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Two-center interference in p–H$_2$ electron-transfer collisions

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Abstract. We report on measurements of transfer excitation in collisions of 0.3-1.3 MeV protons with spatially oriented H$_2$ molecules. Evidences of two center interference are found in the angular distribution of the molecule after a transfer excitation process and directly in the projectile angular scattering distributions. These features can be explained in a way which is analogous to that for the interferences in Young's classical double slit experiment: The fast projectiles preferentially capture electrons close to either of the molecular nuclei, and thereby they change their momenta and de Broglie wavelengths. The waves emerging from the two 'slits' of the molecule interfere yielding the observed interference structure.

1. Introduction

The phenomenon of interference due to a double slit is a clear manifestation of wave-like properties of radiation. Already in the first decade of the 19th century, Thomas Young carried out his classical experiment [1], showing that the light emerging from two slits produces a characteristic fringe pattern on a screen. This observation contradicted Newton's 'corpuscular' theory but was consistent with a wave description of light. More than a hundred years later, after the advent of quantum theory and the postulation of the wave-particle dualism by de Broglie [2], the double-slit experiment was generally viewed as a "Gedankenexperiment", which if it could be performed would illustrate the consequences of the quantum mechanical wave-like behavior of moving particles like, e.g., electrons [3]. However, even though the wave character of electrons was demonstrated in diffraction experiments on crystal surfaces by Davisson and Germer [4] already in 1927, a real double slit experiment did not seem feasible due to the very short wavelength related to moving particles. As Feynman put it in his 1963 lectures, "the trouble is that the apparatus would have to be made on an impossibly small scale to show the effects we are interested in" [3]. Despite the doubts about the feasibility of such experiments, it was actually first performed already in 1961 for electrons [5], but considerably later (in 1988) for neutrons [6] and not until 1991 for atoms [7].

For diatomic molecules, the interference of particle waves emitted from the two nuclear centers was often compared to the diffraction on a double slit like in Young's experiment. One inherent

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advantage with molecules as particle wave sources is that the internuclear distance, i.e. the separations of the two slits, already are on atomic scales and thus also in the range of the wavelength of an electron traveling with an energy of several eV. Two-center interference effects with molecules have been predicted and observed in the case of photoionization [8, 9, 10] and electron capture [11, 12, 13, 14, 15, 16]. Recently interferences have also been found in the electron spectra due to heavy-ion [17, 18, 19] and electron [20] induced ionization of H_2. Also for double electron capture in He^{2+}-H_2 collisions followed by the autoionization of the projectile, interferences were observed and interpreted as scattering of the emitted electrons on the two remaining protons [21].

Here we present measurements of electron transfer from spatially oriented H_2 to MeV protons. In fast collisions, electron-transfer is related to small impact parameters with respect to the target nuclei. Thus, the waves describing the motion of the outgoing neutralized projectiles are emerging from two small regions close to either nucleus. The superposition of the two projectile de Broglie waves is expected to result in interferences as in Young's optical two-slit experiment.

Electron capture in fast proton-H_2 collisions has been investigated theoretically by Tuan and Gerjuoy [11] and reexamined by other authors (e.g. [12, 13, 14, 22]). In these calculations interference between two capture amplitudes associated with the two centers of the molecule is found, and a phase factor between the two amplitudes

$$\pm \exp\left(\vec{\alpha} \cdot \vec{\rho}\right) \tag{1}$$

is obtained, where $\vec{\alpha} = \vec{k}_f - \vec{k}_i$ is given by the difference between the final ($\vec{k}_f$) and initial ($\vec{k}_i$) projectile momentum and $\vec{\rho}$ is the internuclear vector of the molecule. The $\pm$ sign depends on the symmetry of the electronic wave function in the molecule and corresponds to the gerade and ungerade states, respectively.

The phase factor in (1) is precisely the factor which is expected in the above mentioned picture where interference is obtained in terms of two projectile de Broglie waves emerging from locations in close vicinities to the molecular nuclei. In the present work, we report on measurements of the projectile angular scattering distribution (which relates to $\vec{\alpha}_\perp$) and the $\vec{\rho}$-dependent total transfer excitation cross section and discuss the resulting interference phenomena in some detail.

