PHYSICAL REVIEW A **72**, 050703(R) (2005)

Two-center interference in fast proton–H_2-electron transfer and excitation processes

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(Received 15 July 2005; published 7 November 2005)

We present experimental evidence for a strong dependence on the angle between the molecular axis of H_2 and the direction of the incoming projectile (p) in the cross section for transfer excitation in fast p-H_2 collisions. For collision energies of 1.0 and 1.3 MeV we find good agreement between the observed data and an analytical expression based on a two-atomic-center description using Brinkman-Kramers amplitudes. This clearly shows that the observed angular dependence is a result of quantum mechanical interference and not a trivial geometrical effect.

DOI: 10.1103/PhysRevA.72.050703 PACS number(s): 34.70.+e, 34.50.−s

Experimental indications and verifications of quantum mechanical interference effects continue to spur considerable interest and discussions, although such effects are well-known consequences of the wave description of matter (which has been around for almost a century). One example of a recent discussion concerns the observation of interference effects in electron emission from *randomly oriented* H_2 molecules in fast charged-particle collision [1–7]. In this Rapid Communication we report on collisions between fast protons and *fixed-in-space* H_2 where we observe strong interferences of electron-transfer amplitudes for the two centers leading to molecular-orientation dependences in the electron transfer cross section. In 1960 Tuan and Gerjuoy [8] formulated the problem of electron transfer in atom-molecule collisions in terms of a separated-atoms picture with interfering electron-transfer amplitudes. A similar description for scattering amplitudes was employed by Deb *et al.* [9] in 1988 considering the "two-slit" problem of high-velocity electron capture from an oriented hydrogen molecule for which the projectile has to pass close by one of the target nuclei [10]. Shingal and Lin [11] used a semiclassical approach in combination with close-coupling electron transfer amplitudes to calculate strong variations in electron transfer cross sections as functions of the molecular orientations for 1–500 keV. Later Wang, McGuire, and Rivarola [12] deduced an analytical expression, based on Brinkman-Kramers amplitudes [13], and valid for high energies ($\geqslant$1 MeV).

Due to the very small electron-transfer cross sections at high energies [14], experiments are difficult and the only earlier investigation of the molecular orientation dependence was performed with O^{8+} increasing the count rate by a large factor [15] in relation to the proton projectile case. However, using O^{8+} instead of protons makes comparisons with model calculations that rely on small values of the projectile charge-to-velocity parameter Z_p/v_p, more difficult. Furthermore capture to excited projectile states becomes important as the projectile charge is increased [16]. In the present paper we have utilized the unique properties of the ion storage and cooler ring CRYRING with its internal gas-jet target to improve the luminosity by orders of magnitude compared to earlier studies (c.f. [17–20]). This has allowed us to investigate two-center interference effects in electron-transfer for p H_2 with $Z_p=1$ and $v_p/v_0=3.5-7.2$ (v_0 is the Bohr velocity).

Protons were injected at 300 keV and accelerated to energies of 300–1300 keV in CRYRING [21] at Stockholm University. The stored ion beam was electron cooled in order to reduce the velocity spread and the geometrical beam width (to ~1.5 mm).

The ion beam intersected a vertical gas jet at 90°. In order to increase the density in the target, the gas was cryogenically precooled to 165 K, and the driving pressure (2.0 bar) and temperature were chosen in order to avoid cluster formation in the supersonic expansion from the 30-μm nozzle. The gas jet was collimated to a diameter of 1.3 mm at the position of the stored ion beam and did not cause a measurable increase in the 1×10^{-11} mbar background pressure in the storage ring [17].

The collisions took place in the extraction stage of a recoil-ion-momentum spectrometer [22,23] with its axis perpendicular to the ion beam and the gas jet. A position-sensitive recoil-ion detector was mounted at the end of the spectrometer (see Fig. 1). The spectrometer voltages were set in order to collect and detect all ions with kinetic energies up to 12 eV. The fast neutral hydrogen atoms formed in electron-transfer collisions were detected by a second position-sensitive detector 3.2 m downstream of the gas target (Fig. 1).

