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Manipulating Atomic Fragmentation Processes by Controlling the Projectile Coherence

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We have measured the scattering angle dependence of cross sections for ionization in $p + \text{H}_2$ collisions for a fixed projectile energy loss. Depending on the projectile coherence, interference due to indistinguishable diffraction of the projectile from the two atomic centers was either present or absent in the data. This shows that, due to the fundamentals of quantum mechanics, the preparation of the beam must be included in theoretical calculations. The results have far-reaching implications on formal atomic scattering theory because this critical aspect has been overlooked for several decades.

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When Rutherford introduced the concept of a scattering cross section [1], he had in mind a quantity which only depended on the properties of the colliding particles and the collision energy, but not on the experimental conditions such as the target density or the preparation of the projectile beam. However, since the advent of quantum mechanics, we know that an experiment providing information about the system of interest generally alters the system through the observation process. In a strict sense it is thus not possible to define an observable quantity which only depends on the properties of the system, but not on the observation process. Here, we are particularly interested in the consequences of these properties of quantum mechanics for scattering theory, which, in turn, directly deals with the fundamentally important and yet unsolved few-body problem (FBP) [2,3].

One implication of the above analysis for scattering theory is that the projectile should be represented in terms of a three-dimensionally localized wave packet with finite width which depends on the preparation of the beam. This is, however, a challenging task. Therefore, as an approximation the projectile is usually described as a delocalized particle [4], for example, in terms of a plane wave in the Born expansion [5]. In the vast majority of collision experiments analyzed so far this seemed to be a very well justified approximation. For electron impact collisions, for example, the width of the projectile wave packet is almost always large compared to the target dimension. But for ion impact the width of the wave packet can become similar or small compared to the target dimension for large collision energies. However, the projectile parameters which would be sensitive to the beam preparation (scattering angle and energy loss) are very difficult to measure directly and those experiments which determined the scattering angle under these conditions measured single differential cross sections (e.g., [6]). Such data are probably not sufficiently sensitive to reveal any influence of the finite width of the projectile wave packet on the cross sections.

Indeed, for decades of atomic collision research, the assumption of a delocalized projectile did not seem to pose a significant problem.

With the development of cold target recoil-ion momentum spectroscopy (COLTRIMS) [7,8], the sensitivity at which theoretical models can be tested has been significantly enhanced. More specifically, COLTRIMS made possible the measurements of fully differential cross sections (FDCS) for target ionization for the complete three-dimensional space (e.g., [2,9–11]). These studies revealed unexpected discrepancies between experiment and theory, which were particularly surprising for small perturbations η (projectile charge to speed ratio). There, even the first Born approximation (FBA) was believed to provide a good description of the ionization process. The FBA strictly demands that the fully differential angular distribution of ejected electrons must be cylindrically symmetric about the momentum transfer vector \mathbf{q} (difference between the initial and final projectile momenta) (e.g., [10]). In the experiments, in contrast, clear signatures of a breaking of this symmetry were observed. These qualitative discrepancies persisted even in nonperturbative approaches [12]. Even more surprising, an FBA calculation convoluted with classical elastic scattering between the projectile and the residual target ion, where the projectile was completely localized as far as the projectile–residual target ion interaction is concerned, reproduces the data very well [13]. These observations suggest that the difficulties of the fully quantum-mechanical calculations for ion impact originate, at least partly, from the assumption of a delocalized projectile. This shortcoming of formal scattering theory has been completely overlooked for decades.

In this Letter we report experimental evidence that the localization of the projectile can have a significant and qualitative impact on atomic collision cross sections involving ionic projectiles. An atomic collision version of Young's double slit experiment was performed. Diffraction of a proton beam from the atomic centers of H_2 was studied

in ionizing collisions. Depending on the coherence of the incoming projectile beam an interference pattern was either present or absent in the scattering angle dependence of the ionization cross sections. These results show that major parts of formal ion-atom scattering theory have to be revised.

