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Letter

Fully differential study of ionization in p + H_2 collisions near electron—projectile velocity matching

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Abstract

We have performed a kinematically complete experiment on ionization of H_2 by 75 keV proton impact for electrons ejected with a speed close to the projectile speed. The fully differential data are compared to a three-body distorted wave and a continuum distorted wave—eikonal initial state calculation. Large discrepancies between experiment and theory, as well as between both calculations, are found. These probably arise from a strong coupling between the ionization and capture channels, which is not accounted for by theory.

Keywords: Ionization, energy-loss spectroscopy, few-body problem, cold target recoil-ion momentum spectroscopy

(Some figures may appear in colour only in the online journal)

With the advent of cold target recoil ion momentum spectroscopy (COLTRIMS [1, 2]) kinematically complete experiments on target ionization by ion impact became feasible [3, 4]. In this method, the recoil-ion momentum is measured in addition to the momentum of either the ejected electron [e.g. [5–9]] or the scattered projectile [e.g. [10–13]] and the momentum of the third (undetected) collision fragment is obtained using momentum conservation. The fully differential cross sections (FDCSs) that can be extracted from such experiments offer the most sensitive tests of theoretical models (for a recent review see [14]).

Initially, these experiments focused on studying the ionization of helium [e.g. [5–7, 9, 10]]. After theory had made remarkable progress in reproducing experimental data for the ionization of simple targets by electron impact [e.g. [15, 16], it was quite surprising that for ion impact, significant—and in some cases severe—discrepancies were found [e.g. [17–22]. Recently, an experimental study [23] suggested that these discrepancies may be partly due to projectile coherence properties, which are not realistically treated in most of the existing theoretical models. For fast heavy ions the coherence length tends to be very small so that only a small fraction of the target dimension is coherently illuminated. As a result, interference effects, present in calculations assuming a coherent projectile beam, are not experimentally observable. This interpretation has since received further experimental support [11, 13, 24, 25]. Nevertheless, especially for highly charged ion impact, it seems likely that the projectile coherence properties are not the only factor contributing to the discrepancies. For this ion species, higher-order contributions to the transition amplitudes can reach a magnitude large enough to represent an enormous challenge to theory.

Higher-order processes involving the post-collision interaction (PCI) between the outgoing scattered projectile and the electron, which has been lifted to the continuum by a preceding primary interaction, have been studied extensively (e.g. [6, 9, 26–29]). These studies have shown that the role of PCI maximizes when the electron speed v_{el} approaches the projectile speed v_p [29, 30]. Well-known manifestations of PCI are the occurrence of a sharp peak in the energy spectrum (the so-called 'cusp peak') of electrons ejected in the forward direction [26–28] and a narrowing of the scattered projectile angular distribution, which is particularly pronounced when $v_{el} \approx v_p$ [29]. However, FDCSs are very difficult to measure for this kinematic regime, at least for fast-ion impact. The problem is that for projectile energies of the order of MeV/amu, $v_{el} \approx v_p$ corresponds to ejected electron energies of the order of keVs. Direct measurement of such large electron energies within a COLTRIMS set-up is only possible at the expense of a poor recoil-ion momentum resolution. The only experimental FDCSs for $v_{el} \approx v_p$ reported so far were measured for 75 keV $p + H_2$ collisions [13], for which the corresponding electron energy is relatively small (41.6 eV). Furthermore, the electron energy was not measured directly, but obtained from the projectile energy loss. That work was focused on the role of projectile coherence effects; impacts of the PCI on the collision dynamics were not analyzed.

In this paper we present a comparative study between experiment and theory on FDCSs for ionization of H_2 by ion impact in the regime $v_{el} \approx v_p$, which is still a largely unexplored kinematic regime. The measurements and calculations were performed for various electron ejection geometries. Stunning discrepancies not only between the measured and calculated FDCSs, but also between two conceptually similar theoretical models were found. This indicates a large sensitivity of the FDCSs to the details of the dynamics of the ionization process for $v_{el} \approx v_p$. A strong coupling between the ionization and capture channels in this regime could at least partly be responsible for these unusual discrepancies.

