Single- and two-centre effects in fully differential cross sections for single ionization of H_2 molecules by 75 keV protons

2015 J. Phys. B: At. Mol. Opt. Phys. 48 115204

(http://iopscience.iop.org/0953-4075/48/11/115204)

IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. **48** (2015) 115204 (8pp) doi:10.1088/0953-4075/48/11/115204

Single- and two-centre effects in fully differential cross sections for single ionization of H_2 molecules by 75 keV protons

M F Ciappina[1], C A Tachino[2], R D Rivarola[2], S Sharma[3] and M Schulz[3]

[1] Max Planck Institute of Quantum Optics, Hans-Kopfermann Str. 1, D-85748 Garching, Germany
[2] Laboratorio de Colisiones Atómicas, Instituto de Física Rosario, CONICET y Universidad Nacional de Rosario, Bv. 27 de Febrero 210 bis, 2000 Rosario, Argentina
[3] Department of Physics and LAMOR, Missouri University of Science and Technology, Rolla, MO 65409, USA

E-mail: marcelo.ciappina@mpq.mpg.de

Received 3 March 2015, revised 18 March 2015
Accepted for publication 23 March 2015
Published 22 April 2015

Abstract

We present theoretical calculations of single ionization of H_2 molecules by 75 keV proton impact. The computed fully differential cross sections for different electron ejection geometries and projectile kinematical conditions are compared with recent measurements made by Hasan *et al* (2014 *J. Phys. B: At. Mol. Opt. Phys.* **47** 215201). We employ a molecular version of the continuum distorted wave-eikonal initial state model, where all the interactions present in the exit channel are considered on an equal footing. In addition, our approach allows us to incorporate different interference terms and to assess their influence. Overall, the agreement between experiment and theory is better than for the case of more sophisticated schemes for coplanar geometries.

Keywords: atomic collisions, interference effects, distorted wave models

1. Introduction

The interaction of charged particles, such as electrons and ions, or photons with simple and complex molecules is of pivotal interest in the subsequent study in diverse fields of research, e.g. in atmospheric, plasma and biological physics (see e.g. [1,2] and references therein). Furthermore, the study of particle–molecule interactions is also important from a fundamental point of view. For example, information about the molecular structure, e.g. symmetry and characteristics of the orbitals, can be obtained using photons as *probes* [3, 4]. Most importantly, processes occurring in collisions of various particles with simple molecules are particularly suitable to study the complex and highly correlated reaction dynamics in such few-body Coulomb systems.

From an experimental point of view the development of the COLTRIMS (cold target recoil ion momentum spectroscopy) [5] and reaction microscope [6] techniques resulted in a major advancement of our understanding of the few-body dynamics in such collisions. With these techniques it is possible to simultaneously detect and fully momentum-analyze the different particles which participate in the process (for a review see [7]). As a consequence, kinematically complete experiments are feasible from which fully differential cross sections (FDCS) for a variety of processes and for a large fraction of the total phase space can be extracted. Initially, experimental efforts focused on studying the few-body dynamics in collisions of charged particles with atomic targets, especially helium (see e.g. [8–19], for a review see [20]).

The multi-centre nature of molecular targets can result in characteristic features in the reaction dynamics which are not present for atomic targets. Most notably, indistinguishable projectile diffraction off or (in the case of ionization) electron emission from the atomic centres of the molecule can lead to observable interference effects. Since the first experimental evidence of the existence of such interference patterns in single capture from and single ionization of molecules by ion impact was reported by Cheng *et al* [21] and Stolterfoht *et al* [22], a profuse activity, both theoretical and experimental,

have been carried out (see e.g. [23–42], for a recent review see [43]).

In the case of photon-impact, kinematically complete experiments for electron emission from molecular targets have been carried out already in the late nineties [44]. However, for charged-particle impact such experiments are significantly more challenging because the complete momentum vector of one more particle, the projectile (which for photons is usually negligible), has to be determined. For ionic projectiles this represents a particularly severe complication because the large mass (compared to electrons) results in very small (for fast and/or heavy ions immeasurably small) scattering angles and projectile energy losses (relative to the initial energy). As a result, only two kinematically complete experiments on ionization of H_2 by ion impact were reported so far [45–47]. In one, the important role of auto-ionization channels on the emission of very low-energy electrons was demonstrated [45]. In the second, pronounced interference structures were observed, however, the data revealed some features which were previously not expected for molecular two-centre interference and it is presently not clear whether the observed structures are not due to a different type of interference [46].

One important aspect relating to interference effects, which is well established in classical optics, but which was overlooked for a long time in the case of atomic collisions, was recently emphasized by Egodapitiya *et al* [47]. They demonstrated that interference structures in the cross sections for ionization of H_2 are only observable if the projectile beam is coherent, i.e. if the width of the projectile wave packet is large enough to illuminate both atomic centres in the molecule [47], but that they are not present for an incoherent beam. This interpretation was challenged by Feagin and Hargreaves [48], who argued that an experiment of the type reported in [47] is not sensitive to the structure of the projectile wavefunction. However, their analysis was subsequently rebutted by further experimental work, which ruled out the interpretation offered by Feagin and Hargreaves [48]. Furthermore, in the case of neutron reflection from a grating it was demonstrated, using a similar experimental approach as in [47], that the structure of the projectile wave packet can very well have a significant impact on the reflected neutron spectra [49]. Finally, the findings of Egodapitiya *et al* [47] are supported by several recent studies, which reported similar coherence effects for other processes and collisions systems [48, 50–53].

Theoretical approaches to tackle ionization or fragmentation of molecules by ion impact represent an arduous task as well, and most of them were applied predominantly to simple molecules (see e.g. [43] and references therein). However, in recent years theoretical studies of ionization of complex molecules were also reported [54]. For fast charged particles impact the scenario is somewhat more manageable since typically very little momentum is transferred from the projectile to the molecular target during the collision and ionization might become comparable to the ionization by photons (photoionization). This limit has been extensively discussed for atomic targets starting with the original work of Inokuti [55].

