Evidence of Wave-Particle Duality for Single Fast Hydrogen Atoms

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We report the direct observation of interference effects in a Young's double-slit experiment where the interfering waves are two spatially separated components of the de Broglie wave of single 1.3 MeV hydrogen atoms formed close to either target nucleus in $H^+ + H_2$ electron-transfer collisions. Quantum interference strongly influences the results even though the hydrogen atoms have a de Broglie wavelength, λ_{dB}, as small as 25 fm.

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The wave-particle duality is a common property of light and matter and a key concept of quantum mechanics, which almost without exception is illustrated with the two-slit experiment in introductory physics textbooks. Already 200 years ago Young [1] demonstrated the wave nature of light by observing interference patterns behind pinholes. More than 100 years later, in 1923, de Broglie [2] proposed that the propagations of electrons and other material particles also could be described as waves and thus that interference patterns should appear if ''particles'' were allowed to interact with two slits or some other forms of spatially well-defined scattering centers. In 1927 Davisson and Germer provided the first experimental evidence of the electron wave nature [3] by electron scattering on a nickel crystal. The scattering off periodic structures (crystals, gratings, etc.) has since been applied to demonstrate the wave nature of the propagation of still heavier assemblies of matter such as fullerenes [4] and even larger molecules [5]. In surface science, the interference of scattered electrons and neutrons are since long standard analytical techniques and it was recently demonstrated that also grazing incidence neutral atoms may be used for surface analysis [6].

Although there is overwhelming evidence for the wave description of matter, the conceptually important two-slit experiment was for quite some time merely regarded as an illustrative gedanken experiment [7]. Already in 1961, however, Jönsson observed electron-wave interference in an actual double-slit experiment (using an etched mechanical double slit) with electrons [8], and in 1976 Merli *et al.* [9] demonstrated the wave behavior of *single* electrons in the low-intensity limit. Double-slit experiments have further been performed with neutrons [10] and metastable helium atoms [11]. In ion-atom collisions, two-*paths* effects (in which different ''particle trajectories'' may lead to the same final state without spatially localized and separate scattering centers) may give rise to Stückelberg oscillations as demonstrated in, e.g., Refs. [12–15]. Recently two-center interference was also observed in *electron* emissions in ion-H_2 collision experiments [16–18].

Here, we report direct experimental evidence of two-center quantum interference in a parameter region where the classical limit is expected to be strongly approached, using fast *single* hydrogen atoms with a de Broglie wavelength of $\lambda_{dB} = 25$ fm. The hydrogen atoms are formed in electron-transfer reactions in the vicinities of the nuclei in H_2. Wang *et al.* [19] calculated that single-electron capture probabilities are significant only for impact parameters up to about $0.3a_0$ with respect to either proton in H_2 with its $1.4a_0$ internuclear distance (a_0 is the Bohr radius). Thus, the relative phases of the two contributions to the outgoing de Broglie wave should be decisive for the angular scattering distribution and intensity. Here, we determine the orientation of the target molecule at the time of the collision and the correlated projectile (H^0) scattering angle for 1.3 MeV

$$H^+ + H_2 \rightarrow H^0 + (H_2^+)^* \rightarrow H^0 + (H + H^+) \quad (1)$$

transfer excitation collisions. The fast H^0 atoms are detected on a position-sensitive detector 3.2 m ($= 1.3 \times 10^{14} \times \lambda_{dB}$) downstream of the H_2 target, where their single-particle probability distributions are given by the wave description of their propagations to the detector. In fast collisions, like (1) at 1.3 MeV, typical interaction times are in the attosecond regime, and the momentum transfer to the H_2 target becomes extremely small (meV recoil energies). Thus, the momentum (and kinetic energy) of the outgoing H^0 atom is very nearly the same as for the incoming 1.3 MeV proton and the projectile scattering angle is small. Dissociation of $(H_2^+)^*$ typically takes a few femtoseconds and gives eV fragment energies.

Tuan and Gerjuoy [20] calculated an increase due to two-center interference of the total $H^+ + H_2 \rightarrow H^0 + H_2{}^+$ pure single-electron capture cross section (i.e., without $H_2{}^+$ dissociation) in relation to that for two separate hydrogen target atoms. Following Deb *et al.* [21], Wang *et al.* [19], and Shingal and Lin [22] derived equivalent theoretical cross section variations with the H_2 target orientation. Their predictions were recently confirmed for 0.3–1.3 MeV $H^+ + H_2$ collisions [23], while no direct observation of interference in the outgoing projectile wave was possible in [23] or in the only similar work [24].

