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(e,2e) ionization of helium and the hydrogen molecule: signature of two-centre interference effects

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Abstract
Relative (e,2e) triply differential cross sections (TDCS) are measured for the ionization of the helium atom and the hydrogen molecule in coplanar asymmetric geometry at a scattered electron energy of 500 eV and ejected electron energies of 205, 74 and 37 eV. The He experimental results are found to be in very good agreement with convergent close-coupling calculations (CCC). The H_2 experimental results are compared with two state-of-the-art available theoretical models for treating differential electron impact ionization of molecules. Both models yield an overall good agreement with experiments, except for some intensity deviations in the recoil region. Similar (e,2e) works were recently published on H_2 with contrasted conclusions to the hypothesis that the two H nuclei could give rise to an interference pattern in the TDCS structure. Murray (2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 1999) found no evidence for such an effect, whereas Milne-Brownlie *et al* (2006 *Phys. Rev. Lett.* **96** 233201) reported its *indirect* observation. In this work, based on a *direct* comparison between experimental results for He and H_2, we observe an oscillatory pattern due to these interference effects, and for the first time the destructive or constructive character of the interference is observed, depending on the de Broglie wavelength of the ejected electron wave. The experimental finding is in good agreement with the theoretical prediction by Stia *et al* (2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** L257).

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Over the past two decades, the field of electron impact single ionization (SI) of simple one- and two-electron atoms (H and He) has reached a degree of maturity such that sophisticated theoretical models (e.g. the exterior complex scaling ECS [1] or the convergent close-coupling CCC [2] methods) can now accurately predict the behaviour of the triple, *fully* differential cross section (TDCS) for wide ranges of the kinematical parameters (energies and vector momenta). Thus, the interest has now moved to the study of more complex, multi-electron atomic or molecular targets where the situation is by far more challenging. In particular, a renewed interest has emerged for the (e,2e) studies of the

dynamics of molecular ionization, both experimentally [3–7] and theoretically [8–14]. This has led to the development of theoretical approaches meant to deal with the description of the molecular ionization processes, the most sophisticated ones being probably the first Born approximation (FBA)–two centre continuum (TCC) approximation with correct boundary conditions in the entrance and exit channels [9] and the molecular three-body distorted wave approximation (M3DW) coupled with an orientation-averaged molecular orbital approximation (OAMO) [13, 14]. During the course of these developments, considerable interest has been raised by the possibility of observing, in the case of diatomic molecules, quantum mechanical interference effects resulting from the coherent superposition of the scattered waves from the two atomic centres [15]. These Young-type interference effects have been considered for many years in the photon ionization of H_2 [16, 17]. They were recently theoretically predicted by Stia *et al* [18] and by Gao *et al* [19] for electron impact ionization of molecular hydrogen and molecular nitrogen, respectively. Their observation was reported in *double* differential cross section (DDCS) measurements for heavy, multicharged ion impact on H_2 [20–22] and for fast (2.4 keV) electron impact on D_2 [23]. The question of their observation in *fully* differential cross sections was recently addressed by two groups in (e,2e) TDCS measurements (we note a speculative mention made by Jung *et al* [24] to explain the low coincidence rates in the recoil peak as being due to destructive interferences). First, at Manchester University, Murray [5] found no evidence of such effects in 'low' energy (<100 eV) electron impact ionization of H_2. Subsequently, Murray *et al* [6] reported a similar investigation on the $N_2(3\sigma_g)$ state, where they discuss the possibility of the existence of an interference peak in the vicinity of the backscattering angles, as predicted in [19]. However, they could not definitely prove it as the experimental data did not cover the angular range of the expected peak. Later on, Milne-Brownlie *et al* [3] at Griffith University reported the observation of Young-type interference effects in (e,2e) ionization of H_2 at an intermediate incident energy of 250 eV. The observable result is partial intensity suppression in the recoil peak compared with the binary one. The contrasted conclusions from these works on such an important matter called for and warranted a new investigation, in order to contribute to the understanding of such fundamental phenomenon.