For single ionization of molecular targets, the knowledge of the multicentre initial bound and final continuum multielectronic states is required in order to compute the observables, e.g. FDCS. In order to avoid difficulties associated with multielectronic targets the most widely used approach is to reduce the problem to a one-active electron description, i.e. to invoke the so-called single active electron approximation. The latter has been successfully applied in single ionization of complex atoms by multicharged ions [56]. As far as the initial state is concerned, powerful computational codes are available which provide an accurate description of the molecular ground state in terms of a linear combination of atomic orbitals. In contrast, the final continuum state of the ejected electron is more complex to model since the interaction between the electron and the residual molecular multicentre ion represents a Coulomb-based multi-body problem. In a first approach it is possible to consider the ejected electron in an effective field produced by the residual ionic core, screened by the other passive electrons present in the molecular target. This last scheme, known as distorted-wave-Born approximation, has been successfully used to model single ionization processes in di- and poly-atomic molecules (see [39] and references therein). In an even much simpler approach the interaction of the ejected electron with both the residual molecular ion and with the projectile is neglected, i.e. a plane wave approximation is used. The latter has been used extensively in strong field induced processes in molecules (see e.g. [31] and references therein).

More elaborate schemes to describe the single ionization of simple molecules by the impact of heavy ions were developed over the years. For instance, it was shown that the continuum-distorted wave-eikonal initial state (CDW-EIS) approach of Crothers and McCann [57] can be easily extended to the treatment of collisions of heavy particles with mutilectronic targets [58]. Just to cite a couple of examples, single ionization of H_2 molecules by highly charged ions [38] and protons [59] impact was already studied within the so-called molecular version of the CDW-EIS (CDW-EIS-MO). In addition, within the CDW-EIS-MO scheme, it is possible to incorporate the interaction between the heavy nuclei (NN interaction) in a semiclassical way [60–62]. This theory is able to reproduce FDCS reasonably well for single ionization of helium by highly charged ions in a broad range of projectile velocities (see e.g. [63–68]). One of the main advantages of the CDW-EIS MO approach is the possibility to examine single- and two-centre interference effects in a simple and direct way (see e.g. [59]). For instance, it was possible to study the role of two-centre interference by analysing the experimental-to-theoretical cross section ratios for the single ionization of H_2 molecules by heavy charged particles [22], to extract the direct and interference contributions for the case of aligned and randomly oriented molecules [59] and to examine how quantum interference effects modify the main physical features of the ionization reaction associated with classical physics, i.e. the binary and recoil peaks [69].

Very recently fully differential data for single ionization of H_2 molecules by impact of 75 keV protons were produced [46] with the aim to experimentally elucidate the role of single- and two-centre interference effects for different electron ejection geometries. In this article we present a theoretical study of FDCS for this collision system based on the CDW-EIS-MO approach. Since in this model it is not possible to describe an incoherent projectile beam we compare the calculations with the data of [46] obtained for a (partially) coherent beam. In the following section we briefly describe our theoretical framework to deal with ion-H_2 collisions. Next, in section 3, our predictions are compared with recent experimental measurements and with other approaches. Within our framework it is possible to disentangle single- and two-centre interference effects and we will discuss this point in this section. Finally in section 4 we present our conclusions, together with a brief outlook. Atomic units are used throughout unless otherwise stated.

2. Theory

We will compute FDCS as a function of the momentum $\mathbf{k} = (k, \theta_k, \phi_k)$ of the ionized electron, being k the magnitude of the vector and θ_k and ϕ_k the corresponding polar and azimuthal angles, respectively, and of the transverse component $\boldsymbol{\eta}$ of the projectile momentum transfer $\mathbf{q} = \mathbf{K}_\mathrm{i} - \mathbf{K}_\mathrm{f}$, where $\mathbf{K}_\mathrm{i}$ ($\mathbf{K}_\mathrm{f}$) is the initial (final) momentum of the incoming particle. In our context $\boldsymbol{\eta} \cdot \hat{\mathbf{v}} = 0$, with $\hat{\mathbf{v}}$ giving the direction of the velocity vector $\mathbf{v}$ of the projectile. The FDCS is related to the prior-form of the transition amplitude $T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)$ by energy conservation, through the expression

$$\sigma^{(5)}\left(\mathbf{k}, \mathbf{q}, \boldsymbol{\rho}_0\right) = \frac{\mathrm{d}^{(5)}\sigma}{\mathrm{d}\mathbf{k}\mathrm{d}\boldsymbol{\eta}} = \frac{(2\pi)^4}{v}\left|T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)\right|^2 \delta\left(E_\mathrm{f} - E_\mathrm{i}\right), \tag{1}$$

with $\boldsymbol{\rho}_0 = (\rho_0, \theta_\rho, \phi_\rho)$ the internuclear equilibrium vector of the target, and E_i ($E_\mathrm{f} = k^2/2$) the energy of the bound (continuum) state of the ionized electron. It is important to note that, as it happens in the atomic target case, when differential cross sections depend on the transverse component of the momentum transfer or on the projectile scattering angle, for example, the interaction between the incident particle and each molecular nuclei must be included in the formulation since it may play a relevant role in the corresponding calculations, depending on the energies of the emitted electron and momentum transfer values [59]. On the other hand, when differential cross sections depending only on the electron energy and /or angular coordinates are considered, these nucleus–nucleus interactions are not included since their influence in the transition amplitude are reduced to a complex phase factor that gives no contribution to the cross sections (for details, see e. g. [43, 60]).

In order to compute the FDCS given by expression (1), a molecular version of the CDW-EIS-MO approach, within the so called two-effective centre (TEC) approximation, is employed. This method was already applied with success to study electron emission from molecular hydrogen by electron impact [70], and ionization of diatomic molecular targets by ion impact in the intermediate and high energy collision regimes [38, 39, 71–73]. Only the main aspects of this method will be summarized here, and the readers will be referred to, e.g., [39, 43] for a detailed explanation about it.