The details of the experiment have been reported previously [11, 12]. In brief: a proton beam was extracted from a hot cathode ion source and accelerated to an energy of 75 keV plus 57 eV. The beam was collimated by an aperture 1.5 mm in diameter, located at the end of the accelerator terminal, and by a pair of horizontal and vertical slits with a width of 150 μm placed at a distance of 50 cm before the target region. This geometry of the collimating slit corresponds to a transverse coherence length of about 3.3 a.u. After intersecting a very cold ($T \approx 1$–2 K) neutral H_2 target beam, the projectiles passed through a switching magnet to eliminate charge-exchanged beam components. An H_2 target was chosen because part of the original motivation for this work was to study the influence of the projectile coherence properties on molecular two-center interference effects [13].

The protons were then decelerated by 70 keV and energy-analyzed by an electrostatic parallel-plate analyzer [31] (for a review on projectile energy-loss spectroscopy see [32]), which was set to a pass-energy of 5 keV. Therefore, only protons that suffered an energy loss of $\varepsilon = 57$ eV in the collision with the target passed the analyzer and were detected by a two-dimensional position-sensitive micro-channel plate detector. The entrance and exit slits were very narrow (75 μm) in the vertical direction (y-direction) and long ($\approx$2.5 cm) in the x-direction. Therefore, the y-component of the momentum transfer $\mathbf{q} = \mathbf{p_o} - \mathbf{p_f}$ (where $\mathbf{p_o}$ and $\mathbf{p_f}$ are the initial and final projectile momenta) was fixed at zero (within the resolution) and the x-component could be determined from the x-component of the position on the detector. The scattering angle $\theta_p = \sin^{-1}(q_x/p_o)$ was obtained with a resolution of about 0.13 mrad full width at half maximum (FWHM). The z-component of $\mathbf{q}$ is to a very good approximation given by $q_z = \varepsilon/v_p = 1.21$ a.u. The energy loss resolution of 3 eV FWHM corresponds to a resolution in q_z of 0.06 a.u. FWHM.

The H_2^+ recoil ions produced in the collision were extracted by a weak, nearly uniform electric field ($E \approx 8$ V cm^{-1}) pointing in the x-direction applied over a length of 10 cm. The ions then drifted in a field-free region 20 cm in length and were finally detected by a second two-dimensional position-sensitive micro-channel plate detector. From the position information the two momentum components in the plane of the detector (i.e. the y- and z-components) could be determined. The projectile and recoil-ion detectors were set in coincidence. The time-of-flight of the recoil-ions from the collision region to the detector, which is contained in the coincidence time, was used to calculate the x-component of the recoil-ion momentum vector. The resolution in the x-, y-, and z-components was 0.15, 0.5, and 0.15 a.u. FWHM, respectively. The electron momentum was obtained using momentum conservation as $\mathbf{p_{el}} = \mathbf{q} - \mathbf{p_{rec}}$.

The experimental results are compared with the molecular continuum distorted wave Eikonal initial state approximation (CDW-EIS MO) [33] and the molecular three-body distorted wave Eikonal initial state (M3DW-EIS) approximation [34], which have been described in previous publications so only a brief description noting the similarities and differences will be presented here. In the CDW-EIS MO approach [33], the T-matrix is approximated as a sum of two terms corresponding to ionization of effective atomic wavefunctions centered on the two H_2 nuclei. CDW-EIS transition amplitudes are used for the individual atomic terms within an impact parameter approximation. This approximation allows us to treat separately the quantum-mechanical dynamics of the electrons from the classical dynamics of the nuclei. Next, the interaction between the projectile and the target nuclei, the so-called 'N-N interaction', is considered in all orders within this approximation. The average over all molecular orientations yields a cross-section that is a product of an interference term and a CDW-EIS cross-section for the two effective atomic centers.