To observe an interference pattern requires that the incoming projectile wave is coherent in two respects: first, since the phase difference between the waves diffracted from the atomic centers depends on the proton energy, the inherent energy spread ε must be sufficiently small. This can be expressed in terms of the longitudinal coherence length $\Delta z \approx (2\Delta p_z)^{-1} = v/2\varepsilon$ [14] (in atomic units a.u.), which must be on the order of or larger than the internuclear separation D . Here, v is the projectile speed. Second, the width of the proton wave packet, its transverse coherence length Δr , must be large enough to illuminate both atomic centers simultaneously; i.e., Δr must also be larger than D . Δr can be manipulated by a collimating slit of width a at a distance L before the target region and is of the order of $\lambda L/(ka)$ (e.g., [15,16]). Here, λ is the de Broglie wavelength and k is a dimensionless constant which depends on the shape of the projectile wave packet. For a Gaussian wave packet $k = 2\pi$, but $k = 1$ [15] and $k = 2^{1/2}/\pi$ [16] have also been used in Δr . Here, we assume $k = 3$, as an approximate average of these values, to estimate Δr . If L is small enough so that $\Delta r < D$ only one proton in the molecule is illuminated at a time. The projectile is then scattered incoherently and no interference structure is expected. If, on the other hand, L is large enough so that $\Delta r > D$ the projectile is coherently scattered, which can result in an observable interference pattern. Such structures have been predicted several decades ago [17] and reported recently [18,19]. [Interferences in the electron energy spectra due to coherent ejection from the two atomic centers were also reported (e.g., [20,21]).]

In the experiment, a 75 keV proton beam, with an energy spread much smaller than 1 eV, was crossed with a neutral molecular hydrogen beam. The projectile beam was collimated by a set of slits 0.15 by 0.15 mm in size located at a variable distance L before the target region. The recoiling H_2^+ ions were extracted by an electric field of about 50 V/cm and detected by a channel-plate detector. The scattered protons passed through a switching magnet, to separate them from neutralized projectiles, and decelerated to 5 keV. The projectiles were then energy analyzed by an electrostatic parallel-plate analyzer and detected by a two-dimensional position-sensitive channel-plate detector. The entrance and exit slits were long (approximately 2.5 cm) in one direction (the x direction), but narrow (75 μm) in the y direction, which is in the plane of dispersion. Therefore, all ionization events leading to scattering angles between 0 and 1.5 mrad were recorded simultaneously. Data were taken for a fixed projectile energy loss ΔE of 30 eV where a pronounced interference structure was observed earlier

[19]. The projectile detector was set in coincidence with the recoil-ion detector.

Data were taken for two different slit-target distances L under otherwise identical experimental conditions. The larger distance, $L = 50$ cm, corresponds to a transverse coherence length of $\Delta r \approx 2.2$ a.u., which is larger than the internuclear separation in the molecule ($D = 1.4$ a.u.). Therefore, for this L the projectile beam is coherent. An incoherent projectile beam is realized with the smaller distance of $L = 6.5$ cm, corresponding to $\Delta r \approx 0.3$ a.u. The angular resolution for the projectiles was measured for both L with the target gas taken out and the energy analyzer set for $\Delta E = 0$. The same angular width (0.1 mrad full width at half maximum FWHM) was found for both L . Furthermore, the effect of the resolution in angle and energy-loss (3 eV FWHM) on the measured cross sections was tested using a Monte Carlo simulation [22]. Only at angles smaller than 0.1 mrad an observable, but small effect was found.

At smaller pass energies of the projectile energy analyzer than used in this experiment a resolution of less than 1 eV FWHM is achieved. Using this as an upper limit for the energy spread of the proton beam, the longitudinal coherence length is more than an order of magnitude larger than D ; i.e., longitudinal coherence is always realized, regardless of L . However, to see interference in the θ dependence of the ionization cross sections requires both transverse and longitudinal coherence, so that no interference structure is expected at the small L .

Since the x position on the projectile detector defines the scattering angle θ and data were taken for a fixed ΔE , the coincident projectile position spectrum is directly proportional to the double differential cross section $\text{DDCS} = d^2\sigma/d(\Delta E)d\Omega_p$ for target ionization. The data were normalized to the single differential cross section $d\sigma/d(\Delta E)$ calculated using the semiempirical model by Rudd *et al.* [23]. These normalized DDCS are shown in Fig. 1 as a function of scattering angle for $L_1 = 50$ cm (closed symbols) and $L_2 = 6.5$ cm (open symbols). Significant differences between the data sets for the two distances are quite obvious. At small θ the DDCS for large L (in the following referred to as the coherent data DDCS_{coh}) are about a factor of 2 larger than those for small L (incoherent data DDCS_{inc}), at intermediate θ (≈ 0.2 to 0.8 mrad) DDCS_{coh} drops below DDCS_{inc} by up to a factor of 2, to once again become much larger than DDCS_{inc} at $\theta \gtrsim 0.9$ mrad. Since all experimental conditions apart from L were kept identical for both data sets, these differences clearly demonstrate that L , and therefore the projectile coherence, has a major effect on the angular dependence of the DDCS.

