Momentum Transfer to a Free Floating Double Slit: Realization of a Thought Experiment from the Einstein-Bohr Debates

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We simultaneously measured the momentum transferred to a free-floating molecular double slit and the momentum change of the atom scattering from it. Our experimental results are compared to quantum mechanical and semiclassical models. The results reveal that a classical description of the slits, which was used by Einstein in his debate with Bohr, provides a surprisingly good description of the experimental results, even for a microscopic system, if momentum transfer is not ascribed to a specific pathway but shared coherently and simultaneously between both.

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Quantum mechanics poses a major challenge to our intuition which is trained in the macrocosm to the laws of classical physics. Among all the quantum phenomena, the double-slit interference is "a phenomenon which is impossible (...) to explain in any classical way, and which has in it the heart of quantum mechanics" (Feynman). Consequently, from its early days on the double slit has been used as an example to discuss the wave concept of quantum mechanics. Most famously, Einstein challenged quantum mechanics by a thought experiment he proposed to Bohr [1,2]. He argued that it should be possible to determine the pathway of each individual particle passing through a double slit by observing the recoil momentum it imparts onto a first slit used to diffract the particle wave, ensuring it coherently illuminates the double-slit assembly. Einstein construed this to express his "deep concern over the extent to which causal account in space and time was abandoned in quantum mechanics" (quoted from [1]). Bohr countered by asserting that the slits, in addition to the scattered particle, obey the laws of quantum mechanics. A slightly modified version of Einstein's thought experiment, which is better matched to Bohr's arguments, is illustrated in Fig. 1(a). Here one expects a momentum transfer p_A or p_B to the double slit in the case of pathway A or B. However, the uncertainty principle either prohibits the determination of the pathway or implies an uncertainty of the length of the two pathways so that the interference structure disappears, depending on whether the slit momentum or position is fixed initially [3]. Since we report an experiment which shows high interference contrast we have realized the case where (almost) no which way information can be obtained. In modern terms, Bohr argued that the quantum-classical border cannot be drawn between the interfering particle (quantum) and the slit arrangement (classically) [4] as implicitly done by Einstein. Instead, the double slit is part of the quantum mechanical system and has to be treated accordingly.

In the spirit of the original thought experiment we measure both slit and projectile momenta. The observed projectile interference pattern is compared to semiclassical and quantum mechanical calculations. These show that even for *microscopic* "slits" a classical modeling of the slit dynamics can be appropriate if the momentum transfer from the scattered particle to the slits is treated in a way not consistent with classical mechanics.

To relate our experiment to the original Einstein-Bohr considerations, we need to consider the two-dimensional analog of the original thought experiment, as shown in Fig. 1(b). In contrast to the one-dimensional case, recoil from the double pinhole may induce rotation (clockwise or anticlockwise) in the x-y plane in addition to translation. However, for a macroscopic pinhole arrangement the interference pattern still consists of vertical stripes. In the present experiment we replace the macroscopic double pinhole by an ensemble of molecular "microslits" (free-floating HD molecules) and observe the diffraction of helium atoms by them [5]. Because the mass of the microslits is now comparable to that of the individual projectile atoms and the mass difference between the H and D nuclei is considerable, a significant distortion in the interference fringe pattern by rotational excitation of the slit is expected. Isotope labeling of the slits enables the entanglement of the slit-projectile system to be explored.

One might anticipate that the molecules comprising our molecular slits need to be aligned in space to observe interference phenomena. This is not the case. It is sufficient to choose only those scattering events in which the molecules rapidly dissociate and to postselect molecular orientations by measuring the emission angles of their atomic fragments. By the commonly used axial-recoil approximation [6,7] the internuclear vector at the time of collision is inferred from the measured directions of the fragments. Furthermore, given the correlation between

kinetic energy release (KER) of the molecular breakup and the internuclear distance at the instance of its inception [8,9], interference for well-defined slit separations may be investigated.