Thus, in order to resolve the above-mentioned contrast, we have undertaken a new set of measurements similar (though not fully identical) to the case studied by Milne-Brownlie *et al* [3]. These authors used coplanar asymmetric geometry with an electron impact energy $E_0 = 250$ eV, ejected electron energies $E_b = 10$, 20 and 50 eV and a scattering angle for the fast electron $\theta_a = -15^\circ$. Under these kinematics, the interference effect is predicted to always result in a suppression of the recoil intensity with respect to the binary one. In contrast, we use the higher energies, $E_0 \sim 600$ to 700 eV, the ejected electron energies $E_b = 37$, 74 and 205 eV and the smaller scattering angle $\theta_a = -6^\circ$. Under these kinematics, the effect of the interference process is predicted (see below) to reduce the relative intensity of the recoil peak at the two lowest ejected energies and to increase it at the largest one. Hence, the new data allow a more stringent test of the theoretical prediction. Moreover, though we use in the present work, as was done in [3], comparison of the H_2 and the He TDCS, we will see below that our approach does not rely at all on any calculated TDCS neither for H_2 nor for He, as was the case in [3], but it solely relies on the ratio of our measured triple differential cross sections for both targets. Hence, the new data allow a more *direct* test of the theoretical prediction.

2. Experiment

The experimental set-up currently in use in Orsay, whose main characteristic is the combination of three high-efficiency, multi-angle toroidal electrostatic energy analysers, has been described in detail elsewhere [25]. The experimental procedure is identical to that reported in [7]. Briefly, an incident electron beam collides with the gas jet formed at the collision centre. A coplanar geometry is used, where all electrons are observed in the collision plane defined by the incident and scattered momentum vectors $\mathbf{k_0}$ and $\mathbf{k_a}$, respectively. The 'slow' ejected electrons (designated with an index 'b' for convenience) are multi-angle analysed in a double toroidal analyser, with the energies $E_b = 205$, 74 and 37 eV and over the angular ranges $\theta_b = 20$–160° and 200–340°, where 0° is defined by the incident beam direction. In the off-line analysis, the total θ_b angular range is divided into sectors of width $\Delta\theta_b = 5^\circ$. The 'fast', forward-scattered electron (indexed 'a') is collected by the third toroidal analyser [25] at the scattered energy $E_a = 500$ eV. In the present work, the a-electron is simultaneously observed at two symmetrical angles, $\theta_a = +(6^\circ \pm 0.25^\circ)$ and $-(6^\circ \pm 0.25^\circ)$, as set by input slits at the entrance to the electrostatic lenses associated with the toroidal analyser. The incident energy (E_0) is consequently adjusted to fulfil the energy conservation requirement for the target under study, $E_0 = E_a + E_b + \mathrm{IP}$, where IP is its ionization potential (24.6 eV for He and 15.5 eV for H_2). As an example, for the helium target and for the case $E_b = 74$ eV, the corresponding momentum transfer value is $K = 0.88 \pm 0.02$ au and the momentum transfer direction is $\theta_K = 46^\circ \pm 1^\circ$.

Due to the low coincidence rate, especially at the highest ejection energy, the spectrometer was operated at the reduced coincidence energy resolution [26], $\Delta E_{\mathrm{coin}} \sim \pm 2.5$ eV. This value did not allow resolving the final ionic state of the hydrogen molecule.

Finally, we note that the He (e,2e) experiments were performed *under exactly the same experimental conditions* as those used for H_2 (except for a slight change in incident energy, due to the difference in their IP), so that we can readily determine the ratio of the measured TDCS for both targets.

3. Results and discussion

The discussion of the results is organized in two parts. First, the measured angular distributions for He and H_2 are compared with calculated results from state-of-the-art theoretical models. The He results are used to validate our procedure, while