We recorded recoil ions (H^+ and H_2^+) in coincidence with neutralized fast projectiles (H^0). The latter started a multihit time-to-digital converter (TDC) that registered the recoil-ion (H^+ and H_2^+) arrival times. For each event, the position of the first hit was recorded. An analog time-to-amplitude converter (TAC) was used for a complementary high-resolution

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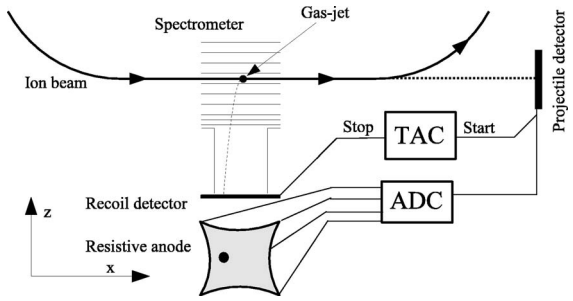

FIG. 1. The experimental setup (see text).

timing measurement for the first recoil ion. All parameters (times and positions) were stored in list mode for later analysis.

We deduced the full three-dimensional momentum vectors (mv_x, mv_y, mv_z) for the first-hit recoil ions, using the position on the recoil detector with respect to the spectrometer axis and the time of flight. The spectrometer was designed so that the axial momentum is linearly proportional to the shift in arrival time with respect to the center of the time-of-flight peak. From the velocity vector the angle between the molecular axis and the ion beam was found.

In the present energy range (0.3–1.3 MeV), the collision times are much shorter than the vibrational and rotational times of H_2. The molecule is thus frozen during the collision and its orientation at the time of impact is revealed by the orientation of a sufficiently large proton momentum vector. In most electron-transfer collisions, the residual molecular ion remains bound in the $^2\Sigma_g^+$ state (H_2^+). However, if this ion is sufficiently vibrationally excited it may dissociate to a proton and a hydrogen atom (3% probability). Only small amounts of energy are released in such *slow proton* processes and the corresponding intensity will appear close to the center of the recoil detector. In the transfer excitation process in which one electron is captured and one is excited to a dissociating H_2^+ state, each fragment (p and H) gets a kinetic energy of 8.5 ± 1.6 eV (starting from the H_2 vibrational ground state). Such *fast protons* allow the determination of the angle between molecule and projectile at the moment of impact.

In Fig. 2 we show a typical time-of-flight (TOF) spectrum (upper part) with a broad proton peak, a narrow H_2^+ peak, and a two-dimensional plot where the TOF is combined with the vertical position on the recoil detector. The main features of Fig. 2 are: (1) Fast protons, which are covering a large part of the area of the recoil detector due to large kinetic energies from excited H_2^+ dissociation. (2) Slow protons, giving the maximum in the middle of the proton peak. (3) H_2^+ in coincidence with neutralized projectiles due to single-electron capture events. (4) Random coincidences between H_2^+ ions with low energies and uncorrelated projectile detector events. In Fig. 3, showing densities in a radial-to-axial two-dimensional velocity plot, we see that the fast protons in the semicircles are clearly separated from the slow protons near the origin.

The experiment was performed at 0.3, 0.7, 1.0, and 1.3 MeV. The relative cross sections as functions of the angle between the internuclear H_2 axis and the direction of

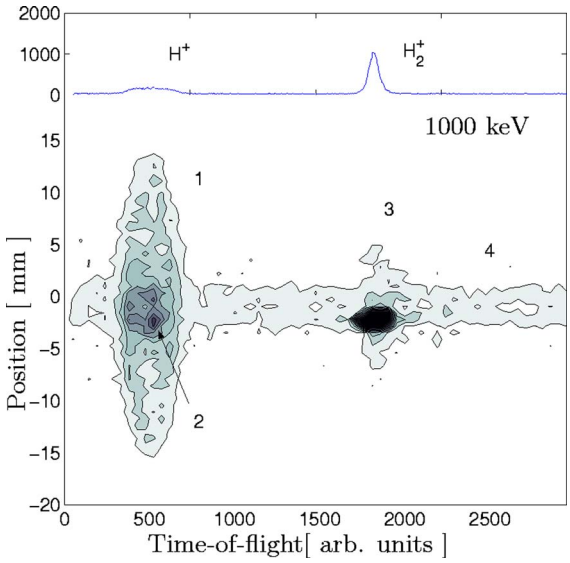

FIG. 2. (Color online) Two-dimensional intensity plot for the vertical recoil-ion position vs time of flight. On top: The projected time-of-flight spectrum.

the projectile are shown in Fig. 4. We find significant dependences of the cross sections on this angle for all four energies with maxima at 90°. The maxima become more narrow with increasing energies, and at the highest energies (1.0 and 1.3 MeV) secondary maxima are found for molecules aligned with the beam direction. In Ref. [12], a method to calculate electron capture from oriented H_2 molecules by coherent addition of two atomic electron transfer probabilities is described. We follow this procedure to calculate the angular dependence at 0.3, 0.7, 1.0, and 1.3 MeV. This calculation only takes the capture part in transfer excitation into account and assumes that the excitation part does not affect the angular dependence.