The M3DW-EIS approach [34] is a fully quantum mechanical approach, with the initial projectile wavefunction being represented as an Eikonal wave and the scattered

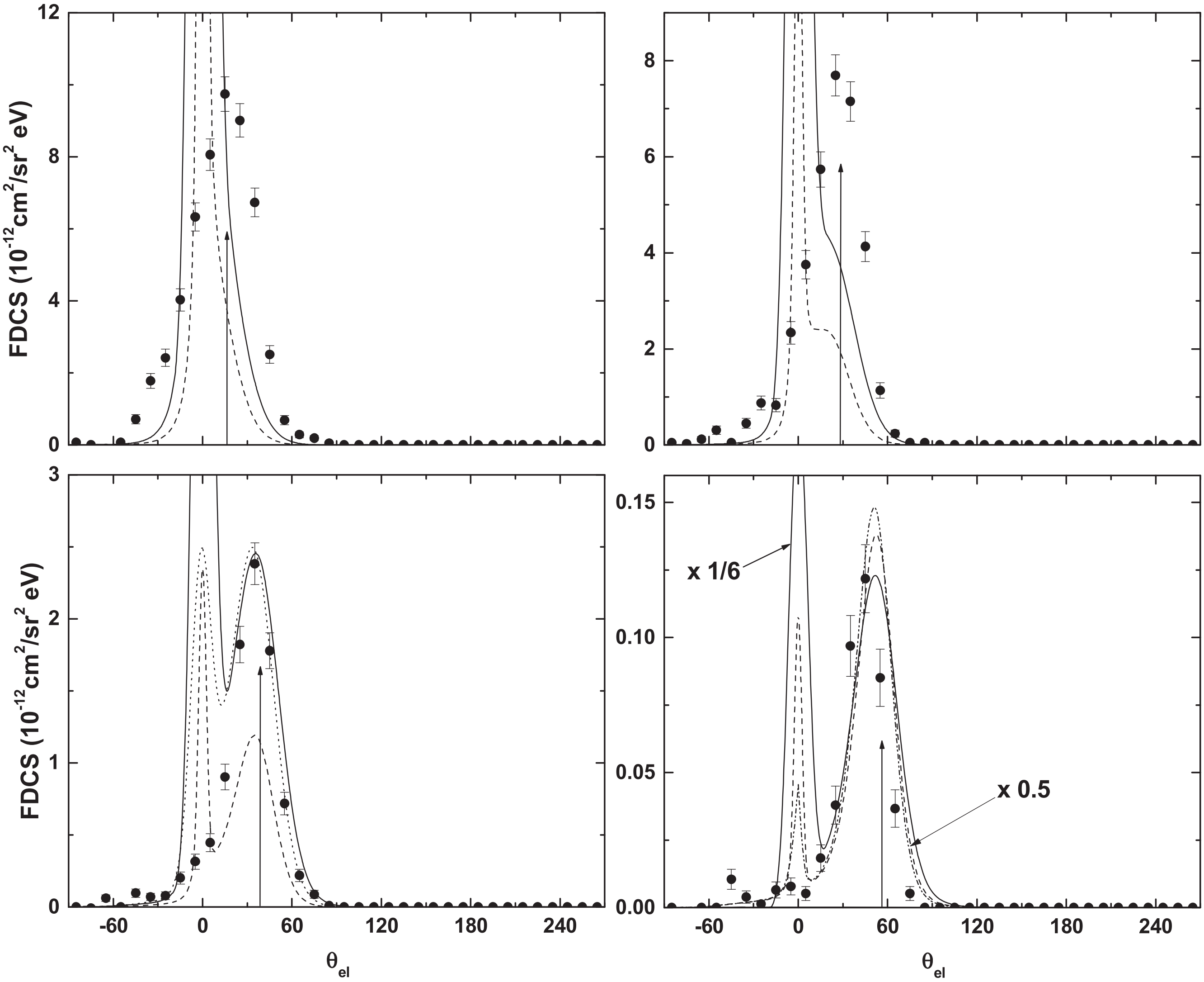

Figure 1. FDCSs for electrons with an energy of 41. 6 eV ejected into the scattering plane as a function of the polar electron emission angle. The projectile scattering angle is fixed at 0.1 mrad (upper left), 0.2 mrad (upper right), 0.325 mrad (lower left), and 0.55 mrad (lower right). Solid curves, M3DW-EIS calculations; dashed curves, CDW-EIS calculations; dotted curve (for 0.325 mrad), CDW-EIS calculation convoluted with an angular resolution of 20° FWHM; dash-dotted curve (for 0.55 mrad), CDW-EIS calculation for an ejected electron energy of 43.6 eV.

projectile being represented as a Coulomb wave for effective charge +1. The initial molecular wavefunction is represented by a numerical Hartree–Fock H_2 ground state Dyson orbital averaged over all orientations. The ejected electron wavefunction is a numerical distorted wave calculated using a two-center distorting potential obtained from the Hartree–Fock H_2 charge density averaged over all orientations. The final state wavefunction for the system is approximated as a product of the Coulomb wave for the projectile, the distorted wave for the ejected electron, and the exact final state projectile-electron interaction (the PCI). Finally, the initial state projectile-target interaction contains both the projectile-active electron interaction and the interaction between the projectile and an effective ion of charge +1.

In figure 1 the FDCSs are plotted for electrons with an energy of 41.6 eV ejected into the scattering plane, spanned by $\mathbf{p_o}$ and $\mathbf{q}$, as a function of the polar electron ejection angle θ_{el}. θ_p was fixed at 0.1 mrad (upper left panel), 0.2 mrad (upper right panel), 0.325 mrad (lower left panel), and 0.55 mrad (lower right panel), respectively. The arrows indicate the direction of the momentum transfer for each θ_p. In the experimental data, in each case a pronounced peak structure, the so-called 'binary peak', is observed near the direction of $\mathbf{q}$. With increasing θ_p the binary peak is increasingly shifted in the forward direction relative to $\mathbf{q}$. A similar trend was also observed in the FDCSs for ionization by fast, highly charged ion impact [9].