Thus, it is possible to show that the prior-form of the transition amplitude $T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)$ for the H_2 molecule can be written as

$$\left|T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)\right|^2 = 2\left\{1 + \cos\left[\mathbf{p}_\mathrm{rec} \cdot \boldsymbol{\rho}_0\right]\right\}\left|T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})\right|^2, \tag{2}$$

where $\mathbf{p}_\mathrm{rec} = \mathbf{q} - \mathbf{k}$. In equation (2), $T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})$ represents a CDW-EIS one-atom transition amplitude related to electron emission from effective hydrogen atoms located at the positions of each molecular nucleus. The initial orbital wavefunction for the H_2 molecule is described by means of a variational single-zeta function of the form [38]:

$$\phi_\mathrm{i}(\mathbf{x}) = N_\mathrm{i}(\rho_0)\left[\xi(\mathbf{x}_1) + \xi(\mathbf{x}_2)\right] \tag{3}$$

with

$$\xi(\mathbf{x}_j) = \left(\frac{Z_\mathrm{eff}^3}{\pi}\right)^{1/2} \exp\left(-Z_\mathrm{eff} x_j\right); \qquad j = 1, 2, \tag{4}$$

$Z_\mathrm{eff} = 1.193$ and $N_\mathrm{i}(\rho_0) = 0.5459$ being the normalization factor of the wavefunction corresponding to an equilibrium distance $\rho_0 = 1.4$ a.u. In the calculation of the scattering matrix element $T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})$, the orbital energy is taken from the experimental value obtained for single ionization of H_2 molecules, i.e. $\varepsilon_\mathrm{i} = -0.566$ a.u. (−15.4 eV). It must be also noted that $N_\mathrm{i}(\rho)\xi(\mathbf{x})$ could be considered as an *effective* atomic H wavefunction. At this point it must be mentioned that the Coulomb interaction among the projectile and each one of the nucleus of the molecular centres is incorporated *exactly* in $T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})$ in a semiclassical way [60–62].

An average over all the molecular orientations should be performed in (1) for the case of randomly oriented molecules. By integrating the transition amplitude $T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)$ given by equation (2) over the molecular angles θ_ρ and ϕ_ρ, it is possible to show that the averaged transition amplitude is given by

$$\left|T_\mathrm{fi}^{(-)}(\boldsymbol{\eta}, \rho_0)\right|^2 = 2\left\{1 + \frac{\sin\chi}{\chi}\right\}\left|T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})\right|^2, \tag{5}$$

where $T_\mathrm{fi}^{\mathrm{A},(-)}(\boldsymbol{\eta})$ is the same atomic transition amplitude presented in (2) for the case of oriented molecules, and $\chi = p_\mathrm{rec}\,\rho_0$. Finally, the averaged two-centre fully differential cross sections $\sigma^{(5),2}(\mathbf{k}, \mathbf{q}, \rho_0)$ can be defined as follows

$$\sigma^{(5),2}\left(\mathbf{k}, \mathbf{q}, \rho_0\right) = 2\left\{1 + \frac{\sin\chi}{\chi}\right\}\sigma^{(5),\mathrm{A}}(\mathbf{k}, \mathbf{q}), \tag{6}$$

where $\sigma^{(5),\mathrm{A}}(\mathbf{k}, \mathbf{q})$ represents a FDCS for single ionization by proton impact of an hydrogen atom. It is worth noting that, in equation (6) all the information about the multicentre nature of the target is included in the factor $2\left\{1 + \frac{\sin\chi}{\chi}\right\}$. Then, if

