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Two-Center Double-Capture Interference in Fast $\text{He}^{2+} + \text{H}_2$ Collisions

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We report the first observation of Young-type interference effects in a two-electron transfer process. These effects change strongly as the projectile velocity changes in fast (1.2 and 2.0 MeV) $\text{He}^{2+} + \text{H}_2$ collisions as manifested in strong variations of the double-electron capture rates with the H_2 orientation. This is consistent with fully quantum mechanical calculations, which ignore sequential electron transfer, and a simple projectile de Broglie wave picture assuming that two-electron transfer probabilities are higher in collisions where the projectile passes close to either one of the H_2 nuclei.

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The wave-particle duality is inherently fundamental to quantum mechanics. This is often illustrated by two-slit interference phenomena, where the passage of a specific slit, an aperture or some localized region in space often is linked to particlelike behavior while the interference of probability amplitudes of two paths describes the wavelike behavior. Well-known examples are Young's original two-slit experiment [1] and its followers where the latter have demonstrated interferences of "particles" like, e.g., electrons [2,3], neutrons [4], atoms [5], and molecules [6,7]. Important aspects here are that the interferences only are observed with both slits open and that the interference patterns remain also when the rates are so low that only *single* particles at the time may be inside the apparatus [2,3]. In this Letter, we report the first observation of molecular two-center interference effects for *double*-electron capture (here in fast $\text{He}^{2+} + \text{H}_2$ collisions). In a remarkable way, this also sheds light on the double-electron transfer mechanism itself.

In 2008, we reported on strong interference effects in fast $\text{H}^+ + \text{H}_2$ collisions when *only one* electron is transferred to a fast H^+ ion to form an (almost) equally fast neutral H^0 atom and a very slow, excited, H_2^+ target ion [8]. The orientation of the H_2 molecule at the time of the collision was obtained via the determination of the momentum vector of the slow proton from H_2^+ dissociation. Interference effects manifested themselves as an orientation dependent variation of both the transfer excitation rate [9] and, notably, of the distribution of the H^0 hits on the detector [8]. Qualitatively, these observations could be explained by considering the very small shifts in the projectile de Broglie wavelength related to one-electron transfers occurring efficiently only for trajectories close to either one of the H_2 target nuclei [10]. In this approach, however, it had to be assumed that the dissociation part in the transfer excitation process could be treated as being

separate from the electron transfer part as the variation of the cross section (with molecular angle) was found to be similar to those calculated earlier for single-electron capture (using fully quantum mechanical [10–12] and semiclassical [13,14] methods). Clearly, however, some uncertainty still remained concerning the exact role of the second active electron. With the present experiment, such ambiguities are removed since *both* electrons in the $\text{He}^{2+} + \text{H}_2$ system are transferred to the projectile.

The presently investigated interference in double-electron capture from H_2 molecules exhibits maxima of the cross section for perpendicular molecular orientations with respect to He^{2+} beams at 1.2 and 2.0 MeV. This is very interesting as the naive picture of sequential electron transfer, where the projectile interacts independently close to both of the target nuclei, rather should give maxima for orientations parallel to the ion beams. Nevertheless, and in spite of the fact that we are dealing with double-electron capture, we do observe one of the strongest interference effects measured to date in any molecular two-slit experiment and a strong velocity dependence. These results compare favorably with parallel fully quantum mechanical calculations of the Oppenheimer-Brinkmann-Kramers (OBK) type [15] and with the simple projectile de Broglie wave description, but only if it is assumed that both electrons are transferred together in collisions where the projectile passes close to either of the target nuclei. One earlier measurement of *double*-electron capture in $\text{He}^{2+} + \text{H}_2$ collisions was carried out by Martínez *et al.* [16] at much lower energies (100 and 400 keV) and only for two fixed molecular orientations of 10° and 90° with respect to the ion beam. At even lower energies, interferences for simultaneous two-electron transfer have been observed but then only for a one-center (atomic) target [17–19].

Molecular double-slit experiments have also shown interference effects in *electron emission* in energetic colli-

sions with ions [20–22], electrons [23], and photons [24–26]. In [26], interferences in the sum momentum of both electrons in photo-double ionization of H_2 was observed and interpreted as being related to the small fraction of the H_2 ground state for which both electrons are centered on the same nucleus. Very recently, Schmidt *et al.* [27] observed nicely resolved interference patterns in the transverse momentum transfer to He^+ in final-state resolved measurements on dissociative *single*-electron capture in 10 keV $H_2^+ + He$ collision and, in addition, they observed changes in these patterns with changing internuclear separation in H_2^+ [27].