2. Experiment

The experiment was carried out at the CRYRING [23] facility at the Manne Siegbahn Laboratory in Stockholm. The protons are injected at $300\,keV$ in the magnetic storage ring, accumulated, accelerated, and electron cooled at the desired kinetic energy (in the present experiments 0.3, 0.7, 1.0, and $1.3\,MeV$). The projectile beam is intersected with a molecular H_2 super-sonic gas jet [24], which provides target densities of about 10^{11} molecules/cm^3 at a temperature of well below $1\,K$ with a background pressure of $10^{-11}\,mbar$ in the storage ring. Neutralized projectiles, which have captured a single electron from a target molecule, leave the storage ring after the next dipole magnet and hit a position sensitive micro-channel-plate detector situated $3.2\,m$ behind the reaction region. The projectile deflection angle is determined from the position on the detector. Positive ions from the target are detected in coincidence with the neutralized projectiles with a COLTRIMS (COld Target Recoil Ion Momentum Spectroscopy) setup [25].

A drawing of the COLTRIMS set up is shown in figure 1 (details are found in [16, 26, 27]). The recoiling target ions are extracted by means of a homogeneous 20 V/mm electric field, perpendicular to the projectile and target beams. After passing a field-free drift region the recoil ions are detected with a second micro-channel plate with a position sensitive resistive anode. The momenta of the target ions along the extraction direction can be calculated by their time of flights, while the momentum components in the other dimensions are determined by the positions on the detector.
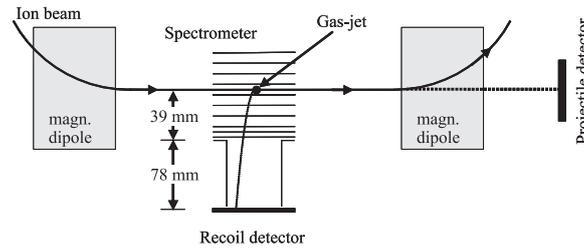
Figure 1. Experimental set up (see text).

In the density plot of figure 2 (a), the number of events is shown as a function of the time of flight and the distance r from the center of the distribution on the detector for 1 MeV proton-H_2 collisions. In this figure several regions of high intensity can be identified. There is a distinct, narrow maximum at a time of flight close to 575 ns with a small spatial width at the detector, i.e. with a narrow distribution in r. This peak is due to the capture of one electron in the collision between the projectile and the target creating a non-dissociating H_2^+ molecular ion (which thus is in its electronic ground state, $1s\sigma_g$). There is another spatially and temporally narrow peak at a time of flight of about 410 ns. This peak is due to capture of one electron, where the H_2^+ molecule is excited to the vibrational continuum of the $1s\sigma_g$ state. This results in dissociation processes with low kinetic energy releases (typically < 1 eV).

In the same figure, a half-ring shaped region of high intensity is also centered at flight times of 410 ns, with distances from the center of the position distribution up to $r > 17$ mm. Here the molecule is excited to one of several possible repulsive states resulting in kinetic energy releases of typically 3 to 15 eV. From previous studies on dissociative collisions involving H_2^+ [28, 29] it may be expected that the molecular ion dissociates predominantly via the $2p\sigma_u$, $2p\pi_u$, and $2s\sigma_g$ states. Also transfer ionization processes in which both electrons are removed promptly from the target yield protons in a similar energy range and may contribute to the data with up to about 20 % [30].