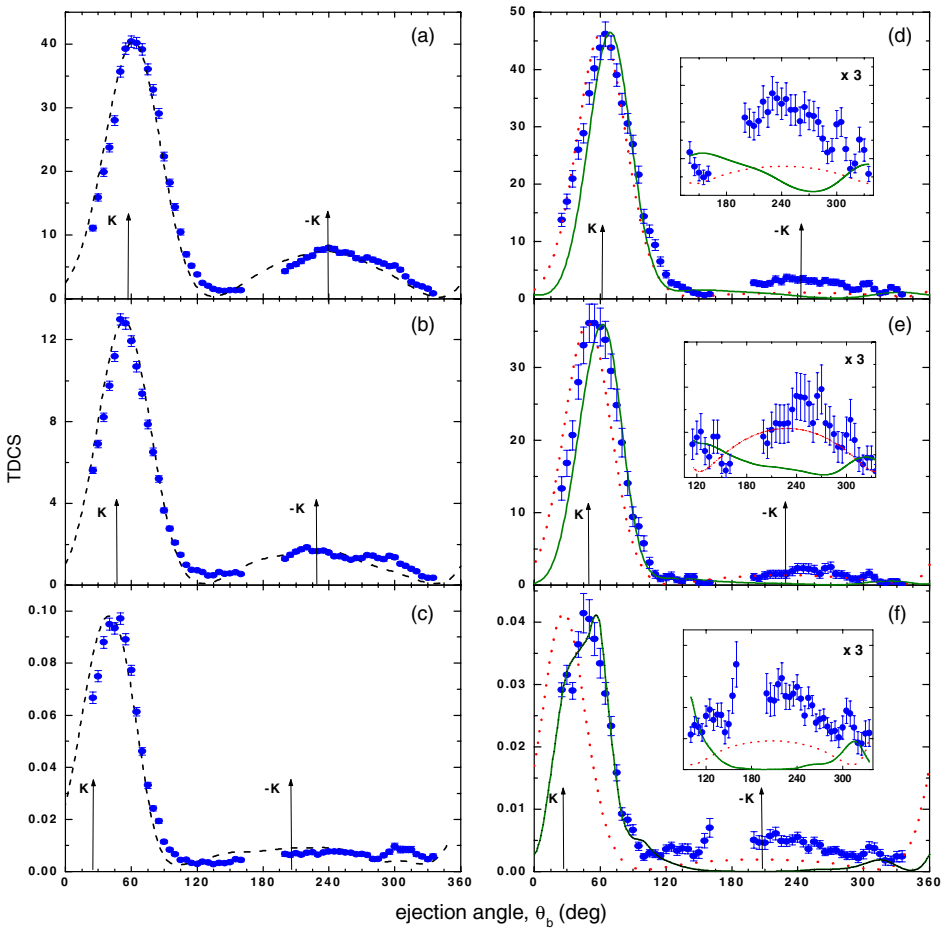

Figure 1. (e,2e) TDCS for ionization of He (left column) and H_2 (right column), plotted versus ejection angle θ_b, at a fixed scattering angle, $\theta_a = -6^\circ$ and the fixed scattering energy, $E_a = 500$ eV. Panels (a) and (d) $E_b = 37$ eV; (b) and (e) $E_b = 74$ eV; (c) and (f) $E_b = 205$ eV. The incident energy (E_0) is consequently adjusted to fulfil the energy conservation. For He, the dashed line represents the results of the CCC calculations. For H_2, the dotted and full lines represent the theoretical results from the FBA–TCC and the M3DW–OAMO models, respectively. Solid circles: experimental data, with one standard deviation statistical error bars. The vertical arrows indicate the momentum transfer direction and its opposite. The insets in the H_2 results represent a zoom on the low intensity recoil region to facilitate comparison. The relative experimental data have been normalized for the best visual agreement with theory. The absolute scale shown is that of the CCC calculations for He and that of the FBA–TCC for H_2, both in 10^{-2} atomic units. The M3DW–OAMO results have been multiplied by 2.5 in (d), 2.8 in (e) and 6.7 in (f).

the molecular results allow to pin-point successes (at the binary peak region) and deficiencies (at the recoil peak region) of the models. Second, the behaviour of the ratio of the *measured* TDCS for both targets is confronted to the theoretical prediction in [3] which allows interpreting this behaviour in terms of molecular two-center interference effects.

3.1. Angular distributions of the TDCS

The experimental results for the TDCS distribution for ionization of He are shown in figures 1(a)–(c) for the three investigated ejected electron energies, whereas figures 1(d)–(f) show the similar results for ionization of H_2. The data are compared with calculated results obtained using the convergent close coupling (CCC) method [2] for the helium atom, and using two state-of-the-art available approaches for the molecular target. Note that for each angular distribution, the relative experimental data have been independently normalized to the absolute scale given by theory, as explained in the figure caption.