Before discussing the detailed comparison between theory and experiment it is worth noting that, if the contributions to the transfer probabilities from the two atomic centers would be added incoherently, no angular dependence would appear and the angular differential cross section would be indepen-

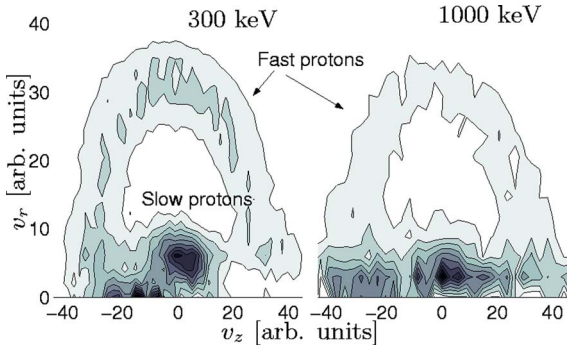

FIG. 3. (Color online) Two-dimensional density plots showing the correlations between radial and axial velocities at 0.3 and 1.0 MeV for fast protons (the ring shapes), slow protons (close to the center), and randoms distributed along v_z with small v_r values. The latter are clearly visible at 1.0 MeV.

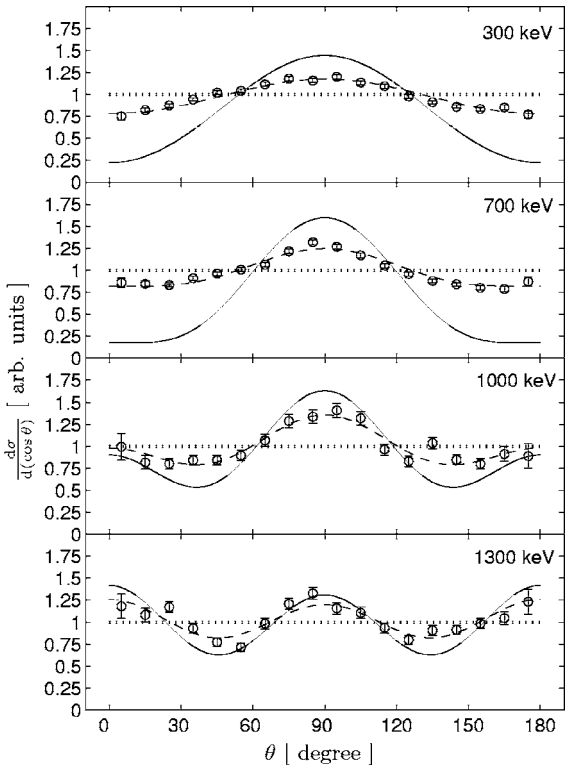

FIG. 4. Angular differential cross sections for transfer excitation in 0.3, 0.7, 1.0, and 1.3 MeV proton-H_2 collisions as functions of the angle θ between the axis of the H_2 molecule and the projectile trajectory. The full curves are the present evaluations within the theory by Wang, McGuire, and Rivarola [12], while the same results but with added (different for different energies) angular independent components are shown as dashed curves. The dotted lines are the cross sections (independently set to 1 in the four panels) for incoherent addition of the transfer amplitudes of Ref. [12].

dent of θ. This is shown by the dotted lines for which $d\sigma/d(\cos\theta)$ are independently set to unity in each one of the four panels of Fig. 4. Thus *all* the observed variations are consequences of quantum mechanical interferences. The calculation using the method of Ref. [12], represented by the full curve of Fig. 4, shows the same qualitative feature and variation with projectile energy as the experimental data. However, quantitative agreement is only found at the highest energies.

It is not surprising that the quantitative agreement improves with increasing collision energy, as the calculation is based on a perturbative approach assuming capture to the 1*s* ground state of the fast hydrogen atom. While capture to other *s* states only affects the angular dependence weakly through the inelasticity [16], capture to $l \neq 0$ states will have a much more dramatic influence on the result. Somewhat *ad hoc* we assume all contributions with non-*s* symmetries to average to angle-independent contributions and thus make fits to the data combining the results of the model calculations with constant offsets representing different relative $l \neq 0$ contributions for the different collision energies. These results are shown as dashed curves in Fig. 4. Except at our highest energy where the agreement is already quite good without the offset (full curve), the introduction of angular independent offsets improves the agreement tremendously for the lower energies. This illustrates that the qualitative features such as the width of the central peak and the appearances of the secondary maxima are similar in experiment and theory and that a strong and effectively angle-independent mechanism may be involved at the lower energies.