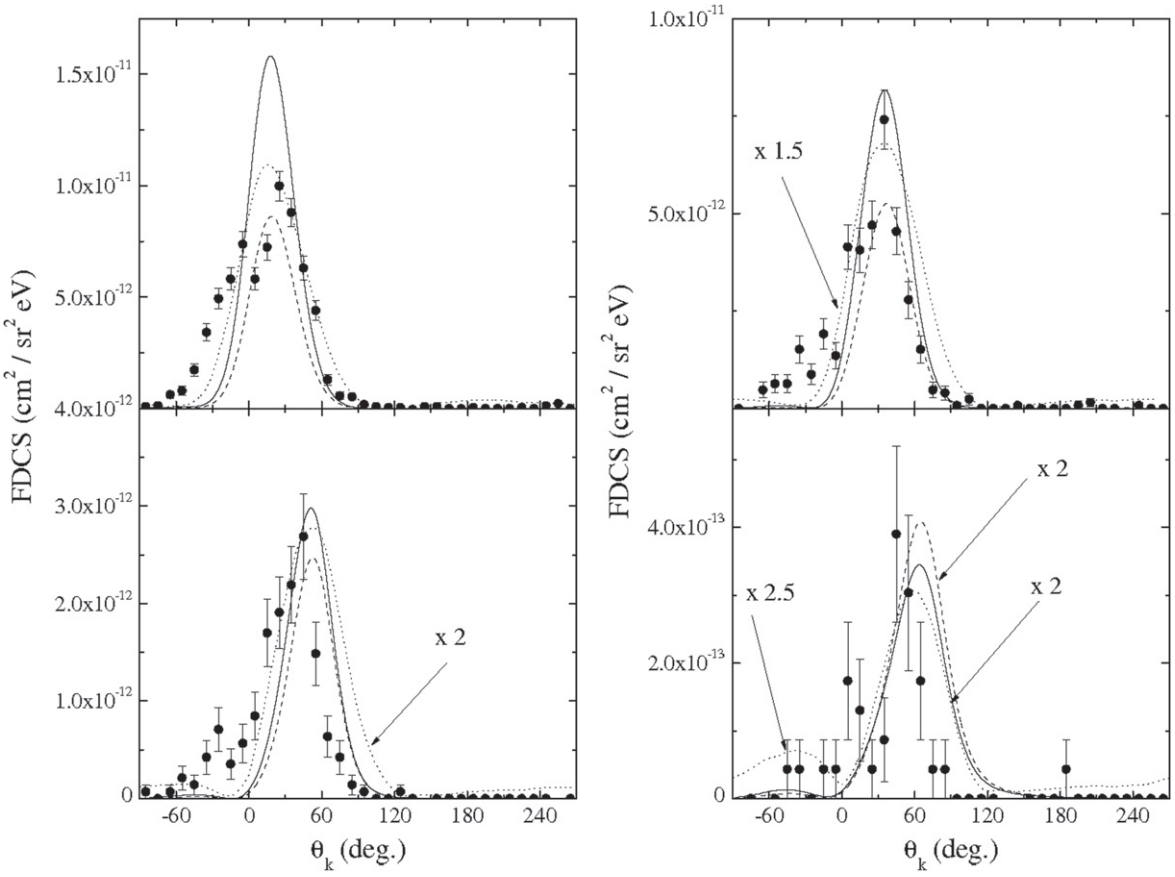

Figure 1. FDCS for electrons emitted with an energy E_k = 14.6 eV (k = 1.04 a.u.) ejected into the scattering plane as a function of the polar electron emission angle θ_k. The magnitude of the momentum transfer q is fixed at 0.71 (upper left panel), 0.9 (upper right panel), 1.21 (lower left panel), and 1.86 a.u. (lower right panel). Dots: experimental data [46], solid curves: TC-FDCS (see text for details), dashed curves: SC-FDCS (see text for details), dotted curves: M3DW results [46].

we neglect this oscillatory factor-which is related to the interferences arising from the two-centre character of the H_2 molecule, we can define an equivalent single-centre FDCS as

$$\sigma^{(5),1}(\mathbf{k}, \mathbf{q}) = 2\sigma^{(5),A}(\mathbf{k}, \mathbf{q}). \quad (7)$$

In the next section we compute $\sigma^{(5),2}(\mathbf{k}, \mathbf{q}, \rho_0)$ and $\sigma^{(5),1}(\mathbf{k}, \mathbf{q})$ for different electron emission geometries in order to assess the role of two- and single-centre interferences, respectively.

3. Results and discussion

The CDW-EIS-MO approach has been applied in order to compute FDCS for single ionization of H_2 targets by impact of protons at 75 keV (v = 1.73 a.u.). All the theoretical results presented in this section were performed following the experimental configuration reported in [46].

In figure 1, theoretical and experimental FDCS for electrons emitted with an energy of E_k = 14.6 eV (k = 1.04 a.u.) into the scattering plane, as a function of the polar electron emission angle θ_k, are shown. It should be noted that in the present work, $\theta_k \in [0°, 180°]$ corresponds to the binary peak side of the scattering plane and $\theta_k \in [180°, 360°]$ corresponds to the recoil peak side. Following [46], the magnitude of the momentum transfer was chosen to be q = 0.71 (upper left panel), q = 0.9 (upper right panel), q = 1.21 (lower left panel), and q = 1.86 a.u. (lower right panel). It is worth noting that the shape of the molecular FDCS resemble the ones obtained earlier for the same proton impact energy but with He as a target [9, 18].

For the experimental data presented in this and all following figures the full bin widths over which the data were integrated for each q are 0.14, 0.22, 0.4, and 0.89 a.u. for q = 0.71, 0.9, 1.21, and 1.86 a.u., respectively. In the electron ejection angle the corresponding bin width is 10° in all cases. The projectile energy loss (and thereby the ejected electron energy) is not determined from any measured spectrum, but rather by the voltage applied to the energy analyzer. Here, the effective bin width is given by the energy resolution, which is about 2–3 eV full width at half maximum (FWHM).

From a theoretical point of view both single-centre based FDCS (SC-FDCS) $\sigma^{(5),1}(\mathbf{k}, \mathbf{q})$, and two-centre based FDCS (TC-FDCS) $\sigma^{(5),2}(\mathbf{k}, \mathbf{q}, \rho_0)$, are computed. For completeness, M3DW theoretical results [46] are also included. It should be

noted that, within our single-centre approach, higher-order contributions associated with the interaction of the projectile with the active electron and with the target nuclei are taken into account as we use continuum factors for both of them. In the present study, the active electron initial wavefunction employed to calculate FDCS is approximated by a sum of simple exponential variational wavefunctions, as it was done in [39]. However, it is feasible to improve this description by using a more elaborated molecular orbital wavefunction, without loosing this simple two-centre image, as it was already done for the asymmetric HeH^+ molecular ion and for N_2 and O_2 molecules [71–75]. In the mentioned cases, the package Gaussian98 was employed [76] in order to optimize the coefficients and exponents of the Slater-type functions conforming each molecular orbital as well as the equilibrium internuclear distance and the orbital energies. Nevertheless, it must be taken into account that all this information is referred to the centre of mass of the molecule, resulting it in 'effective' atomic one-centre wavefunctions, which are observed from the centre of mass of the molecule and preserve the two-molecular centre nature of the target.