The direction of the momentum vector of a proton, which is emitted in a fast dissociation process, reveals the orientation of the molecular axis at the instant of the collision, as two
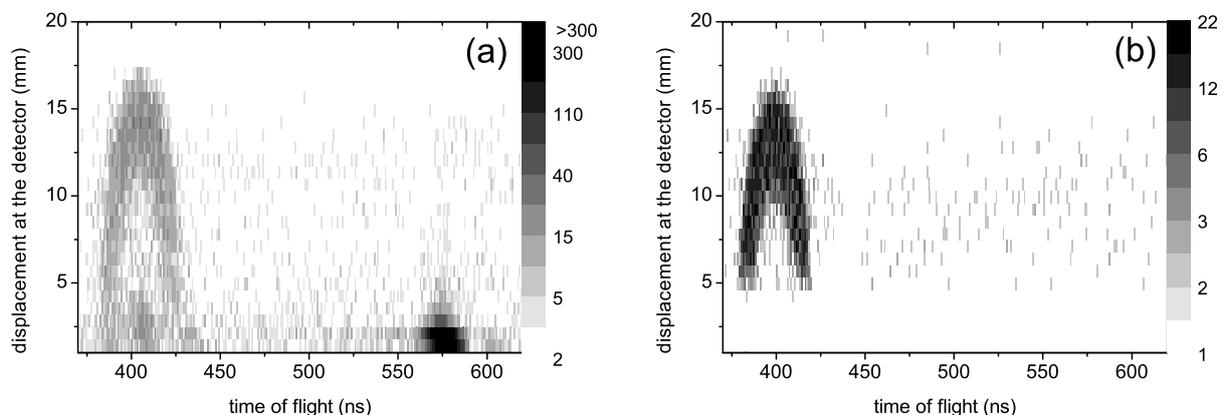
Figure 2. Intensity plots of the recoil ion yields as function of their time of flights and their radial positions r on the detector (with respect to the center of the position distribution). (a) is for 1 MeV proton-H_2 collisions, and (b) is for 1.3 MeV protons using a foil in the center of the recoil ion detector (see text).

conditions are fulfilled in the present energy range: (i) The recoil ion momentum, which is transferred from the projectile to the target system, is much smaller than the momentum which the proton gains through the dissociation process itself, and (ii) the molecular rotation is much slower than the dissociation process. As the projectile velocity is much higher than that of the vibrational motion, the two nuclear centers of the molecule can be regarded as having fixed positions during the collision process.

One experimental difficulty lies in the fact, that the cross section of electron capture is much smaller than the total electron loss probability of the target (dominated by ionization without electron capture) in the present energy range (about 5 orders of magnitude at a projectile energy of 1 MeV). In order to avoid random coincidences, the experiment would in principle have to be operated with low recoil ion count rates. However, we avoid such luminosity limitations by means of a foil in the center of the recoil ion detector. In this way, the detection of low-energetic recoil ions, which are mainly produced in direct target ionization events, is strongly suppressed (see figure 2 (b)). Protons emitted due to fast dissociation processes are only masked out by the foil when the molecule is oriented along the extraction direction. The data for this orientation may, however, be reconstructed due to the cylindrical symmetry of the experiment with respect to the projectile beam direction.

3. Results and discussion

The experiment has been performed for four projectile energies between 0.3 and 1.3 MeV. In figure 3, the total cross section for transfer excitation leading to H_2^+ dissociation is plotted as a function of the angle ϑ between the molecular axis and the projectile beam direction. For all four energies, the experimental data features significant variation of the cross section with respect to the orientation of the molecule and there are maxima at 90° where the molecule is oriented perpendicular to the projectile beam. These maxima become more narrow with increasing projectile velocities and for the highest energy secondary maxima are measured for molecular orientations along the projectile beam.

These results may be qualitatively understood by considering changes in projectile momenta and de Broglie wavelengths at two spatially well defined regions around the nuclei of the H_2 target molecules: For electron capture at high velocities v_p the longitudinal projectile momentum transfer is $\alpha_\parallel = v_p/2 - Q/v_p$, where the change of binding energies is Q (cf. e.g. [25]). According to expression (1) there is a longitudinal phase shift of $\alpha_\parallel \rho_\parallel = \alpha_\parallel d \cos \vartheta$ (d is the internuclear separation of the molecule) between the two projectile waves emerging from the nuclear centers of the molecule. For small values of the transverse projectile momentum change $\alpha_\perp$ (the main contribution to the capture cross section at high velocities) the superposition of these waves results in constructive interference when the phase shift $\alpha_\parallel \rho_\parallel$ is equal to 0 (or to n2π where n is an integer). This is the case, e.g. when the molecular axis is perpendicular to the projectile beam and for such orientations maxima in the cross sections are observed. For molecular orientations other than 90° the phase shift is $\alpha_\parallel \rho_\parallel \neq 0$. Destructive interference then occurs when the phase shift is equal to π (or to $(2n+1)\pi$ where n is an integer). For a projectile energy of 1.3 MeV, for instance, this is fulfilled for a molecular angle ϑ of about 50°, where also a minimum in the experimental cross section is observed. Thus, this simplified picture describing the projectile final state as the superposition of two phase shifted outgoing waves readily accounts for the angular locations of the maxima and minima in the experimental cross sections. Note that this reasoning only considers the longitudinal phase shift and the scattering in the forward direction. A somewhat more detailed discussion would need to consider also transversal phase shifts (i.e. outgoing spherical rather than only planar projectile waves). Still, the simplified discussion yields the main experimental features.