The solid curve in Fig. 1 shows a calculation based on the molecular 3-body distorted wave (M3DW) approach [24]. Like the experimental data, this calculation is averaged over all molecular orientations. Most importantly for the present context, the projectile is treated as fully

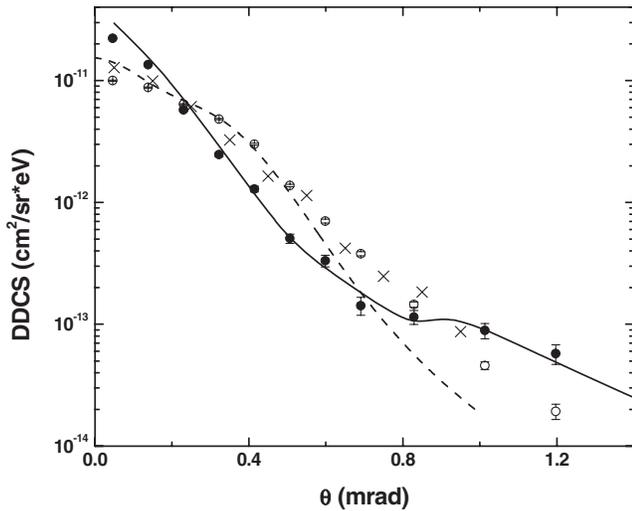


FIG. 1. Double differential cross sections for ionization of H_2 by ion impact as a function of scattering angle for a projectile energy loss of 30 eV. The closed symbols show the data for a large slit-target distance L corresponding to a transverse coherence length Δr larger than the internuclear separation D , the open symbols show the data for small L corresponding to $\Delta r < D$. The crosses are data for ionization of atomic hydrogen [25,26]. The solid curve is a molecular 3-body distorted wave (M3DW) calculation [24], which assumes a completely coherent projectile beam. The dashed curve is a second Born approximation with Coulomb waves (SBA-C) calculation for ionization of atomic hydrogen [26,28].

coherent. This calculation reproduces the measured $DDCS_{\text{coh}}$ very well, but is in poor agreement with the $DDCS_{\text{inc}}$. At the same time, the shape of the angular distribution of the $DDCS_{\text{inc}}$ agrees nearly perfectly with the $DDCS$ for atomic hydrogen ($DDCS_{\text{H}}$) measured earlier [25,26] and which are shown as crosses in Fig. 1. The $DDCS_{\text{H}}$ were multiplied by 2 to account for the presence of 2 electrons in H_2 . (The data in Refs. [25,26] were not normalized to calculated $d\sigma/d(\Delta E)$ of Rudd *et al.*, but to experimental values by Park *et al.* [27]. As a result, the $DDCS_{\text{H}}$ shown in Fig. 1 divided by 2 differ by about 25% from the data of Refs. [25,26].) The shape of the angular dependence of the $DDCS_{\text{inc}}$ is also well reproduced by $DDCS_{\text{H}}$ calculated using a modification of the second Born approximation, except for large θ (dashed curve in Fig. 1). In this model, which was labeled SBA-C, the projectile is described by a Coulomb wave rather than a plane wave [26,28]. Although this also represents a fully coherent treatment of the projectile, its effect on the $DDCS$ is strongly suppressed, if visible at all, compared to the molecular target. The interference for a molecular target is a particularly prominent manifestation of the projectile coherence, which is obviously not present for atomic hydrogen, even if the projectile beam is fully coherent. Furthermore, the ionization potentials of H and H_2 are very similar. Therefore, if $\Delta r < D$, i.e., if only one H atom in the molecule is illuminated at a time, the

ionization process should basically behave like ionization of H and one would expect the $DDCS_{\text{inc}}$ to exhibit the same angular dependence as the $DDCS_{\text{H}}$, assuming that the projectile coherence has no significant effect on the latter. That this is indeed observed supports the conclusion that, at the smaller slit-target distance, it is the incoherence of the projectile beam which makes the $DDCS_{\text{inc}}$ so different from the $DDCS_{\text{coh}}$.