A proton beam from a plasma ion source was accelerated to 300 keV, injected in the ion-storage ring CRYRING [25] where it was further accelerated to 1.3 MeV and electron cooled [26] to a diameter well below 0.8 mm (at 500 nA). This circulating proton beam intersects a cold narrow supersonic H_2 jet [27,28] at right angles and inside the extraction region of a recoil ion momentum spectrometer [29] with its axis perpendicular to the fast proton beam and the H_2 jet. We measure the momentum vectors for slow H^+ ions (eV energy range) from $(H_2{}^+)^*$ dissociations by means of their time of flights to, and positions on, the recoil detector. Our standard spectrometer configuration is described in Refs. [23,30,31], but here we suppress a strong random background from single ionization with a 10 mm diameter foil mounted on a grid in front of the detector and on the spectrometer axis. In this way, we block ions with low momenta perpendicular to this axis and strongly suppress the low-energy $H_2{}^+$ ions from single ionization. The foil also blocks H^+ ions from dissociations *along* the spectrometer axis, but this can be compensated for by means of the azimuthal symmetry with respect to the projectile beam. The transfer excitation rate never exceeded 0.05 s^{-1}. With a projectile flight time of 200 ns, the probability for two fast H^0 atoms simultaneously in flight between the target and the detector is $<10^{-8}$. This ensures that we indeed probe the interference of individual single H^0 atoms with themselves.

The proton momentum vector gives the orientation (θ, φ) of the H_2 molecule in the collision as dissociation processes are very fast compared to molecular rotations. For each event (1) this information, in the form of proton time of flight and position, is recorded in coincidence with the (x, y) position of the fast H^0 atom on the projectile detector. Using (x', y') coordinate systems, defined by individual azimuthal orientations φ as shown in Fig. 1, we expect interference patterns in the probability distributions for H^0 hits along the x' axis. The present primary results are variations in the x' distributions as functions of θ (cf. Figs. 2 and 3).

Although we have performed measurements on transfer excitation processes (1) we note that similar periodic variations were calculated in Refs. [19,21] for single-electron capture (which is identical to (1) except that $H_2{}^+$ does not fragment). Further, although the present experiment resembles the optical double-slit experiment, there is one crucial difference in that electron transfer leads to a change in λ_{dB} and thus a θ-dependent phase difference which may give destructive interference also for zero degree projectile scattering. This does not occur in the optical case where a change in θ would change the effective slit separation but preserve the constructive interference at zero degrees. To illustrate this unique feature of the electron-transfer two-slit situation we will discuss the $\theta = 90°$ and $\theta = 51°$ cases, which are expected to give constructive and destructive interference at zero scattering angle, respectively.

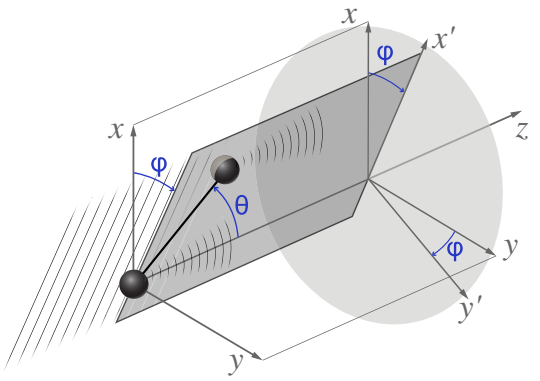

FIG. 1 (color online). The collision geometry: The interference for this single scattering event is expected along the x' axis which is defined by the azimuthal orientation φ of the target molecule, which lies in the $x'z$ plane. The incoming and outgoing wave fronts are shown schematically in this plane for a projectile propagating in the z direction.

For $\theta = 90°$ we expect a central maximum (with capture from the dominant gerade-gerade component of the H_2 ground state [23,32]) and side minima and maxima for

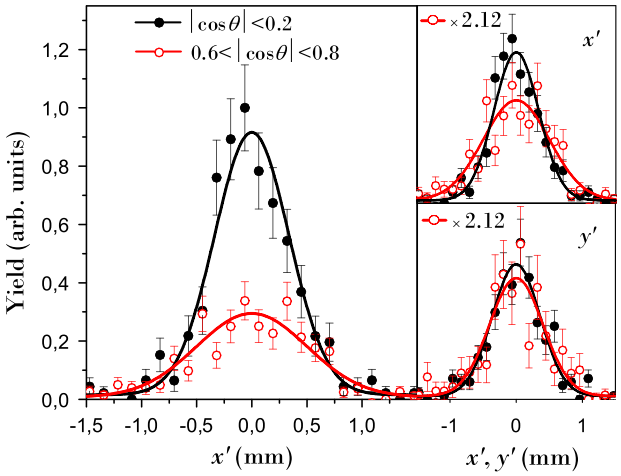

FIG. 2 (color online). The x' distribution of fast H^0 atoms. Solid and open circles: $|\cos\theta| < 0.2$ and $0.6 < |\cos\theta| < 0.8$, respectively. Upper right panel: As main panel but with open circle data ×2.12. Lower right panel: The y' distributions normalized as above. The curves are Gaussian fits (cf. text).