In figure 3, the experimental results are compared to the model from [14] in which the molecule is described by a linear combination of atomic orbitals (LCAO) at a fixed internuclear distance

of 1.4 a.u. For the capture process, atomic Brinkman-Kramers amplitudes are used. If the atomic transfer probabilities are added incoherently, no angular dependence of the cross section appears in their calculations as indicated by the dotted lines in the graphs. This indicates again, that the present oscillations in the experimental transfer excitation (and dissociation) cross sections are due to quantum mechanical interferences and independent of trivial geometrical effects. The full curves show, as comparisons, the calculated cross sections for single-electron capture (without excitation and dissociation of the H_2^+ molecule) in which the interference term is included. Here strong angular variations are observed and the general agreement with the present experimental data is good. This agreement becomes better also on a quantitative level when the projectile energy is increased as expected from a theoretical point of view (the Brinkman-Kramers approximation is better fulfilled at higher velocities).

The comparison of experimental results on transfer excitation with theoretical results on single-electron capture naturally raises the question as to what extent the excitation of the molecular ion contributes to the orientation dependence. This question was analyzed in detail in [22] where this two-electron process was treated as a two-step process in the independent-electron approximation. While it was found that the excitation process does depend on the molecular orientation, the final result was that the expected orientation dependence is very similar for single-electron capture and transfer excitation. The authors of [22] further pointed out that the inclusion of electron correlation effects would allow for a one-step mechanism in which the H_2^+ ion is promoted to an ungerade excited dissociating state. Interestingly, such a mechanism would give a reversed molecular-alignment dependence with respect to that of the present results, to that of an earlier study on O^{8+}-H_2 collisions [15], and to that of the
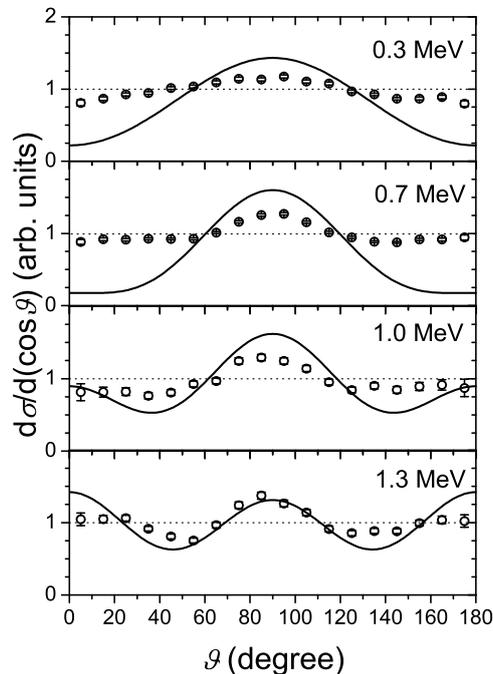
Figure 3. Cross sections for electron transfer with subsequent fast dissociation of H_2^+ as functions of the initial H_2 orientation. ϑ is defined as the angle between the molecular axis and the projectile beam. Circles are experimental data from [16], the curves show results using the model for single electron capture (without dissociation) by [14] including (solid curves) and excluding (dotted curves) the interference term.
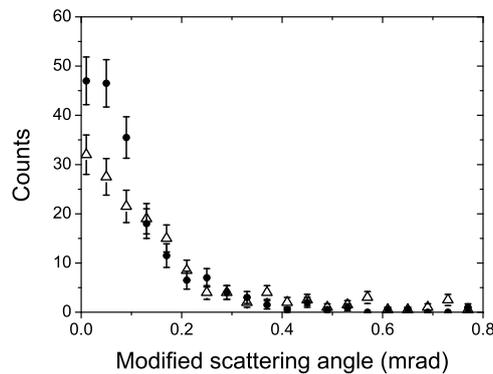
Figure 4. Modified projectile angular scattering distributions (see text) for a molecular orientation of $90°\pm5°$ (filled circles) and $50°\pm5°$ (open triangles).

independent-electron approximation. In fact, already the authors of [22] concluded that the experimental results from [15] are consistent with the independent electron excitation picture, and such a conclusion could apply also to the present results.