The CCC calculations are performed separately for each incident energy. Due to the vastly different energies of the outgoing electrons and large incident energy on the single ground state, exchange needs not be included. For the kinematics considered, the Born approximation is reasonably accurate with a distorted wave Born approximation yielding further improvement. In such cases the close-coupling formalism converges with increasing basis sizes, N_l, relatively rapidly and we simply need to ensure sufficient number of angular momenta l for the ejected electron and L for the scattered electron. We find that $l \leqslant 5$ is sufficient for the smaller ejected energies and $l \leqslant 7$ required for the 205 eV case. With increasing incident energy we require larger L, which ranges from 30 to 50. Lastly, the CCC calculations presented were performed in the frozen-core approximation which keeps one of the He electrons fixed, as a He^+ 1s

orbital. We checked that relaxing this approximation by adding more configurations, significantly improved the quality of the ground state, but had no substantial effect on the ionization cross sections presented.

The first theoretical model used for H_2 is based on a first Born approximation (FBA) in which the two-centre continuum (TCC) approximation with correct boundary conditions in the entrance and exit channels [10] is applied. Special care is taken in the description of the slow ejected electron in the field of the residual diatomic ion by a two-center Coulomb function, which has given [27] excellent results compared to those obtained by the exact solutions of the two-centre Schrödinger equation in prolate spheroidal coordinates [28]. Here, the relatively fast incident and the scattered electrons are described by plane waves. For the initial and final state bound electrons the wavefunctions given in [9] are used. Owing to the high incident energy and the large difference in energies of the outgoing electrons, exchange effects between these electrons are not expected to be significant and hence were not included.

The second model used for H_2 is the molecular three-body distorted wave (M3DW) approximation coupled with an orientation-averaged molecular orbital (OAMO) approximation [13, 14]. The M3DW–OAMO is a two-centre approach in which all three continuum electron wavefunctions are represented by distorted waves calculated on a spherically symmetric potential obtained from the Hartree–Fock charge distribution for H_2 averaged over all molecular orientations. For the incoming electron, the neutral charge density is used and for the two final state electrons the ionic charge density is used. The nuclear contribution to the distorting potential is equivalent to the potential of a thin metal spherical shell of radius $0.7a_0$ containing a total charge of 2. The polarization and correlation potential of Perdew and Zunger [29] and the Furness–McCarthy [30] exchange-distortion potential are added to the static Hartree–Fock distorting potential. The electron–electron Coulomb factor is included in the final state wavefunction which means that the final state post-collision-interaction (PCI) between the two continuum electrons is included to all orders of perturbation theory. In the OAMO approximation, an orientation averaged molecular orbital is used for the initial-state wavefunction. The OAMO approximation has been shown to be valid for ionization of H_2 as long as the momentum transferred to the residual ion is less than unity [31]. Here again, exchange was not included due to the large energy difference of the outgoing electrons.

We first comment on the He (e,2e) results in figures 1(a)–(c). It is nowadays a well established fact that at high and intermediate impact energy the ionization process is very well described by the CCC method. Indeed, the agreement between experiments and theory is very good at the three considered energies, both in the shape of the distributions and in the position of the binary lobes. The small deviations seen in the recoil region might at least partly be of the statistical nature (the count rates being there rather small), reflecting the difficulty involved in performing measurements of processes characterized by low cross sections. The CCC results show a shift of the binary lobe of some 10° from the momentum transfer direction (θ_K), and so do our data, though at the highest energy the CCC theory yields a slightly smaller shift than experiments. These observations are consistent with known trends for He [32, 33], where peak shifts away from θ_K direction are to be expected whenever the first Born approximation is not sufficiently accurate. We thus believe that the experiments are free from any significant error or artefact. Our experimental procedure can thus be applied with good confidence to the other target studied here, since the H_2 data were obtained *under exactly the same experimental conditions* as those used for He.