In an impact parameter treatment, it must be taken into account that for each molecular orientation the projectile sets an impact parameter vector with respect to the centre of mass of the target and, at the same time, it defines also two other impact parameter vectors related to each nucleus of the hydrogen molecule. As a consequence, in spite of the fact that we are considering an impact parameter with respect to the H_2 centre of mass, it should be kept in mind that actually the impact parameters associated to each target centre are not independent of each other. In the present study, the more crude approximation is related to the final continuum wavefunction. In the TEC approximation employed here to obtain the molecular transition amplitude $T_{\rm fi}^{(-)}(\boldsymbol{\eta}, \boldsymbol{\rho}_0)$ (see equation (2)), the active electron is assumed to be emitted from a region close to one of the nucleus of the H_2 molecule at a time, while the other one is screened completely by the non-ionized electron. In consequence, and since the 'complete' molecular orbitals has been constructed using a linear combination of Slater-type orbitals centred on each target nuclei, it is possible to consider a continuum wavefunction for the active electron related to the nucleus from which it is emitted. In fact our two-centre model does not consider that the ionized electron could feel simultaneously the influence of both molecular centres. At a first sight, this could configure a very coarse approximation. However, Weck *et al* [77] have shown that, for example, in the case of electron impact the use of two-centre continuum wavefunctions did not involve major improvements in the molecular averaged FDCS. Nevertheless, more significant differences appeared when FDCS were calculated for fixed molecular orientations.

From figure 1, the first conclusion we can draw from the comparison between both sets of theoretical results corresponding to the single- and the two-centre CDW-EIS MO formulation and the experimental data, is the very good agreement between them for the cases of $q = 0.71$ a.u. (although for this value of the momentum transfer, two-centre calculations overestimates the experiments by a factor of 1.5 approximately), $q = 0.9$ and $q = 1.21$ a.u.. In contrast, for $q = 1.86$ a.u., the magnitudes of both SC-FDCS and TC-FDCS are smaller than the experimental data by a factor of 2. On the other side, it is observed that the curves obtained within the M3DW theory [46] for $q = 0.9$, $q = 1.21$ and $q = 1.86$ a.u. need to be multiplied by larger values to be closer to the measurements, where these multiplicative factors are of the order of 1.5, 2 and 2.5, respectively. The exception is the M3DW-FDCS corresponding to the momentum transfer of magnitude $q = 0.71$ a.u., that agree quite well with the experiment. The second important observation is that for $q = 0.71$ and $q = 0.9$ a.u. the SC-FDCS results are in better agreement with the experimental data than the TC-FDCS results [46]. Furthermore, for $q = 1.21$ and $q = 1.86$ a.u. there are only small differences between both theoretical data sets. Although we cannot make definite conclusions, this suggests that the contributions from single-centre interference are larger than those from two-centre interference. In this sense our calculations are consistent with the conclusions of Sharma *et al* [46, 53].

As it was mentioned earlier in the text, another experimental data configuration presented in [46] was considered by plotting FDCS as a function of the azimuthal electron emission angle ϕ_k, for fixed values of the polar angle θ_k. In figure 2, results obtained for the same values of the momentum transfer analysed in figure 1, namely $q = 0.71$ (upper left panel), $q = 0.9$ (upper right panel), $q = 1.21$ (lower left panel) and $q = 1.86$ a.u. (lower right panel), are shown. In the first three cases, the electron polar angle was chosen to be $\theta_k = 35°$, whereas for $q = 1.86$ a.u. this value was set at $\theta_k = 45°$. The azimuthal angular axis in the plots of figure 2 is in some unconventional way, since ϕ_k is referred to the angle between the projection of k onto the x–y plane and the positive y-semi-axis, rather than the positive x-semi-axis. With this convention, $\phi_k = 90°$ coincides with the direction of the q_x component of the vector $\mathbf{q}$. Reasonably good agreement between both SC-FDCS and TC-FDCS and the experimental data is found in the absolute magnitude (except for $q = 1.86$ a.u.); however, the calculations significantly underestimate the width of the peak structure. In addition, it can be seen that, as in the θ_k dependence of the FDCS, both sets of theoretical results become increasingly similar to each other with increasing q, both qualitatively and quantitatively. Overall, the agreement of our calculations with the experimental data is similar to the M3DW model. Perhaps, in magnitude our calculations do slightly better while the M3DW model has the edge as far as the shape of the ϕ-dependence is concerned. As for the case of single ionization of atoms, these discrepancies suggest an incomplete description of higher-order contributions [78]. The two-centre interference CDW-EIS MO approach appears to reproduce better the binary peak region, in particular for the cases of $q = 0.9$ and $q = 1.21$ a.u. This could be an indication that two-centre interference is more important for electron emission outside the scattering plane. However,