The solid and dashed curves in figure 1 show calculations based on the M3DW-EIS [34] and the CDW-EIS [33] approaches, respectively. As described above, the two models represent different approximations for essentially the same physics. Although both of them treat the ionization process perturbatively to first-order in the operator of the T-matrix, they both also incorporate higher-order contributions (both in the projectile-electron and in the projectile-target nucleus interaction) in the final-state wavefunction. There are dramatic discrepancies to the experimental data: in both calculations a very sharp and large peak structure is found at $\theta_{el} = 0$ for all scattering angles, which is absent in the measured FDCSs. Furthermore, the binary peak, i.e. the only structure observed in the experimental data, is completely missing in both calculations for $\theta_p = 0.1$ mrad. For all θ_p the height of the 0^o peak in the calculations is much larger than the magnitude of the binary peak in the experimental FDCSs (for $\theta_p = 0.1$ mrad, by about an order of magnitude in the case of the M3DW-EIS results). Considering the conceptual similarity of the two models, the large differences in magnitude between the theoretical curves are quite surprising. On the other hand, for the two larger θ_p, the shape of the measured binary peak is very well reproduced by both calculations and for $\theta_p = 0.325$ mrad, the M3DW-EIS model also predicts the magnitude correctly.

A strong 0° peak for $v_{el} = v_p$ appears to be a feature that one may have expected since the attractive PCI is known to produce an enhanced electron flux in the forward direction. This raises the question of whether the discrepancies between experiment and theory are simply due to inaccuracies in the calibration shifting the peak, which should be located at 0^o, to larger θ_{el}. However, this does not explain the comparison between experiment and theory at the two larger θ_p, where in the calculations two peak structures are observed, but only one is seen in the measured FDCSs. Furthermore, the accuracy of the calibration has been thoroughly tested [35] and also seems to be confirmed by the good agreement between experiment and theory regarding the location of the binary peak. Finally, we found that the shape of the FDCS near $\theta_{el} = 0$ is surprisingly insensitive to the calibration.