For H_2, figures 1(d)–(f), the comparison between experiments and theory is less satisfactory, with a somehow better agreement reached by the FBA–TCC model with respect to the M3DW–OAMO at the two lowest energies, while the M3DW–OAMO is doing better in describing the binary peak at the highest energy. The shape of the binary lobes is essentially correctly reproduced by both model calculations. For the highest ejected-electron energy (figure 1(f)), we note that the M3DW–OAMO predicts a shoulder on the low angle side of the binary peak, which might possibly be also present in the experimental data though the statistics do not allow to be more affirmative. The origin of this shoulder was found to be mostly due to final state elastic scattering of the projectile electron from the target. However, both models predict a too small recoil intensity (except for the FBA-TCC at 74 eV), or even the absence of a recoil lobe in the M3DW–OAMO results. Since the recoil peak corresponds to the largest momentum transferred to the ion, the incorrect behaviour of the M3DW–OAMO in the region of the recoil peak most likely stems from the breakdown of the OAMO approximation, as also discussed in [7]. Moreover, as expected FBA–TCC predicts a binary lobe aligned with the momentum transfer direction, θ_K, being a first Born model. The M3DW–OAMO includes final state PCI between the two continuum electrons, whose effect is to rotate the lobes in the backward direction. However, the effect seems to be overestimated, the M3DW–OAMO shift of the binary lobe from the **K**-direction being larger than the about 10° measured shift (the latter is similar to the observed and the CCC-calculated shift for He in figures 1(a)–(c)).

We note that both theoretical models (TCC and M3DW) were recently found [7] to be less successful in describing (e,2e) experiments on N_2 under very similar kinematics as the present ones, a failure which thus must be attributed to the difficulty of describing the more complex nitrogen molecule. On the other hand, the TCC model behaved very well [9] in describing high energy (∼4.1 keV) (e,2e) processes on H_2 [34], so that its deficiencies here must be attributed at least in part to the different impact energy regime (∼600 eV in this work) where non-first-Born effects are expected to start playing a role.

3.2. Interference effect

At first glance, the TDCS distributions obtained for He and H_2 (figure 1) may look very similar as far as the shape of the lobes is concerned. However, a closer inspection shows that the recoil peak in H_2 is substantially smaller than that in He, relative to the height of the binary peak, for the cases E_b = 74 and 37 eV, whereas the recoil peak in H_2 is larger than

that in He for the case at $E_b = 205$ eV. This recoil intensity suppression on the one side and enhancement on the other side in the molecular case is attributed to Young-type interference effects, and is the subject of the discussion in this section.

In their theoretical investigation of the (e,2e) single ionization of H_2, Stia *et al* [18] (see also an earlier derivation by Dal Cappello *et al* [35]), have shown that the angular distribution of the ejected electrons exhibits interference structures arising from the coherent emission from the two molecular centres. Moreover, they predicted that these interference structures should be observed even in the TDCS distribution from non-oriented molecules (as is the case in the present study). They showed that, provided a two-effective-centre description is used, the TDCS distribution for molecular hydrogen, $\sigma_{e,2e}(H_2)$, can be expressed as twice the TDCS distribution of the one-centre atomic hydrogen, $\sigma_{e,2e}(H)$, modulated by an interference factor, I, that is

$$\sigma_{e,2e}(H_2) = 2^*\sigma_{e,2e}(H)^* I$$

where I is given by

$$I = 1 + \frac{\sin(q\rho)}{q\rho}. \qquad (1)$$

Here, $\mathbf{q}$ is the momentum imparted to the recoiling ion, $\mathbf{q} = \mathbf{k_0} - \mathbf{k_a} - \mathbf{k_b}$ and ρ is the equilibrium internuclear distance of the H_2 molecule, $\rho = 1.4$ au [36]. In other words, the ratio $\sigma_{e,2e}(H_2)/2^*\sigma_{e,2e}(H)$ should display the same oscillatory behaviour as the I factor. This is the basic idea of the present study. However, instead of using twice the atomic hydrogen cross section, we have used the He cross section (an equivalent two-electron-single centre atom), hence comparing the ratio $R = \sigma_{e,2e}(H_2)/\sigma_{e,2e}(He)$ to I. We emphasize that this procedure does not rely on any theoretical calculations neither for He nor for H_2, as was the case in [3]. The whole argument hinges on the behaviour of the recoil peak relative to the binary one and does not need any support from the theoretical calculations presented above, which anyway fail to properly predict the recoil intensity.