In the simplified picture above, the projectile final state wave packet is discussed in terms of a superposition of two plane waves. However, the waves emerging from the two molecular centers should be better described as being spherical. The superposition of two spherical waves in turn gives rise to an interference pattern at the projectile detector as a function of the scattering angle [12] similar to the results of the original optical double-slit experiment by Young. The direct observation of the interference of the outgoing projectile waves requires a very low beam emittance, as the projectile scattering angle between two bright fringes may be very small (0.34 mrad is expected for a projectile energy of 1.3 MeV).

In the case of one run at 1.3 MeV, a very narrow projectile beam was achieved and a width of 0.9 mm was obtained on the projectile detector 3.2 m behind the reaction region. This corresponds to an upper limit of the angular resolution of 0.28 mrad. In figure 4 the cross section is shown as a function of the modified scattering angle, which is the angle between the projectile axis and the projection of the projectile final state momentum vector onto the plane of the molecular axis and the projectile beam. This corresponds to a projection of the two-dimensional diffraction pattern on the axis perpendicular to the fringe structure and in this representation oscillations due to interference should in principle be visible. The experimental data plotted in this graph contains events with kinetic energy releases below about 16 eV, due to limitations in resolution at higher energies (trajectories far from the center in the time-of-flight spectrometer).

For a molecular orientation of $90°$ (filled circles in the graph) there is a maximum in the forward direction, i.e. at a projectile scattering angle of 0 mrad. No secondary maximum is observed, where the first bright fringe would be expected (at 0.34 mrad). This is, however, due to the fact that the cross section decreases strongly with increasing scattering angle such that there are hardly no counts in this angular range at the present level of statistics. For molecular angles of about $50°$ (open triangles), where the destructive interference is observed in the total cross section (cf. figure 3), there is a maximum expected at scattering angles of about 0.15 mrad while there should be a minimum in the forward direction. Again there are no distinct corresponding structures in the cross section which is mainly related to the experimental resolution, the integration over a range of angular orientations of the target molecule ($\pm5°$) and possibly also due to variations in the actual internuclear separations in the H_2 molecule due to vibrations. Still, the data for the two molecular orientations are distinctly different. For molecular orientations of $50°$ the data is about 30% wider than in the $90°$ case. This effect is

evidence of two-slit interference in direct analogy to Young's optical experiment.

4. Conclusion

We have measured the transfer excitation process in collisions of 0.3-1.3 MeV protons and H_2 molecules. Both the molecular orientation and the projectile deflection angle have been determined experimentally. In the experimental cross section, a strong dependence on the molecular orientation is found. This can be understood in a simple picture as the result of quantum mechanical interference of the two projectile de Broglie waves emerging from the molecular nuclei. The experimental results have been compared to a model developed by Wang *et al* [14] and qualitative agreement has been found. Further, the projectile angular distributions for two different molecular target orientations have been measured. The comparison of these angular distributions shows in a direct way the effect of quantum mechanical interference at the projectile detector.

It is worth mentioning, that the two-slit scenario of the experiment described here is very different to those of ionization experiments of molecules found e.g. in [17, 18, 19]. In the latter cases the wavelength λ and the slit distance d are typically of similar magnitude. Constructive interference occurs for $\varphi = n \cdot \lambda/d$ (n can be any positive integer or 0), thus, maxima are expected at angles φ in the order of radians. In our experimental situation, however, the de Broglie wavelength of the projectile is more than three orders of magnitude smaller than the slit distance i.e. the internuclear separation of the molecule. This leads to interference fringes which are very close to each other in the range well below 1 mrad. The variation of the total cross section with the molecular orientation could still be observed in the present experiment, as the product of the longitudinal projectile momentum change $\alpha_\parallel$ and the internuclear distance d is of the order of π.

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