In analogy to optical double slit interference, the $DDCS_{\text{coh}}$ can be expressed in terms of the $DDCS_{\text{inc}}$ multiplied by an interference term (IT); i.e., the interference term is given by the cross section ratio $R = DDCS_{\text{coh}}/DDCS_{\text{inc}}$. The phase difference φ between the projectile waves diffracted at the two centers is a function of θ , the molecular orientation δ , and D . In our experiment δ was not determined and the $DDCS$ therefore have to be averaged over all δ . This averaging leads to a damping, but not to a complete elimination of the interference structure [26,28]. The measured ratio R , i.e., the interference term, is shown in Fig. 2 as a function of the scattering angle. A pronounced maximum can be seen at $\theta = 0$ and a minimum near $\theta = 0.5$ mrad. R then steeply rises again to approach a second interference maximum, which, however, lies only partly within the angular range covered in the experiment. The solid line shows the ratio between the $DDCS_{\text{coh}}$ calculated with the M3DW model (solid line in Fig. 1) and the $DDCS_{\text{H}}$ calculated with the SBA-C model (dashed line in Fig. 1). As mentioned above, the $DDCS_{\text{H}}$ should to a very good approximation exhibit the same θ dependence as the $DDCS_{\text{inc}}$; i.e., like the data this theoretical ratio should represent to a good approximation the interference term. These theoretical R are in excellent agreement with the measured values for scattering angles smaller than about 0.7 mrad, but they considerably overestimate the experimental R at larger θ . However, these

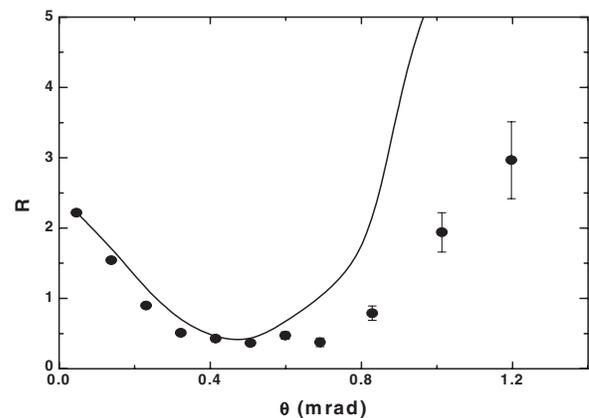


FIG. 2. Ratios between the double differential cross sections for ionization of H_2 for a large slit-target distance (closed symbols in Fig. 1) and for a small distance (open symbols in Fig. 1). The solid curve shows the ratio between the double differential cross sections calculated for ionization of H_2 using the M3DW model and for ionization of atomic hydrogen calculated using the SBA-C model.

discrepancies are not primarily due to an incorrect description of the interference, but mainly result from an underestimation of the experimental DDCS_H at large θ (see Fig. 1). Overall, the interpretation of the differences between DDCS_{coh} and DDCS_{inc} as due to the interference is qualitatively supported by the theoretical R .

In summary, an interference structure due to indistinguishable scattering of a proton beam from the two atomic centers of H_2 was observed if a collimating slit was placed at a large distance from the target, but not for a small distance. We do not consider the presence of interference effects *per se* to be the most significant result. Rather, we believe that the most important conclusion to be drawn from this work is that the preparation of the projectile beam affects the scattering cross sections, not because of imperfections in the experiment, but because of the fundamentals of quantum mechanics. Many decades of atomic scattering theory are based on the assumption that the projectile beam is prepared coherently. In many cases (like, e.g., electron scattering or cross sections integrated over projectile parameters) this assumption may represent a very good approximation; however, it is not sustainable in general. Here, we presented an example, namely, incoherent proton scattering leading to ionization of H_2 , where this assumption leads to qualitatively incorrect results.

Our results demonstrate that the projectile has to be described by a three-dimensionally localized wave packet with finite width. Collision systems involving atomic targets are potentially also significantly affected by the projectile coherence. For example, the long-standing puzzle regarding discrepancies between theory and experiment in the FDSC for ionization in 100 MeV/amu $\text{C}^{6+} + \text{He}$ collisions [2] could probably be solved by properly accounting for the localization of the projectile. More specifically, the incorrect assumption of a fully coherent projectile beam probably leads to artificial path interference between two (or more) different impact parameters, resulting in the same scattering angle, in theory [29].

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