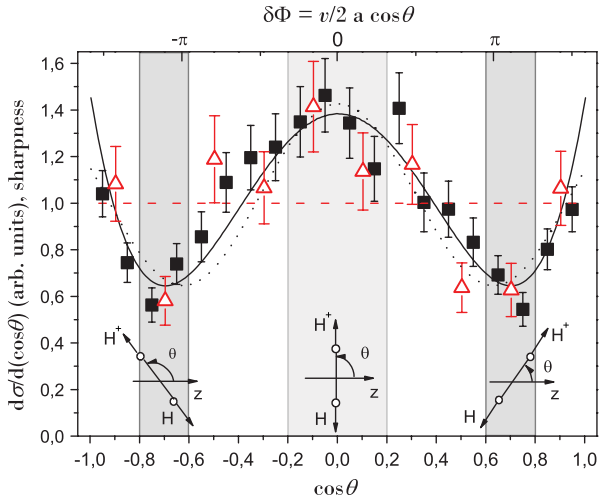

FIG. 3 (color online). Relative transfer excitation cross sections $d\sigma/d(\cos\theta)$ (solid squares), and peak sharpness (open triangles cf. text) as functions of molecular orientation. The upper horizontal axis gives the phase shift $\delta\Phi$ according to Eq. (3) for zero degree projectile scattering. The full curve is the theoretical result of Ref. [19] for single-electron capture and the dotted curve is $1 + \xi\cos(\delta\Phi)$ with $\xi = 0.34$ (cf. text). The shaded areas indicate $\cos\theta$ intervals for the data in Fig. 2.

destructive and constructive interference along x' as the path length difference of the two wave components from either center reaches half-integer and integer values of $\lambda_{dB} = 25$ fm. Deb *et al.* [21] calculated such interference patterns, but found less than 1% of the total intensity in the first secondary maxima (in our case located at $|x'| = 1.1$ mm from the central peak). At the present statistical level of data, we thus expect a single peak in the x' distribution for $\theta = 90°$. The shape and magnitude of this peak is, however, affected by quantum mechanical interference. At zero scattering angle ($x' = 0$) the two contributions to the de Broglie wave are in phase and interfere to increase the peak intensity. Away from exact forward scattering the interference is less favorable and thus the signal falls of more rapidly than in the absence of interference effects. Thus, for $\theta = 90°$, quantum interference is expected to increase the integral value and to make the width of the x' distribution more narrow.

For $\theta \neq 90°$ there will be a phase difference between the two parts of the neutralized projectile wave due to electron transfer from the vicinity of either H_2 nucleus. At the velocity v, the projectile gains forward momentum (in atomic units):

$$\delta k = v/2 + Q/v, \tag{2}$$

where Q is the inelasticity in (1). For 1.3 MeV protons, the first term dominates and thus the change in projectile wave number due to electron transfer is $\delta k \approx v/2 = 3.6$ a.u. (a very small change in relation to $k = 1.3 \times 10^4$ a.u. for 1.3 MeV protons). This yields the phase difference

$$\delta\Phi = \delta k a\cos\theta = 5.0\cos\theta, \tag{3}$$

where $a\cos\theta$ is the distance of propagation between the target nuclei along the beam direction and $a = 1.4a_0$ is the H_2 internuclear distance. For $\theta = 51°$, $\delta\Phi = \pi$ and we expect *destructive* interference in the exact forward direction. More generally, we expect the cross section to vary with the molecular orientation and, thus, $\delta\Phi$ as

$$d\sigma/d(\cos\theta) \propto 1 + \xi\cos(\delta\Phi), \tag{4}$$

where $\xi = 1$ in the idealized case of interference of two plane waves, while it is smaller in the real situation.

In Fig. 2 we show the measured distributions of neutralized projectiles along the x' axis due to reactions (1). The solid and open circles are for H_2 orientations with $|\cos\theta| < 0.2$ and $0.6 < |\cos\theta| < 0.8$ (expected *constructive* and *destructive* interference at zero degree H^0 scattering, respectively). With two such equally wide $|\cos\theta|$ ranges (shaded areas in Fig. 3) we would, in the absence of quantum interference, expect the two data sets in Fig. 2 to be equal in shape and magnitude.