The present experiment was performed in the ion storage ring CRYRING at Stockholm University (see Ref. [8] and references therein). The He^{2+} ions were injected, accelerated, stored and electron cooled [28] at energies of 1.2 and 2.0 MeV. These He^{2+} beams were crossed with a supersonically cooled H_2 jet [29]. Resulting molecular target ions H_2^{2+} , produced by double-electron capture to He^{2+} , Coulomb explode as two back-to-back protons with equal energies of about 9 eV. These 9 eV protons were then extracted perpendicular to the projectile beam and the gas jet in a Recoil-Ion-Momentum Spectrometer [30–32] and one of them was detected by a 2D position sensitive, resistive anode detector with 60% of its active area covered by a foil (c.f. Fig. 1). The fast, neutralized, He^0 , atom was detected in coincidence by a similar detector (projectile detector) 3.2 meters downstream of the collision region.

The projectile detector signal was used to start a multihit time-to-digital converter (TDC) while the stop signal was generated from a proton hit on the recoil-ion detector. For

each event the position and the time-of-flight for the proton was recorded. All parameters (times and positions) were stored in list mode. The full three-dimensional momentum vectors (mv_x, mv_y, mv_z) of the single 9 eV protons were then deduced using the position on the recoil detector with respect to the spectrometer axis and the time-of-flight [33]. In our energy range 300–500 keV/u, the collision times are much shorter than the H_2 vibrational and rotational times and, thus, the molecule may be viewed as *frozen* during the collision. Furthermore, the axial recoil approximation [34,35] is valid, i.e., the short time scale for dissociation (few fs) compared to that for rotations (few ps) strongly indicates that the momentum vector of the single 9 eV proton gives the direction of the molecular axis in the collision.

For our projectile velocities of $v_p = 3.46$ and 4.46 a.u., single ionization (SI), double ionization (DI), single capture (SC), transfer excitation (TE) and transfer ionization (TI) event rates are together way larger than the double-capture (DC) rate. Therefore, they may in principle give high random stop signal rates and make it difficult to record the true DC coincidences. However, the strongly dominating SI events give very small momentum transfers and thus the corresponding recoil ions would be concentrated in a small area close to the center of the detector and are stopped by the foil. (A similar technique has been successfully used before [36].) This tremendously improves the signal-to-noise ratio of the coincidence spectrum. As we only detect *one* 9 eV proton from each H_2^{2+} Coulomb explosion, we only record half of the angular distribution. However, except for angles close to $\theta = 90^\circ$ no information is lost due to the symmetry of the process (θ is the angle between the molecular axis and the ion beam). In Fig. 1, we show a schematic of the experimental arrangement, and the maximum, foil-limited, acceptance angle of $\theta = 75^\circ$ is indicated.

In Fig. 2, we show $d\sigma/d(\cos\theta)$, as functions of $\cos\theta$, where we have integrated over the full kinetic energy release distributions of the H_2^{2+} dissociation. The shaded region, for which $75^\circ \leq \theta \leq 105^\circ$ and thus $-0.22 \leq \cos\theta \leq 0.22$ (c.f. Fig. 1), is inaccessible due to the foil as seen from the very low measured intensities in this region. The data points for $0.22 \leq \cos\theta \leq 1.0$ were mirrored from the measured values for $-0.22 \geq \cos\theta \geq -1.0$. The experimental cross section for 300 keV/u (filled triangles in Fig. 2) increases monotonically from $\theta = 0^\circ$ to 90° and from 180° to 90° . This behavior changes strongly at 500 keV/u where the cross section goes through a strong oscillation (open squares) which becomes even more pronounced (not shown) when selecting the lower part of the kinetic energy release distribution for the H_2^{2+} fragmentation. The solid curves in Fig. 2 are our theoretical, fully quantum mechanical, OBK type calculations for double-electron capture from H_2 by He^{2+} ions. They are based on a simple model incorporating a linear combination of products of hydrogenic 1s wave functions to describe the

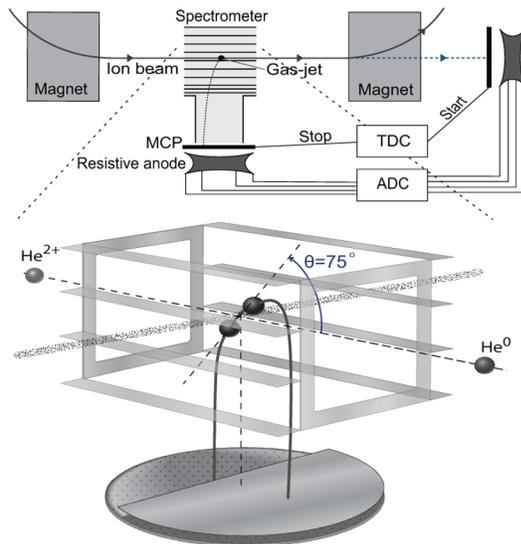


FIG. 1 (color online). Schematics of the experimental setup. The extraction, acceleration and drift regions are not shown to scale. The position sensitive detector is partly covered ($\sim 60\%$) by a foil. With this arrangement, random coincidences due to single ionization are avoided. Coulomb explosions for molecular axis orientation, θ , in the range $75^\circ \leq \theta \leq 105^\circ$ cannot be detected (c.f. text).