Another possibility is that the absence of the 0° peak in the data could be due to its very small width (6° FWHM in the CDW-EIS calculation). Since data points are taken in steps of 10°, the peak structure could simply have been 'missed'. However, since the angular resolution ($\approx 15^\circ$ FWHM) is larger than the step size, the 0° peak should have been observed, although with a much larger width than in the calculation, and therefore, since the integrated peak content cannot change, with a reduced height. That leaves the possibility that the 0° peak is not resolved from the binary peak and/or is so much reduced in height because of the resolution that it is no longer visible. We have therefore convoluted the CDW-EIS calculation for $\theta_p = 0.325$ mrad with a pessimistic resolution of 20° FWHM and the result is shown, after renormalizing to the height of the experimental binary peak, as the dotted curve in figure 1. Indeed, in the convolution the height of the 0° peak is significantly reduced relative to the binary peak, but nevertheless a well-resolved structure as tall as the binary peak remains. A further increase in resolution would make the binary peak too broad compared to the experimental data. We therefore conclude that the poor agreement between the measured and calculated FDCSs cannot be explained by experimental artifacts, at least not for the two larger θ_p. For $\theta_p = 0.1$ mrad (and possibly also for 0.2 mrad), on the other hand, it is quite possible that a significant 0° peak, unresolved from the binary peak, exists in the experimental data. But even if this is the case, the magnitude of the 0° peak relative to the binary peak would still be severely overestimated by theory since the maximum in the measured FDCS is much closer to the direction of $\mathbf{q}$ than to 0°.

Some discrepancies between experiment and the M3DW-EIS model, although not nearly as large as in the present study, was also found in the FDCS for $\varepsilon = 30$ eV [12]. In that work we considered the possibility that the capture channel, which is not accounted for in the calculation, may be responsible at least for part of the discrepancies. Electrons promoted to the continuum by a primary interaction with the projectile can eventually get captured to the projectile by subsequent interactions. However, the theoretical model does not contain bound projectile states and all electrons removed from the target therefore must remain in the continuum. The capture probability steeply increases with decreasing relative velocity between the electron and the projectile. For $\mathbf{v_{el}} \approx \mathbf{v_p}$ (i.e. $\varepsilon = 57$ eV and $\theta_{el} = 0$) this probability could saturate, leading to a strong depletion of the FDCS for ionization in this kinematic regime. This would explain the absence of the 0° peak in the experimental data. In order to test this explanation, a non-perturbative coupled-channel calculation, using a two-center basis set, would be very helpful. Since such a model would account for the capture channel, the 0° peak should be at least strongly reduced.

A similar feature as in the present data, with reversed roles between experiment and theory, was found in the FDCS for 3.6 MeV amu^{-1} Au^{53+} + He collisions [6]. There, a pronounced 0° peak was observed in the measured FDCS in addition to the binary peak, which was completely absent in the calculations. One important difference was that the speed of the ejected electron was much smaller than the projectile speed. Nevertheless, due to the very large projectile charge, PCI played a similarly important role as in the present data. However, the electron flux in the 0° peak probably did not get depleted as strongly by the capture channel because of the large relative speed of about 10 a.u. between the electron and the projectile. On the other hand, based on this argument, the missing capture channel in the perturbative models should not pose a serious problem and the question arises why the 0^o peak could not even qualitatively be reproduced by theory. We cannot offer a definite answer to this question; however, we allude to the extremely large perturbation parameter (projectile charge to speed ratio ≈ 4.4) for this collision system, for which the validity of perturbative approaches is not clear. For example, the importance of PCI may be severely underestimated.

