J. Phys. B: At. Mol. Opt. Phys. 36 (2003) L257-L264

PII: S0953-4075(03)65685-3

LETTER TO THE EDITOR

Interference effects in single ionization of molecular hydrogen by electron impact

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Received 4 July 2003 Published 14 August 2003 Online at stacks.iop.org/JPhysB/36/L257

Abstract

A recently developed molecular three-continuum approximation is employed to compute differential cross sections for the ionization of hydrogen molecules by electron impact. Within the framework of this approximation, the chosen final electronic wavefunction takes into account the molecular character of the target as well as the correlate motion between the aggregates in the final channel of the reaction. Fivefold-differential cross sections as a function of both the electron momenta in the final state and the molecular orientation are studied for different kinematical arrangements. Interference structures coming from the two-centre geometry of the molecule are predicted in this case. Integrated cross sections over all molecular orientations are also calculated. It is shown that interference patterns remain, even for this case.

During the last decades, a large number of theoretical and experimental studies have treated the (e, 2e) reactions with atomic targets, providing a general understanding of these processes (see, for example, [1]). In contrast, the research on ionization of molecules has advanced in a slower way due to some extra complications appearing in this case. For instance, on the experimental side, it has not been possible yet to prepare the molecular target in a particular rovibrational state. Another difficulty is imposed by the finite resolution of the electron beam. On the theoretical side, the molecular structure must be fully included in any realistic description of the process.

The main purpose of the present investigation is the study of interference effects which appear as a consequence of the two-centre character of the target, and may be related to the Young's two-slit experiment. This effect was theoretically predicted for ionization by photon impact on H₂ [2] (see also Walter and Briggs [3]), and recently measured for electron emission by fast multicharged ion impact on the same target [4, 5]. Different theoretical

models were introduced to describe these experiments [4, 6, 7]. In all of them, it was found that molecular transition amplitudes may be obtained as a coherent sum of *effective* atomic amplitudes. However, the interference behaviour has not been studied so far for the case of electrons as projectiles. So, the interest is focused here on the (e, 2e) reactions for H_2 targets.

(e, 2e) multiple-differential cross sections in the coplanar geometry are studied at high incident energies. In this arrangement, the internuclear vector as well as the incident, the ejected and the scattered electron momenta are all on the same plane. In particular, fivefold-differential cross sections (5DCSs) as a function of the scattered and the ejected electron momenta, and of the internuclear orientation, are computed in this work. As experiments have only been performed to date with low energy resolution, the ionization process may be considered as a pure electronic transition. Moreover, at the energies studied in this work, it can be assumed that the internuclear distance, ρ , remains unchanged during the collision [8–10]. Unfortunately, the orientation of the molecule in the available experiments cannot be resolved. However, the rapid development of multi-detection techniques [12] made possible recently the determination of the momenta of several particles resulting from the collision allowing thus to discern the molecular orientation [13, 14]. Therefore, these advancements on the experimental side indicate that a comparison between theory and experiments might be possible in the near future. This fact encourages the computation of 5DCSs in which most of the kinematical properties of the single ionization process are determined.

The recently introduced MBBK approximation (hereafter referred to as I) [15] is used in the calculations. In this model, the single ionization process is assumed to be produced in the proximity of one of the two molecular nuclei, while the *passive* electron completely screens the other molecular nucleus. Thus, the molecular amplitude is reduced to a coherent sum of two three-body amplitudes (where the three bodies are the ionized electron, the scattered electron, and one or the other molecular nucleus). Moreover, the correlated motion of these three unbound particles in the final channel is considered in the same way as was previously done for the atomic case [16]. The MBBK model was used with success to describe absolute experimental triple-differential cross sections for single ionization of H₂ [17] under the same conditions of interest in the present calculation [15].