The measured H^0 distributions in Fig. 2 are, however, different. First, the *integral* intensity is much larger when the molecular axis is perpendicular to the beam direction (left main panel of Fig. 2). Second, the *shapes* of the distributions are different as Gaussian fits yield widths (FWHM; 95% confidence level) of 0.80 ± 0.06 mm for $|\cos\theta| < 0.2$ and 1.18 ± 0.12 mm for $0.6 < |\cos\theta| < 0.8$ (cf. Fig. 2 upper right panel where the latter data are multiplied by 2.12 in order to better compare peak *shapes*). The absence of a minimum at $x' = 0$ in the latter case, where destructive interference is expected at zero degree projectile scattering, is mainly due to the finite width of the fast proton beam. For projections on the orthogonal y' axis the widths are the same within error bars (Fig. 2 lower right panel). This is exactly what is expected—the H^0 scattering distribution is influenced by two-center effects only in the direction given by the azimuthal orientation φ of the molecule.

The data in Figs. 2 and 3 are for kinetic energy releases in the range 5–8 eV for H_2^+ dissociations. The lower limit at 5 eV excludes contributions from H_2^+ ions formed in the vibrational continuum of the H_2^+ electronic ground state $(1s\sigma_g)^2\Sigma_g^+$, while the upper, 8 eV, limit excludes effects of aberrations for trajectories too far off the spectrometer axis. The upper limit favors transfer excitation via the $(2p\pi_u)^2\Pi_u$ and $(2s\sigma_g)^2\Sigma_g$ states of H_2^+ over those from $(2p\sigma_u)^2\Sigma_u^+$. The population of different final target (and projectile) states implies that not all transfer excitation events have exactly the same Q value. However, this only weakly affects δk [cf. Eq. (2)] and thus the single-event x' probability distributions are similar and the interference effect appears also in the x' histograms of many events.

The present transfer excitation cross sections (integrated over x') and peak sharpness of the x' distributions, defined as the ratio of the intensities for H^0 scattering angles

smaller and larger than 0.1 mrad ($|x'|$ smaller and larger than 0.32 mm), are shown as functions of $\cos\theta$ in Fig. 3. The locations of the minima, close to but not exactly at $\cos(51°)$ and $\cos(129°)$, and the amplitudes of the oscillations of the transfer excitation cross section are in agreement with the quantum mechanical calculations for single-electron capture by Wang *et al.* [19]. Thus, the probability for excitation of the remaining H_2^+ ion does not appear to depend strongly on θ. Using a self-consistent field method with a second order perturbation treatment of electron correlation [33] we calculate the overlap between the H_2 ground state and the H_2^+ wave functions at $a = 1.4a_0$. This gives a shakeup probability as large as $(9 \pm 1)\%$ indicating that electron transfer with shakeup may well be the dominant transfer excitation mechanism and, thus, that the relevant theoretical description for the θ dependence of this process is identical to that for single-electron capture.

The result of the qualitative description given above in the form of Eq. (4) (with ξ arbitrarily set to $\xi = 0.34$) is shown in Fig. 3 (dotted curve). Though far less sophisticated than the full quantum treatment by Wang *et al.* [19] our simple picture with two interfering outgoing projectile wave components captures the essence of the orientation dependence of the transfer excitation cross section. The $\cos\theta$ dependence for the H^0-peak sharpness (open triangles in Fig. 3) is similar to that for the integrated cross section and thus a narrow projectile angular distribution is correlated with a large cross section. This is consistent with the discussion above, which suggests that interference at small projectile scattering angles controls the magnitude of the total cross section. Steric (geometrical shadowing) effects, on the other hand, appear to be relatively weak as they would favor minima at $\theta = 0°$ and 180°.

In this Letter, we have reported the direct observation of two-center interference for *single* protons capturing electrons from H_2 to form hydrogen atoms with 25 fm de Broglie wavelengths. Single-particle detections add up to position distributions which clearly depend on the orientation of the target molecule at the time of the collision and which only can be accounted for if the wave properties of the propagating fast H^0 atoms are taken into account. This unambiguously demonstrates the wave-particle duality for single hydrogen atoms with extremely small de Broglie wavelengths in relation to the atomic length scale and the distance between the scattering centers. The outgoing hydrogen atom wave is a superposition of contributions from the vicinities of either proton in H_2. While there are large conceptual similarities with Young's original two-slit experiment the electron-transfer process has a further intriguing feature as the interference is governed by a *change* in de Broglie wavelength at either scattering center (''slit''). Still, the two-slit dilemma remains: There is no answer to the question from which atomic center the electron was captured.

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