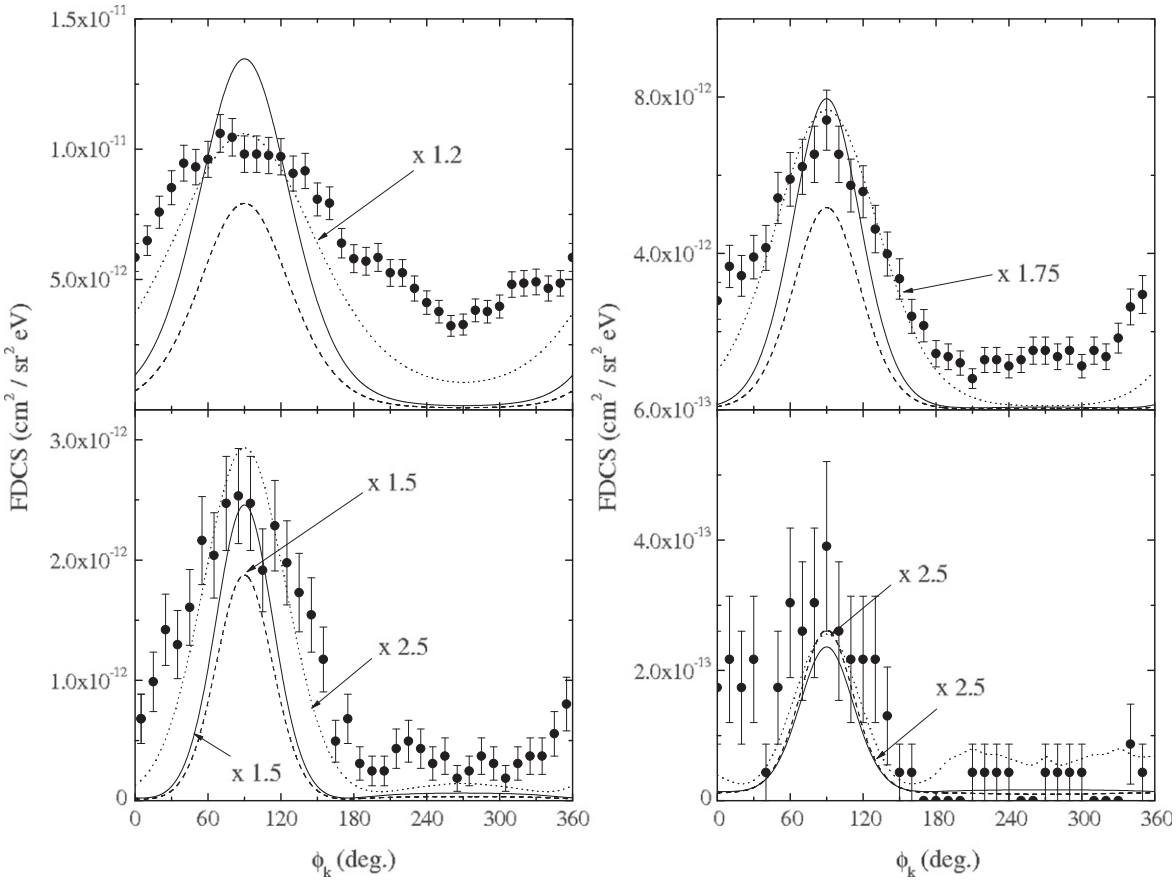

Figure 2. FDCS for electrons with an energy of E_k = 14.6 eV ejected along the surface of a cone with an opening angle $\theta_k = 35°$ ($\theta_k = 45°$ for q = 1.86 a.u.) as a function of the azimuthal electron emission angle ϕ_k. Here, the values of q are the same as in figure 1. Dots: experimental data [46], solid curves: TC-FDCS, dashed curves: SC-FDCS, dotted curves: M3DW results [46].

given the discrepancies in the shape of the θ_k-dependence of the FDCS we cannot make any conclusive statement to this end. Furthermore, approximating the final state wavefunction as a one-centre continuum wavefunction might contribute to these discrepancies. This will be matter of future investigations.

In figure 3, SC- and TC-FDCS as a function of the polar angle θ_k, for electrons emitted into the perpendicular plane, are shown together with the experimental data. In this case, the same values of the momentum transfer q presented in figures 1 and 2 are considered. Similar to the case of electron emission into the scattering plane, it can be seen that the general behaviour of the curves is comparable to the 75 keV p + He collisions [9, 18]. A fairly bad agreement between the experimental data and both CDW-EIS curves is observed. These discrepancies are even larger than the ones obtained when the theoretical M3DW-FDCS are considered, especially for the two largest values of q, i.e. q = 1.21 and q = 1.86 a.u.. On the other hand, there is a reasonable good agreement in magnitude between the experimental data and the two-centre CDW-EIS-MO results for the case of q = 0.71 a.u., in particular for values of $\theta_k \approx 0°$. In conclusion, and taken into account the previous discussion, it is not possible to us to elucidate which of the two effects, namely the single- or the two-centre electron emission, have more influence in the case of electron ionization in the perpendicular plane.

4. Conclusions and perspectives

We have carried out computations of FDCS for single ionization of H_2 molecules by 75 keV proton impact and compared them with recent experimental data. The theoretical results presented in this work were obtained within a molecular version of the CDW-EIS-MO approach. In this approximation, all the interactions present in the exit channel are considered on an equal footing. Such a model allows us to disentangle the so-called single- and two-centre interference effects. A reasonable good agreement between our theory and the measurements is achieved, in particular for coplanar geometries. Overall, the agreement between the CDW-EIS theory and the experiments is not worse than the one observed when more sophisticated schemes are employed. Substantial differences appear, however, for non-coplanar emission geometries and for larger values of the momentum transfer. Our approach appears to be perfectly suitable to tackle other *unconventional* electron emission geometries, for instance, those presented in [47] and recently in [79], where the interference effects are clearly enhanced.