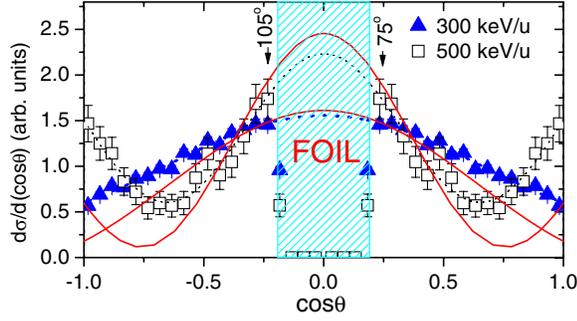


FIG. 2 (color online). Molecular axis orientation-angle dependent differential cross sections for 300 and 500 keV/u $\text{He}^{2+} + \text{H}_2$ double-electron capture reactions at 300 (filled triangles) and 500 keV/u (open squares). The dotted curves are fits [Eq. (1)] to the experimental data. The solid curves are the present OBK results, which should be multiplied by $8.4 \times 10^{-21} \text{ cm}^2$ and $4.8 \times 10^{-23} \text{ cm}^2$ for absolute scales at 300 and 500 keV/u, respectively.

two-electron two-center state of the H_2 molecule. In this OBK approach, the transfer of the two electrons can be understood as follows: one electron is captured as a result of the direct interaction with the projectile close to one of the nuclei, while the transfer of the other electron becomes possible through the nonzero overlap between the target and projectile states in a “shakeover” process—and thus this other electron does not need to be from a region close to a nucleus. Note that neither (the small) two-electron, one-center, component (non-Heitler-London part) of the H_2 ground state wave function, nor the sequential two-electron transfer is included in this OBK calculation. Still, the main features of the experimental results are reproduced (c.f. Fig. 2).

In a second, more qualitative approach, the experimental results are fitted with the function

$$d\sigma/d(\cos\theta) = A[1 + \mathcal{V} \cos(c\delta\phi)], \quad (1)$$

where A , \mathcal{V} , and c are free parameters. Here, $\delta\phi$ is the expected phase shift between two projectile de Broglie waves emerging from the molecular centers [8,11] and

$$\delta\phi = \vec{\delta k} \cdot \vec{a} \quad (2)$$

when we assume that *both* electrons are captured when the projectile is in the vicinity of either H_2 -target nucleus (internuclear vector \vec{a}). For small projectile deflection angles the momentum change is

$$\delta k \approx \delta k_{\parallel} = n \frac{v_p}{2} + Q/v_p, \quad (3)$$

where Q is the inelasticity. For $v_p \sim 4$ a.u. and $Q \sim 1$ a.u., $\delta k_{\parallel} \approx nv_p/2 = v_p$ for a two-electron transfer. Using this and Eq. (2) in Eq. (1) we reproduce the present experimental results (c.f. dotted curves in Fig. 2).

Based on the similarity of the presently observed oscillatory behavior for double-electron capture, with theoretical single-electron capture [10–12], and experimental

transfer excitation cross sections [9], it appears that the mechanisms behind these three processes have important features in common. In Fig. 3 we thus compare our DC results with those of Støchkel *et al.* [9] for the $\text{H}^+ + \text{H}_2$ TE process. In Figs. 3(a)–3(d), we show the TE results [9] for 1300 keV/u, 1000 keV/u, 700 keV/u, and 300 keV/u corresponding to perturbation strengths $S_p = q/v_p = 0.14$ –0.29 (q is the projectile charge state). Figures 3(e) and 3(f) show the present $\text{He}^{2+} + \text{H}_2$ DC results for 500 keV/u and 300 keV/u ($S_p = 0.45$ and 0.58). The curves are fits [Eq. (1)] and the vertical dotted lines indicate $\delta\phi = \pm\pi$ where the first minimum is expected for destructive interference in the exact forward scattering direction of the projectile [12] and $c = 1$ in Eq. (1). The positions of the measured minima do, however, vary systematically from the top to the bottom panel. There is a very slight deviation from $\pm\pi$, for $S_p = 0.14$ while a much larger deviation is found for $S_p = 0.58$. This effect can be studied in a somewhat more quantitative manner as a function of c [c.f. Eqs. (1) and (2)] or the “effective slit distance”, $d = ca$, for a one- and two-electron capture from H_2 . The latter is related to two separate spatial regions in H_2 within which projectile trajectories should pass for electron transfer to become more favored than otherwise. In Fig. 4, d is very close to $a = 1.41a_0$ (a_0 is the Bohr radius) for our smallest value of S_p , while it decreases