Another possible explanation for the missing 0° peak in the present measured FDCS is related to a study by Shah *et al*, where the shape of the cusp peak was studied for low-energy p + H_2 and p + He collisions [36]. They found that the cusp peak was shifted in position to an electron speed that was about 5%–10% smaller than v_p. They interpreted this shift as being due to a post-collisional interaction of the residual target ion with the ejected electron (which we refer to as target PCI), which acts in the opposite direction to the projectile

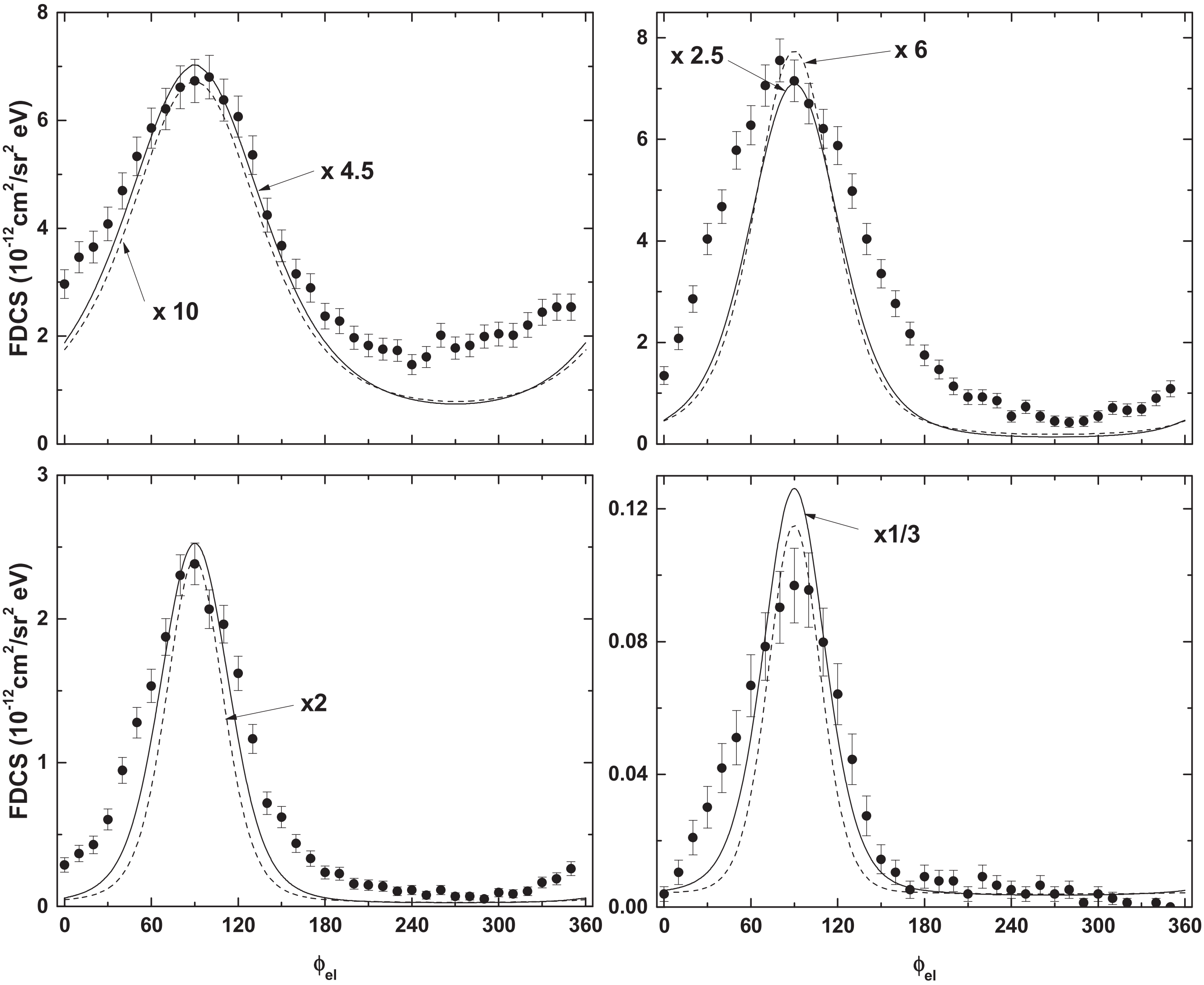

Figure 2. FDCSs for electrons with an energy of 41.6 eV ejected along the surface of a cone with an opening angle of 35° as a function of the azimuthal electron emission angle. The projectile scattering angle was fixed at the same values as in figure 1. Solid and dashed curves as in figure 1.

PCI. This conclusion was disputed by Sarkadi and Barrachina [37]. They argued that the bulk of the shift can be explained by the finite acceptance angle of the electron analyzer and misalignment effects. However, to the best of our knowledge the question of whether the target PCI can lead to a significant shift of the cusp peak has never been conclusively settled. In our case v_{el} is actually slightly larger than v_p (by about 1%), which according to the data of Shah *et al* would already be outside the (shifted) cusp peak. However, it should be noted that in the present study v_p was nearly a factor of 2 larger and any shift of the cusp peak due to the target PCI should be reduced. Nevertheless, we cannot entirely rule out that the target PCI may also contribute to a suppression of the 0° peak.

In order to test how sensitive the shape of the FDCS is to a shift of the cusp peak, we performed the CDW-EIS calculations also for $\varepsilon = 59$ eV, which corresponds to a change in the ejected electron speed by only 2%. The results are shown in figure 1 for $\theta_p = 0.55$ mrad as the dash-dotted curve. Relative to $\varepsilon = 57$ eV the 0° peak is reduced by almost a factor of 3, while the binary peak is slightly increased. For the other θ_p similar differences between the FDCSs for these two energy losses were found. Therefore, if the target PCI shifts the cusp peak to smaller electron energies, as asserted by Shah *et al*, then it could sensitively affect the shape of the FDCSs for electron speeds close to the projectile speed. However, the vast overestimation of the 0° peak for $\varepsilon = 57$ eV would then suggest that either the shift of the cusp peak is not accurately described by CDW-EIS or this model would probably predict an even larger 0° peak at the actual cusp peak energy.