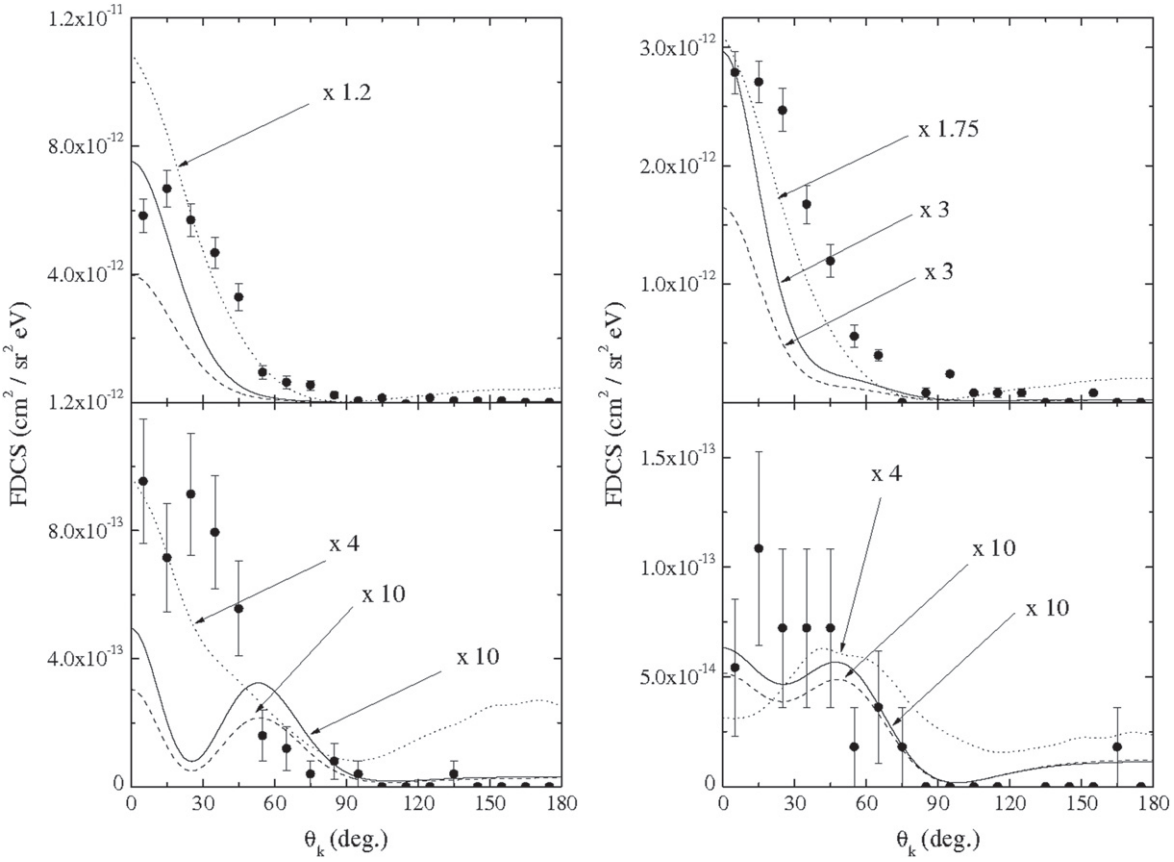

Figure 3. Same as figure 1, but for the electron emitted into the perpendicular plane.

Acknowledgments

This work was supported by the National Science Foundation under grant nos. PHY- 1401586. CAT and RDR acknowledge financial support from PICT 2145 of the Agencia Nacional de Promoción Científica y Tecnológica.

References

[1] Stolterfoht N, DuBois R D and Rivarola R D 1997 *Electron Emission in Heavy Ion-Atom Collisions* (Berlin: Springer)
[2] Drake W F (ed) 2006 *Springer Handbook of Atomic, Molecular, and Optical Physics* (Berlin: Springer)
[3] Itatani J *et al* 2004 *Nature* **432** 867
[4] Kanai T, Minemoto S and Sakai H 2005 *Nature* **435** 470
[5] Ullrich J *et al* 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** 2917
[6] Ullrich J *et al* 2003 *Rep. Prog. Phys.* **66** 1463
[7] Dörner R, Mergel V, Jagutzski O, Spielberger L, Ullrich J, Moshammer R and Schmidt-Böcking H 2000 *Phys. Rep.* **330** 95
[8] Schulz M, Moshammer R, Fischer D, Kollmus H, Madison D H, Jones S and Ullrich J 2003 *Nature* **422** 48
[9] Maydanyuk N V, Hasan A, Foster M, Tooke B, Nanni E, Madison D H and Schulz M 2005 *Phys. Rev. Lett.* **94** 243201
[10] An L, Khayyat K and Schulz M 2001 *Phys. Rev.* A **63** 030703
[11] Schulz M *et al* 2001 *J. Phys. B: At. Mol. Opt. Phys.* **34** L305
[12] Schulz M, Moshammer R, Perumal A N and Ullrich J 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** L161
[13] Fischer D, Moshammer R, Schulz M, Voitkiv A and Ullrich J 2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** 3555
[14] Voitkiv A B, Najjari B, Moshammer R, Schulz M and Ullrich J 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** L365
[15] Schulz M, Moshammer R, Voitkiv A, Najjari B and Ullrich J 2005 *Nucl. Instrum. Methods* B **235** 296
[16] Schulz M *et al* 2013 *Phys. Rev.* A **88** 022704
[17] Hubele R *et al* 2013 *Phys. Rev. Lett.* **110** 133201
[18] Schulz M, Hasan A, Maydanyuk N V, Foster M, Tooke B and Madison D H 2006 *Phys. Rev.* A **73** 062704
[19] Hasan A, Maydanyuk N V, Fendler B J, Voitkiv A, Najjari B and Schulz M 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** 1923
[20] Schulz M and Madison D H 2006 *Int. J. Mod. Phys.* A **21** 3649
[21] Cheng S, Cocke C L, Frohne V, Kamber E Y, McGuire J H and Wang Y 1993 *Phys. Rev.* A **47** 3923
[22] Stolterfoht N *et al* 2001 *Phys. Rev. Lett.* **87** 023201
[23] Stia C R, Fojón O A, Weck P F, Hanssen J and Rivarola R D 2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** L257
[24] Kamalou O, Chesnel J-Y, Martina D, Hanssen J, Stia C R, Fojón O A, Rivarola R D and Frémont F 2005 *Phys. Rev.* A **71** 010702(R)
[25] Gao J F, Madison D H and Peacher J L 2005 *Phys. Rev.* A **72** 020701
[26] Gao J F, Madison D H and Peacher J L 2005 *Phys. Rev.* A **72** 032721
[27] Gao J F, Madison D H and Peacher J L 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 1275
[28] Milne-Brownlie D S, Foster M, Gao J F, Lohmann B and Madison D H 2006 *Phys. Rev. Lett.* **96** 233201
[29] Fojón O A, Fernández J, Palacios A, Rivarola R D and Martín F 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** 3035
[30] Rolles D *et al* 2005 *Nature* **437** 711