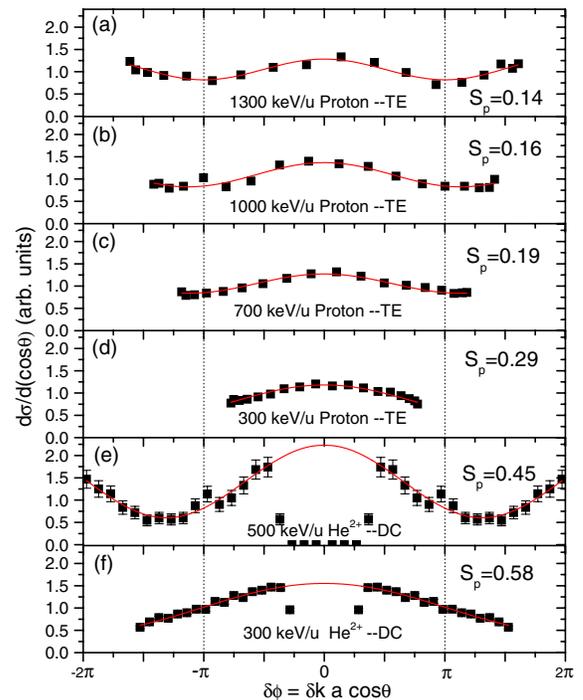


FIG. 3 (color online). Panels (a)–(d) experimental results for 1300 keV/u – 300 keV/u protons on H_2 —the TE process, from Ref. [9]. Panels (e),(f) present results for 500 and 300 keV/u He^{2+} on H_2 —the DC process. Curves are fitted functions [Eq. (1)] to the experimental data (filled squares). Vertical dotted lines at $\pm\pi$ show the expected positions for the first minima according to Eqs. (1)–(3) with $c = 1$.

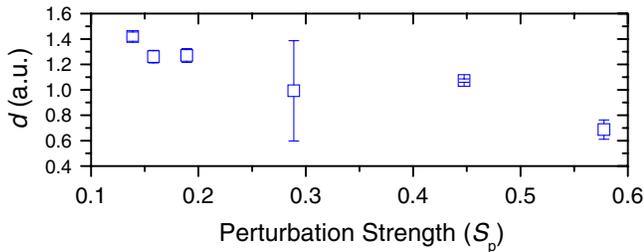


FIG. 4 (color online). Effective distance, d , between two favored regions for electron transfer from the H_2 molecule as seen by the incoming H^+ and He^{2+} ions as functions of the perturbation strength $S_p = q/v_p$ (c.f. text).

rather strongly with increasing S_p . The latter concerns two-electron capture and may be qualitatively understood in the following way: The probability density of the target electron cloud is higher at a given distance from one nucleus and between the two nuclei than at the same distance from the same nucleus in the other direction. This presumably also means that the two-electron transfer probability is asymmetric with respect to either target nucleus and that it is higher between them. In principle, this implies an effective slit (or scattering center) separation smaller than a . The larger the two localized regions within which such simultaneous two-electron transfer processes are effective (i.e., the larger S_p is), the larger the deviation from a slit distance equal to the internuclear separation for H_2 becomes. To a large extent, the variation with S_p displayed in Fig. 4 may thus be attributed to larger capture radii (with respect to either nucleus) and lower impact velocities for the present $He^{2+} + H_2$ study of two-electron capture in relation to the $H^+ + H_2$ TE case [8,9] (where one electron is transferred).

In this Letter, we report on double-electron capture from H_2 molecules. We observe a strong oscillatory behavior of the cross section as a function of the molecular orientation which, furthermore, significantly varies when the projectile velocity is reduced by only 20%. The observed features are attributed to Young-type interference effects arising from the two-center nature of H_2 which is, for the first time, observed for *double*-electron capture from molecular targets. Our theoretical model calculation, based on the OBK approximation and incorporating a two-electron, two-center wave function for the H_2 ground state, qualitatively reproduces the experimental results when neither the non-Heitler-London part of the H_2 wave function nor sequential electron transfer processes are included. This thus defies the naive intuitive picture of two sequential electron transfers and, instead, it indicates that the two electrons are captured in a correlated fashion for projectile trajectories close to either target nucleus.

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