Following I, the transition matrix element t_{fi}^{e} may be expressed as a sum of direct and indirect terms. The direct term describes ionization of the *active* electron from one of the two molecular centres (denoted by j) by means of the interaction between the projectile with this electron and with the centre j. The indirect term may be considered as giving ionization of the *active* electron from the same nucleus j but now through the interaction of the projectile with the *passive* electron and with the other centre from which ionization is not produced. Of course, electrons are shared by both nuclei in the molecule but matrix elements admit this interpretation. Moreover, it is also stated in I that, for the geometries considered here, the indirect term can be neglected in the computation of the differential cross sections. Accordingly, the square modulus of the transition matrix element (see equation (18) in I) can be written in the convenient form

$$|t_{\rm fi}^{\rm e}(\rho_0)|^2 \simeq 2[1 + \cos(\chi \cdot \rho_0)]|t_{\rm fi}^{\rm A}|^2 \tag{1}$$

where $\chi = \mathbf{k}_e - \mathbf{K}$, and $\mathbf{K} = \mathbf{k}_i - \mathbf{k}_s$ is the momentum transferred to the ionized electron. The emitted electron is ejected with momentum \mathbf{k}_e into the differential solid angle Ω_e with respect to the incidence direction defined by the initial momentum of the projectile, \mathbf{k}_i . The projectile is scattered with momentum \mathbf{k}_s into the solid angle Ω_s . ρ_0 denotes the equilibrium internuclear vector of the molecular target. As only highly asymmetric arrangements are considered, exchange effects are neglected. In equation (1), the function $t_{\rm fl}^{\rm A}$ plays the role of a transition matrix element corresponding to effective hydrogen-like atoms placed at the position of each one of the molecular nuclei. The effective atomic transition element given by equation (1) is obtained by using the variational charge and the molecular binding energy corresponding to a Heitler–London type wavefunction [18].

Consequently, the 5DCS reads

$$\sigma^{(5)} = \frac{\mathrm{d}^{5}\sigma}{\mathrm{d}\Omega_{\rho}\,\mathrm{d}\Omega_{\mathrm{e}}\,\mathrm{d}\Omega_{\mathrm{s}}\,\mathrm{d}(\mathrm{k}_{\mathrm{e}}^{2}/2)} \cong 2[1 + \cos(\chi \cdot \rho_{0})]\sigma_{\mathrm{A}}^{(3)} \tag{2}$$

where $\sigma_A^{(3)}$ represents a one-centre differential cross section computed with the transition matrix element t_{fi}^A . In order to take into account the two electrons of the H₂ molecule, an extra factor of two is included in expression (2). The interference pattern due to coherent emission from both molecular centres appears explicitly in equation (2).

Triple-differential cross sections corresponding to a coplanar geometry are also obtained averaging over all possible molecular orientations, i.e.,

$$\sigma^{(3)} = \frac{\mathrm{d}^3 \sigma}{\mathrm{d}\Omega_{\mathrm{e}} \,\mathrm{d}\Omega_{\mathrm{s}} \,\mathrm{d}(\mathrm{k}_{\mathrm{e}}^2/2)} \cong 2 \left[1 + \frac{\sin(\chi\rho_0)}{\chi\rho_0} \right] \sigma_{\mathrm{A}}^{(3)}. \tag{3}$$

Therefore, interference patterns remain even for this case.

It is worth emphasizing that interference structures might also be found by choosing a first-order final wavefunction, where correlations of the projectile with the *active* electron and with the molecular nuclei are neglected. In fact, only the advantages of a two-effective-centre description and the negligible character of the indirect terms in the transition matrix element are required to obtain similar expressions to those appearing in equations (2) and (3).

Differential cross sections for electron-impact ionization of hydrogen molecules are computed for coplanar geometry. In figures 1(a)-(d), MBBK 5DCSs as a function of the ejection angle θ_e are calculated for some fixed molecular orientations ($\theta_o = 0^\circ, 30^\circ, 60^\circ, and$ 90°). The incident and emission energies are $E_i = 4087 \text{ eV}$ and $E_e = 150 \text{ eV}$, respectively, whereas the scattering angle is fixed at $\theta_s = 1^\circ$. Interference patterns coming from the electron emission from the different scattering centres in the molecule are observed. Particular regions in which the electron emission is more likely to happen are revealed in the present figures. Thus, 5DCS profiles may be considered, in this case, as composed mainly by two lobes in the ρ_0 direction and two others appearing in the normal direction, showing a strong dependence on the internuclear vector. As the molecular orientation changes, oscillations seem to rotate in a reference frame fixed to the molecule. This is in accordance with the recently presented ionization cross sections for ion impact on H_2 [11]. However, in that case, only an increase of the electron ejection in the direction of the molecule is observed as the molecule turns. As stated in [11], the additional integration over the transverse momentum performed there tends to blur the interference patterns in the molecular angular distributions. In the present case, the observed behaviour in the 5DCS may be exclusively attributed to the interference factor containing the function $\cos(\chi \cdot \rho_0)$ (see equation (2)), which is also represented in the figures after being properly scaled. In this way, it is easy to see that zeros in the 5DCS correspond to zeros in the interference factor. Of course, the molecular emission pattern comes from the superposition of the interference factor and the TDCS corresponding to two effective atomic centres (the latter is also shown in the figures in order to highlight the behaviour of both contributions). At the energies considered here, effective atomic results, which do not depend on the molecular orientation, exhibit the usual two-peaked structure: the binary peak located around the momentum transfer direction and the recoil one that appears in the opposite direction. Hence, the molecular cross section profiles may be understood as the differential cross sections corresponding to one-centre contributions modulated by the interference factor.