The interference factor, I, is plotted in figure 2 as a function of the ejected electron angle, θ_b, for the kinematics of the present experiments. As expected, the factor I has an oscillatory behaviour, passing through a maximum at θ_b angles in the vicinity of 50–60°, that is close to the maximum of the binary peak as observed in figure 1. Our cross section measurements are obtained on a relative scale, and so is their ratio, R, to which I should be compared. Hence, we arbitrarily normalized the I values to unity in the region of the binary peak. We note that, for the ejected electron energies $E_b =$ 37 and 74 eV, the I factor passes through a minimum in the angular range where the recoil peak is at maximum, i.e. at θ_b angles in the vicinity of 230–240°. The secondary maximum observed in this angular range for the 37 eV case is too small to be meaningful for the present discussion. But a remarkable fact is that for the ejected electron energy $E_b = 205$ eV, the I factor displays a maximum in the recoil region instead of a minimum. Consequently, we might expect the recoil peak to be diminished in H_2 (with respect to that of the He atom) for the two lowest energies, and in contrast to be enhanced in the case of the highest E_b value. This is exactly the analogue of

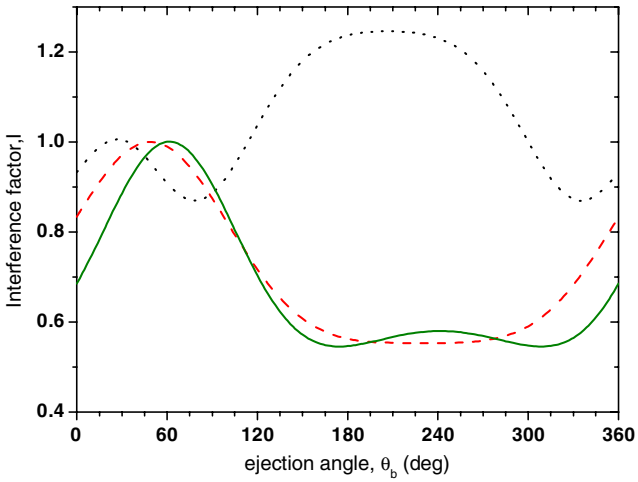

Figure 2. Interference factor, I, predicted by equation (1), plotted versus ejection angle, θ_b, at the ejected electron energies $E_b = 37$ eV (full line), $E_b = 74$ eV (dashed line) and $E_b = 205$ eV (dotted line). The I vaues are arbitrarily normalized to unity in the region of the binary peak.

Young-type double slit interference effects, which might be either destructive or constructive at a given scattering angle, depending on the ratio λ/ρ, where λ is the light (here ejected electron) de Broglie wavelength and ρ is the distance between the two slits (here the two nuclei). We note that for the three considered energies, the ejected electron wavelength λ varies between 1.6 and 3.8 au, that is λ is close to the 'inter-slit distance' $\rho = 1.4$ au, which is the condition of the existence of interference effects

In figures 3(a)–(c), the experimental ratio $R = \sigma_{e,2e}(H_2)/\sigma_{e,2e}(He)$ is plotted as a function of the ejected electron angle, θ_b, for the three ejection energies considered in the present experiments. Comparison is made with the interference factor, I. Qualitative good agreement is seen in the three cases between R and I, in spite of the large error bars due to the fact that, in certain angular ranges, we are taking the ratio of two small quantities, and considering the approximations made in the Stia *et al*'s model. Figures 3(a) and (b) clearly show a suppression of the recoil peak intensity with respect to the binary one, while figure 3(c), where $E_b =$ 205 eV, displays its prominent enhancement. In figures 3(b) and (c) the effect is more pronounced in the experiments than in the theoretical prediction, but the effect is qualitatively the same. The reasonably good agreement of the experimental results with the predictions of Stia *et al* [18] suggests that the present observations can be ascribed to the destructive ($E_b =$ 37, 74 eV) or constructive ($E_b = 205$ eV) interference effects arising from the two-centre nature of H_2. We note that, to our best knowledge, this is the first time that both the destructive and constructive characters of the interference process are simultaneously observed in the same (e,2e) experiments.