In figure 2 the FDCSs are shown for fixed $\theta_{el} = 35°$ and the same values of θ_p as in figure 1 as a function of the azimuthal electron ejection angle ϕ_{el}, where $\phi_{el} = 90°$ coincides with the azimuthal angle of **q**. Since $\theta_{el} = 35°$ is close to the polar angle of **q**, at least for $\theta_p = 0.2$ and 0.325 mrad, the data points at $\phi_{el} = 90°$ are close to the binary peak. For all θ_p the entire data sets of figure 2 are far from the 0° peak predicted by theory. Therefore, for this geometry the comparison between measured and calculated FDCSs (the same calculations are shown by the same curves as in figure 1) shows how well the collision dynamics is described by theory in a regime that should not be as strongly affected by the capture channel as the forward direction. Significant discrepancies, especially to the CDW-EIS calculations, are still present, but they are not as severe as in the scattering plane. For the same geometry, but an energy loss of 30 eV, the discrepancies, especially in the magnitude, were significantly smaller [12, 34]. This hints that even far away from the 0° peak the capture channel could present a problem to perturbative calculations.

In summary, we have presented the first fully differential comparative study between experiment and theory on target ionization by ion impact in the regime of velocity matching of the ejected electron with the projectile. Remarkable discrepancies between experiment and theory and between two conceptually similar theoretical models were found. This indicates a large sensitivity of the cross-sections to the details of the collision dynamics in this regime. A possible explanation for the discrepancies between the measured and calculated cross-sections is a strong coupling between the ionization and capture channels, which is not accounted for in both theoretical models. This should be particularly important for electrons ejected to a state just above the continuum limit of the target, but our data suggest that it may still be significant far away from this regime.

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