We have extended the available theoretical description by describing the internuclear motion explicitly. Two immediate advantages emerge from this procedure. First, in the earlier descriptions the H_2 internuclear coordinate was assumed to be fixed at the equilibrium value $1.4a_0$ during the collision. While the nuclear motion is slow compared to the projectile velocity and thus the frozen nuclei picture is well justified, it seems a rather drastic approximation to assume that the value always equals the equilibrium distance. Second, our description also allows for the explicit inclusion of the nuclear wave function in the dissociating H_2^+ state as described in [24]. Within this framework, two different mechanisms for transfer excitation were identified. When electron correlation in the H_2 ground state is described in the configuration mixing picture a small amount of doubly excited ungerade-ungerade symmetric states is mixed in. Therefore it is in principle possible to capture an electron from a state that, in an independent-electron description, has ungerade symmetry. In order to conserve the total symmetry of the initial state, the remaining electron is then necessarily promoted to the antibonding excited ungerade state. This possibility was identified and briefly discussed by Wang *et al.* [25]. The other possibility is that a gerade electron is captured and that the excitation to the ungerade dissociating state takes place in an independent projectile-electron interaction as assumed in the earlier calculations. While the latter mechanism predicts an angular dependence rather similar to that of the earlier calculations the one-step mechanism (capture from an ungerade state) would lead to maxima at 45° and 135° instead of at 0°, 90°, and 180° (which we observe). The comparison with the experimental data thus immediately tells us that even at the present high velocities the independent capture and excita-

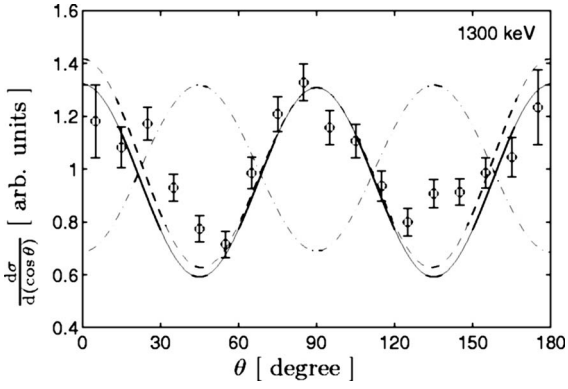

FIG. 5. Angular differential cross sections for p-H_2 transfer excitation in 1.3 MeV collisions as function of the angle θ between the H_2 axis and the projectile trajectory. Open circles: the present experimental results; the dashed curve: theoretical results using Ref. [12]; the full and dash-dotted curves present calculations treating the initial H_2 and final H_2^+ nuclear wave functions for the two-step (full curve) and one-step (dash-dotted curve) transfer excitation mechanisms.

tion mechanism dominates over the one-step mechanism, the effects of which may be detectable only at still higher velocities. In Fig. 5 we show the experimental data together with the results calculated directly from Ref. [12] and with our method taking the distribution of possible internuclear distances in H_2 at the moment of impact into account for the two-step (full curve) and the one-step (dash-dotted curve) transfer excitation processes.

We have measured the cross section for the transfer excitation process in collisions between fast ($v_p/v_0=3.5-7.2$) protons and H_2 as a function of the angle between the molecular axis and the projectile trajectory. The measurements reveal a strong angular dependence, due to interferences between two atomic electron-transfer amplitudes related to the two atomic centers. A comparison with the theoretical description by Wang, McGuire, and Rivarola [12] yields quantitative agreement at the highest energy. At lower energies, where the validity of the perturbative approach is questionable, no quantitative agreement is found. However, the qualitative features and their velocity dependences are reproduced as predicted.

Including the spread in possible internuclear distances at the moment of the collisions gives a minor improvement in comparison with the experimental results. From the phase of the observed interference we conclude that the transfer excitation process is due to independent capture and excitation processes as a one-step process would lead to an inverted interference pattern.

We thank Professor N. Elander for many lively and fruitful discussions. This work is supported by the Knut and Alice Wallenberg Foundation, and the Swedish Research Council (Contracts 2001-2226 and 2002-4693).

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