It might be tempting to attribute the peak observed at about 60° in the experimental ratio of figure 3(b) to the broader initial state momentum distribution in He with respect to H_2 which results in a larger width of the He binary peak. However, such interpretation implies that a similar peak should be observed

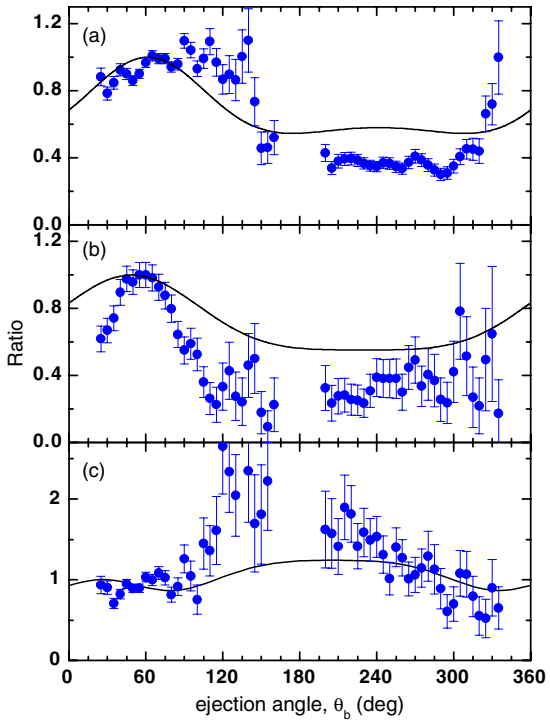

Figure 3. Solid circles: the experimental ratio, $\sigma_{e,2e}(H_2)/\sigma_{e,2e}(He)$, of the (e,2e) TDCS for ionization of H_2 relative to that of He, plotted versus the ejection angle θ_b at (a) $E_b = 37$ eV; (b) $E_b = 74$ eV; (c) $E_b = 205$ eV. The full lines represent the predicted interference factor of figure 2.

at the three considered energies, which is not the case. Rather, our measured peak is in qualitative agreement with the Stia *et al*'s predicted interference factor, which does not depend on the target momentum distribution. We thus believe that the origin of this peak is mostly of a geometrical/kinematical nature, as is the factor *I*.

The reduction of the recoil peak relative to the binary one observed in figures 3(a) and (b) might have other plausible explanations. For example, the recoil peak is known to be very sensitive to the properties of the initial target state. Since it involves elastic backscattering of the ejected electron from the target core, the more diffuse nuclear charge in H_2 compared to He might result in a reduction of the recoil peak. Alternatively, the interaction between the faster electron and the target core might also contribute to a reduction of the molecular recoil peak. However, our argument above is based on the simultaneous observation of a reduction *and* an enhancement of the recoil peak, and we think that there is no way that a diffuse nuclear charge or any alternative argument might yield to an enhanced recoil peak for H_2. It would certainly be interesting to investigate the role of the different electron charge distributions or nuclear charge distributions, but this would not be a meaningful task here since the molecular models considered do not even get the recoil peak correctly described.

Similarly, our analysis relies, as was done in [3], on the assumption that the interference term, *I*, can be compared to the ratio of the TDCS for H_2 to that for He instead of twice that of atomic hydrogen. Although this assumption might be questionable, the good agreement achieved with the Stia *et al* [18] ratio factor provides further support for the validity of this approach.

4. Conclusion

Relative (e,2e) TDCS for ionization of the H_2 molecule at ~600 eV incident energy are reported. Similar data obtained for the ionization of He are found to be in good agreement with the well-established CCC results, thus providing a validation of our experimental procedure. The H_2 results are compared with the most elaborate available molecular calculations. Reasonable agreement is found between measured and calculated distributions in the binary region. However, clear discrepancies are observed between theories and experiments, in particular, (i) for the position of the binary lobes, which calls for a proper treatment of second-order effects in the theories and (ii) for the intensity distribution in the recoil region which calls for a better modelling of the interaction with the nucleus. These discrepancies demonstrate the need for further development of the theoretical models in order to accurately describe the ionization process, even for the simplest molecular target, H_2.

Our H_2 data were analysed in a *direct* comparison with the He ones, showing a diminution or an enhancement of the recoil intensity with respect to the binary one, in the molecular case. Though the reduction of the recoil intensity could have several plausible explanations, the simultaneous observation of a reduction and an enhancement depending on the ejected electron de Broglie wavelength supports on the one hand the interpretation of these effects as being the signature of the presence of interference effects as theoretically predicted in [18] and experimentally observed in [3], while on the other hand bringing further indication by showing the destructive or constructive character of these interferences.

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