[31] Jaroń-Becker A, Becker A and Faisal F H M 2004 *Phys. Rev.* A **69** 023410
[32] Fojón O A, Palacios A, Fernández J, Rivarola R D and Martín F 2006 *Phys. Lett.* A **350** 371
[33] Della Picca R, Fainstein P D, Martiarena M L and Dubois A 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 473
[34] Della Picca R, Fainstein P D, Martiarena M L and Dubois A 2007 *Phys. Rev.* A **75** 032710
[35] Martín F *et al* 2007 *Science* **315** 629
[36] Fernández J, Fojón O, Palacios A and Martín F 2007 *Phys. Rev. Lett.* **98** 043005
[37] Laurent G, Fainstein P D, Galassi M E, Rivarola R D, Adoui L and Cassimi A 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** L495
[38] Galassi M E, Rivarola R D, Fainstein P D and Stolterfoht N 2002 *Phys. Rev.* A **66** 052705
[39] Galassi M E, Rivarola R D and Fainstein P D 2004 *Phys. Rev.* A **70** 032721
[40] Dimopoulou C *et al* 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 593
[41] Dimopoulou C *et al* 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 3173
[42] Alexander J S, Laforge A C, Hasan A, Machavariani Z S, Ciappina M F, Rivarola R D, Madison D H and Schulz M 2008 *Phys. Rev.* A **78** 060701
[43] Ciappina M F, Fojón O A and Rivarola R D 2014 *J. Phys. B: At. Mol. Opt. Phys.* **47** 042001
[44] Martín F 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** R197
[45] Dimopoulou C *et al* 2004 *Phys. Rev. Lett.* **93** 123203
[46] Sharma S, Arthanayaka T P, Hasan A, Lamichhane B R, Remolina J, Smith A and Schulz M 2014 *Phys. Rev.* A **90** 052710
[47] Egodapitiya K N, Sharma S, Hasan A, Laforge A C, Madison D H, Moshammer R and Schulz M 2011 *Phys. Rev. Lett.* **106** 153202
[48] Feagin J M and Hargreaves L 2013 *Phys. Rev.* A **88** 032705
[49] Majkrzak C F, Metting C, Maranville B B, Dura J A, Satija S, Udovic T and Berk N F 2014 *Phys. Rev.* A **89** 033851
[50] Wang X, Schneider K, LaForge A, Kelkar A, Grieser M, Moshammer R, Ullrich J, Schulz M and Fischer D 2012 *J. Phys. B: At. Mol. Opt. Phys.* **45** 211001
[51] Sharma S, Hasan A, Egodapitiya K N, Arthanayaka T P and Schulz M 2012 *Phys. Rev.* A **86** 022706
[52] Schneider K, Schulz M, Wang X, Kelkar A, Grieser M, Krantz C, Ullrich J, Moshammer R and Fischer D 2013 *Phys. Rev. Lett.* **110** 113201
[53] Sharma S, Arthanayaka T P, Hasan A, Lamichhane B R, Remolina J, Smith A and Schulz M 2014 *Phys. Rev.* A **89** 052703
[54] Lüdde H J, Spranger T, Horbatsch M and Kirchner T 2009 *Phys. Rev.* A **80** 060702(R)
[55] Inokuti M 1971 *Rev. Mod. Phys* **43** 297
[56] Fainstein P D, Ponce V H and Rivarola R D 1988 *J. Phys. B: At. Mol. Opt. Phys.* **21** 287
[57] Crothers D S F and McCann J F 1983 *J. Phys. B: At. Mol. Phys.* **16** 3229
[58] Gulyás L and Fainstein P D 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 3297
[59] Ciappina M F and Rivarola R D 2008 *J. Phys. B: At. Mol. Opt. Phys.* **41** 015203
[60] Fainstein P D, Ponce V H and Rivarola R D 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 3091
[61] Rodríguez V D and Barrachina R O 1998 *Phys. Rev.* A **57** 215
[62] Sanchez M D, Cravero W R and Garibotti C R 2000 *Phys. Rev.* A **61** 062709
[63] Ciappina M F and Cravero W R 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 1091
[64] Ciappina M F and Cravero W R 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 2183
[65] Ciappina M F, Cravero W R and Schulz M 2007 *J. Phys. B: At. Mol. Opt. Phys.* **40** 2577
[66] Fainstein P D and Gulyás L 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 317
[67] Foster M, Madison D H, Peacher J L and Ullrich J 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** 3797
[68] Pedlow R T, O'Rourke S F C and Crothers D S F 2005 *Phys. Rev.* A **72** 062719
[69] Fojón O A, Stia C R and Rivarola R D 2011 *Phys. Rev.* A **84** 032715
[70] Weck P, Fojón O A, Hanssen J, Joulakian B and Rivarola R D 2001 *Phys. Rev.* A **63** 042709
[71] Tachino C A, Galassi M E, Martín F and Rivarola R D 2009 *J. Phys. B: At. Mol. Opt. Phys.* **42** 075203
[72] Tachino C A, Galassi M E, Martín F and Rivarola R D 2010 *J. Phys. B: At. Mol. Opt. Phys.* **43** 135203
[73] Tachino C A, Martín F and Rivarola R D 2012 *J. Phys. B: At. Mol. Opt. Phys.* **45** 025201
[74] Nandi S, Agnihotri A N, Tachino C A, Rivarola R D, Martín F and Tribedi L C 2012 *J. Phys. B: At. Mol. Opt. Phys.* **45** 215207
[75] Nandi S, Agnihotri A N, Kasthurirangan S, Kumar A, Tachino C A, Rivarola R D, Martín F and Tribedi L C 2012 *Phys. Rev.* A **85** 062705
[76] Frisch M J *et al* 1999 *Gaussian 98, Revision A11* (Pittsburgh, PA: Gaussian Inc.)
[77] Weck P F, Fojón O A, Joulakian B, Stia C R, Hanssen J and Rivarola R D 2002 *Phys. Rev.* A **66** 012711
[78] Colgan J, Pindzola M S, Robicheaux F and Ciappina M F 2011 *J. Phys. B: At. Mol. Opt. Phys.* **44** 175205
[79] Arthanayaka T P, Sharma S, Lamichhane B R, Hasan A, Remolina J, Gurung S and Schulz M 2015 *J. Phys. B: At. Mol. Opt. Phys.* **48** 071001