In figures 2 and 3, molecular 5DCSs as a function of the emitted electron angle are presented for different ejection energies, in the particular cases in which the molecule is



Figure 1. 5DCSs as a function of the ejection angle θ_e for different molecular orientations. (a) $\theta_{\rho} = 0^{\circ}$; (b) $\theta_{\rho} = 30^{\circ}$; (c) $\theta_{\rho} = 60^{\circ}$; $\theta_{\rho} = 90^{\circ}$. Impact energy, $E_{i} = 4087 \text{ eV}$; emission energy, $E_{e} = 150 \text{ eV}$; scattering angle, $\theta_{s} = 1^{\circ}$. Full curve: MBBK results. Dashed curve: two effective H atoms. Dotted curve: interference factor (from equation (2)).

aligned parallel and perpendicular to the incident beam, respectively. In figures 2(a)-(d), the incident energy is $E_i = 4087$ eV, the scattering angle is fixed at $\theta_s = 1^\circ$, and the ejection energy takes the values $E_e = 20, 57, 100$, and 500 eV, respectively. At the lowest ejection energy considered in the figures, 5DCS results present, qualitatively, a similar behaviour to the ones corresponding to differential cross sections for two effective atomic targets. As will be seen below, under the energetic conditions studied in figure 2(a), the ejected electron is not able to resolve the two-nucleus structure of the molecule and, therefore, molecular results present no observable interference effects for this case. However, as E_e increases, extra features in the 5DCS profiles appear. At the particular ejection energy value ($E_e = 57 \text{ eV}$) considered in figure 2(b), a zero at $\theta_e = 180^\circ$ is observed in the molecular spectrum. This fact may be interpreted as clear evidence of the interference phenomenon, because the atomic pattern

10[°]

270



Figure 2. 5DCS as a function of the ejection angle, θ_e , for H₂ molecules aligned parallel to the incident beam. Impact energy, $E_i = 4087 \text{ eV}$; scattering angle, $\theta_s = 1^\circ$; ejection energy, (a) $E_e = 20 \text{ eV}$; (b) $E_e = 57 \text{ eV}$; (c) $E_e = 100 \text{ eV}$, and (d) $E_e = 500 \text{ eV}$. Same notation as figure 1.

270

10

emission is broken by a new structure appearing at $\theta_e = 180^\circ$. Ejection energies (equivalently, ejection momenta) at which these new structures are observed can be determined. For instance, when the molecule is aligned along the incidence direction (only the component of χ in this direction contributes to the argument of the cosine function) and $\theta_e = 180^\circ$, it is easy to show from equation (2) that interference structures appear for ejected electron momenta larger than $k_e = \frac{\pi}{\rho_0} - K_{\parallel}$, K_{\parallel} being the longitudinal component of the transfer momentum. For the cases under study, K_{\parallel} is small, and then the criteria discussed above imply that the de Broglie wavelength associated with the ejected electron is smaller than twice the internuclear distance, i.e., the ejected electron begins to discern the two-centre geometry of the molecule. This result can be directly related to the ones corresponding to the photo-ionization case [3] in which the interference factor depends only on the product $\mathbf{k}_e \cdot \rho_0$. It is not surprising that analogies between electron impact ionization and photo-ionization may be expected for collisions with





