi^{3/2}} \Gamma(1-i\gamma)\exp\left(\frac{\pi\gamma}{2}\right) F_1\left[i\gamma, 1, -i(\vec{k}_e \cdot \vec{r}_i + \vec{k}_e \cdot \vec{r}_i)\right]; \quad i = a, b . \tag{6}
\]

Neglecting indirect contributions which consider that the electron is ionized from one of the molecular center due to the interaction of the projectile with the other center or with the passive electron (see reference [27]) and after some algebra, the four-fold differential cross section (FDCS) can be obtained,

\[
\sigma^{(4)} = \frac{d^4\sigma}{d\Omega_{\rho_0} d\Omega_{\rho_0} d\Omega_{\rho_0} d\Omega_{\rho_0} d(k^2_e/2)} \approx 2[1 + \cos(\vec{\rho} \cdot \vec{\rho}_0)]\sigma^{(3)}_{\text{eff}}, \tag{7}
\]

with \( \Omega_{\rho_0}, \Omega_{\rho_e} \) and \( \Omega_{\rho_s} \), the solid angles subtended by the molecular orientation and by the ejected and scattered electrons, respectively and, \( \vec{\rho} = \vec{k}_e - \vec{k}_s \), where \( \vec{k} = \vec{k}_e - \vec{k}_s \) is the momentum transferred to the target and \( \sigma^{(3)}_{\text{eff}} \) represents an effective one-center triple differential cross section. Moreover, a TDCS can be calculated after integration on \( \Omega_{\rho_0} \), so that

\[
\sigma^{(3)} = \frac{d^3\sigma}{d\Omega_{\rho_e} d\Omega_{\rho_s} d(k^2_e/2)} \approx 2I\sigma^{\text{eff}}_{\text{a}} = 2 \left[ 1 + \frac{\sin(\chi \rho_0)}{\chi \rho_0} \right] \sigma^{(3)}_{\text{eff}} \tag{8}
\]

In Equation (8), \( I \) is known as the interference factor. Further integration on \( \Omega_{\rho_s} \), allows us to obtain DDCS under the expression

\[
\sigma^{(2)} = \frac{d^2\sigma}{d\Omega_{\rho_e} d(k^2_e/2)} \approx 2 \left[ 1 + \frac{\sin(\chi \rho_0)}{\chi \rho_0} \right] \sigma^{(3)}_{\text{eff}} \tag{9}
\]
Moreover, SDCS, \( \sigma^{(1)} = d\sigma/d\Omega_e \), can be calculated integrating \( \sigma^{(2)} \) on the energy of the ejected electron.

3. Results and discussions

According to expression (8), constructive interferences will be found in TDCS for \( \chi \approx 0 \) values. It corresponds to soft collisions (for which \( K \approx 0 \) and \( k_e \approx 0 \)) or to electrons ejected in the Bethe region (\( \bar{k}_e \approx \bar{K} \)).

In the left side of Fig. 1, absolute experimental [26] and theoretical DDCS in a 8 keV \( e^- + H_2 \) collision are presented as a function of the polar emission angle at fixed emission energies of 9, 70 and 280 eV. Effective one-center double differential \( \sigma_{(2)}^{(2)} \) calculations, obtained after integration of \( \sigma_{(3)}^{(3)} \) on \( \Omega_e \) are included. Theoretical molecular results are in good agreement with measured data, particularly for low-energy ejected electrons (soft collisions) as well as for binary electrons (which correspond to the peaks in DDCS). In the right side of Fig. 1, the corresponding DDCS ratios obtained dividing experimental and theoretical DDCS by the effective one-center calculations are presented. Partial constructive interferences due to coherent emission from the molecular centers are evidenced by experiments and supported by theoretical results so well for soft collisions as for binary electrons, for which the ratios take values larger than 1.5 and smaller than 2.

**FIGURE 1.** (a), (b) and (c) show angular distributions of secondary electrons. Open circles, experimental data from [27]; solid lines, theoretical molecular cross sections; dashed lines, twice the calculations for effective one-center DDCS. (d), (e) and (f) show the corresponding ratios (for details see the text).

As the result of integration of DDCS on emitted electron energies ranging from 9 eV to 400 eV, experimental [28] and theoretical single differential cross sections (SDCS) are determined and presented in Fig. 2 together with the corresponding SDCS ratios. Again experimental and theoretical ratios are obtained by dividing the cross sections with effective one-center SDCS predictions. A reasonable agreement between theory and experiment is found for SDCS as well as for SDCR ratios.
It is very interesting to observe that fingerprints of Young-type interferences are preserved in SDCS. This behaviour can be explained by the fact that the contributions of low energy and binary encounter electrons dominate SDCS.

Finally, we must mention that for (e,2e) collisions an oscillatory behaviour in the angular asymmetry parameter

$$
\alpha(k_e, \theta_e) = \frac{\sigma^{(2)}(k_e, \theta_e) - \sigma^{(2)}(k_e, \pi - \theta_e)}{\sigma^{(2)}(k_e, \theta_e) + \sigma^{(2)}(k_e, \pi - \theta_e)}
$$

associated with coherent emission from the centers of the target has been obtained, as it was previously observed for ion impact [33]. In Equation (10), $\theta_e$ is the polar angle of the ejected electron. A comparison of the asymmetry parameter $\alpha(k_e, \theta_e)$ for electron and ion impact is given in reference [27].

![Diagram](image-url)

**FIGURE 2.** (a) Angular distribution of SCDS. Open circles, experimental data from [28]; solid line, theoretical molecular SDCS; dashed line, twice the calculations for effective one-center SDCS. In (b) the corresponding ratios are shown (for details see the text).

4. **Conclusions**

Young type interference effects are studied in (e,2e) collisions for impact of fast electron beams on H$_2$ targets. Angular distributions of ejected electrons both in double and single differential cross sections present evidence of partial constructive interferences for low-energy emitted electrons and for binary encounter ones, due to coherent emission from the proximities of the centers of the molecular target. The present theoretical description takes account of recent experimental data.
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