Figure 3. The same as figure 2, but considering the molecule oriented in the orthogonal direction to the incident beam. The ejection energy is (a) $E_e = 20 \text{ eV}$, (b) $E_e = 51.2 \text{ eV}$, (c) $E_e = 100 \text{ eV}$, and (d) $E_e = 500 \text{ eV}$.

sufficiently small momentum transferred to the ejected electron. In figures 2(c) and (d), 5DCS results for increasing ejection energies are illustrated. In both cases, it is evident that the number of nodes increases as the emission energy increases. For $E_e = 100 \text{ eV}$ four nodes are predicted, at $\theta_e \simeq 21^\circ$, 136°, 224°, and 339°, whereas for the case of $E_e = 500 \text{ eV}$ six nodes are observed, at $\theta_e \simeq 56^\circ$, 100°, 156°, 204°, 260°, and 304°.

In figures 3(a)–(d), the molecule is aligned along the orthogonal direction to the incoming beam. The incident energy is $E_i = 4087 \text{ eV}$, the scattering angle is fixed at $\theta_s = 1^\circ$, and the ejection energy $E_e = 20$, 51.2, 100, and 500 eV. At the lowest emission energy, interference effects are not observed, and 5DCS profiles do not differ too much from those for effective H atoms. In this case, the involved electron wavelengths are larger than the internuclear separation, and, as a consequence, the molecule is seen as a whole body. However, as the ejection energy increases, the formation of a node at $\theta_e = 90^\circ$ is observed in the molecular



Figure 4. Normalized MBBK TDCS results as a function of both the ejection energy and angle θ_e . The incident energy is $E_i = 4087 \text{ eV}$, and the scattering angle is (a) $\theta_s = 1^\circ$ and (b) $\theta_s = 8^\circ$.

angular distribution. As a matter of fact, the ejection energy, $E_e = 51.2 \text{ eV}$ was chosen in figure 3(b) in order to obtain a zero at 90°. When the molecule is oriented in the perpendicular direction, the argument of the cosine function depends exclusively on the normal component of χ . In the same way as was previously done, it is easy to show from equation (2) that interference occurs at particular electron momenta satisfying $k_e \ge \frac{\pi}{\rho_0} + K_{\perp}$, with K_{\perp} the transverse transfer momentum. As can be seen in figures 3(c) and (d), the number of nodes increases as the ejection energy increases.

Finally, triple-differential cross sections for electron-impact ionization of H_2 are studied. According to equation (3), oscillations are expected even after averaging over all possible molecular orientations. However, these interference effects cannot be clearly seen directly from TDCSs. On one hand, molecular spectra decrease strongly, by several orders of magnitude, at increasing ejection energies, washing out small effects due to the two-centre coherent emission. On the other hand, the argument of the interference factor as a function of the emission angle varies significantly only in the Bethe region, where all the momentum is transferred to the ejected electron. Then, it is expected that the interference factor only influences the magnitude of the characteristic peaks appearing in molecular angular distributions corresponding to asymmetric geometries. In consequence, instead of absolute molecular results, theoretical ratios between molecular TDCSs and twice those corresponding to effective H atoms are plotted in figure 4 as a function of both the ejection energy and angle θ_e . The incident energy is $E_i = 4087 \text{ eV}$ and the scattering angles considered are $\theta_s = 1^\circ$ and 8° . Oscillations are clearly seen outside the binary peak region. In particular, normalized TDCSs as a function of the ejection energy show marked oscillations depending on the electron ejection angle. Moreover, an increase in the frequency of the oscillations is found as the backward emission region is reached. The same behaviour was observed for ions as projectiles [6]. Maximum interference values occur at particular emission angles of about 270° for which the Bethe condition is satisfied.

To sum up, it has been shown that the angular distribution of the ejected electrons exhibits interference structures arising from the coherent emission from the two molecular centres. Moreover, it has been found that the oscillatory patterns depend markedly on the molecular orientation, in the same way as was recently obtained for heavy ion impact [11]. In addition, it has also been shown that interference phenomena appear even in the integrated differential cross sections.

This work was partially supported by the French–Argentinean ECOS-Sud programme (No A98E06). CRS, OAF, and RDR also acknowledge support from the Agencia Nacional de Promoción Científica y Tecnológica (BID 802/OC-AR PICT No 03-04262) and the Consejo Nacional de Investigaciones Científicas y Técnicas de la República Argentina.

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