Figure 1(c) shows a schematic representation of our experiment. Helium atoms are used as the projectiles and the double pinhole is replaced by the two nuclei of the diatomic molecular ion HD^+. Scattering with high momentum transfer is localized close to the nuclei. Only those scattering events corresponding to dissociative electron attachment

$$\begin{aligned} \mathrm{He}(1s^2) + \mathrm{HD}^+(1s\sigma_g) &\rightarrow \mathrm{He}^+(1s) + \mathrm{HD}(b^3\Sigma_u^+) \\ &\rightarrow \mathrm{He}^+(1s) + \mathrm{H}(1s) + \mathrm{D}(1s) \end{aligned}$$

are analyzed, as these lead to an accurate determination of slit orientation. One electron is transferred from the He to the empty $2p\sigma_u$ orbital of the HD^+, forming electronically exited neutral HD on the repulsive $b^3\Sigma_u^+$ potential energy curve [10]. The transition of the electron from an even to an odd state effects a phase shift of π between the two scattering pathways, which inverts the interference maxima to minima and vice versa [5,6,11].

The experiment is performed in inverse kinematics; i.e., a fast beam of molecules, which constitute the "slits", collides with a helium gas target. HD^+ molecules, produced in a Penning source and in a mixture of several vibrational and rotational excited states [10] are accelerated to 30 keV before colliding with helium atoms prepared within a supersonic jet. H and D fragments, formed through the process of charge transfer are measured on position- and time-sensitive delay-line detectors [12] located behind the reaction region, enabling their momenta to be reconstructed and the slit geometry (HD internuclear distance and molecular orientation) at the time of collision to be inferred. The momenta of the He^+ ions, which show interference stripes in the plane perpendicular to the direction of impact, were measured by cold target recoil ion momentum spectroscopy (COLTRIMS) [13,14].

The interference pattern of the scattered helium is shown in Fig. 2. This representation in momentum space is

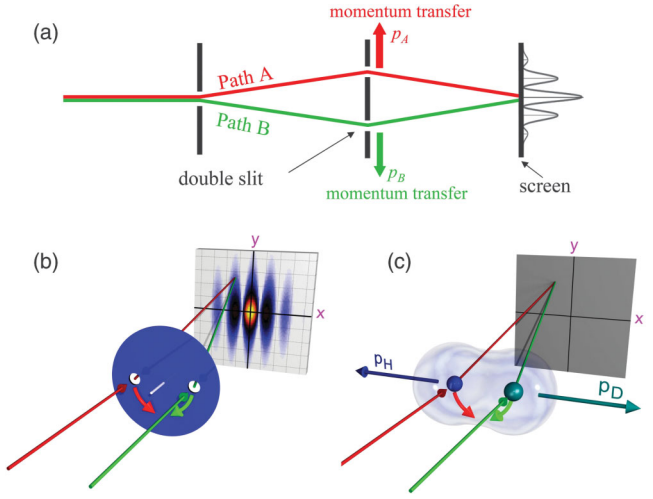

FIG. 1 (color). Thought experiment for a kicked double slit. (a) A coherent particle wave travels through the double slit to the screen where its probability distribution shows interference if no which-way information can be gained. Along path A (B) the particle is deflected downward (upward), therefore measurement of the double-slit momentum for each particle should allow determination of their paths. Equivalently, the momentum transfer to the first slit could be measured as was originally proposed by Einstein [1]. (b) Two-dimensional version of the arrangement in (a). Momentum transfer from the projectile can cause a clockwise- or anticlockwise-rotation of the pinholes. (c) Experimental implementation of (b). Atoms collide with a HD molecule. Rutherford scattering at one of the molecular nuclei establishes the two interfering pathways. The scattered atom is observed in coincidence with the molecular fragments. In contrast to the macroscopic slit arrangement in (b) with the rotation axis located exactly in between the apertures, the unequal nuclear masses of our molecular microslits lead to a curving of the interference fringes.

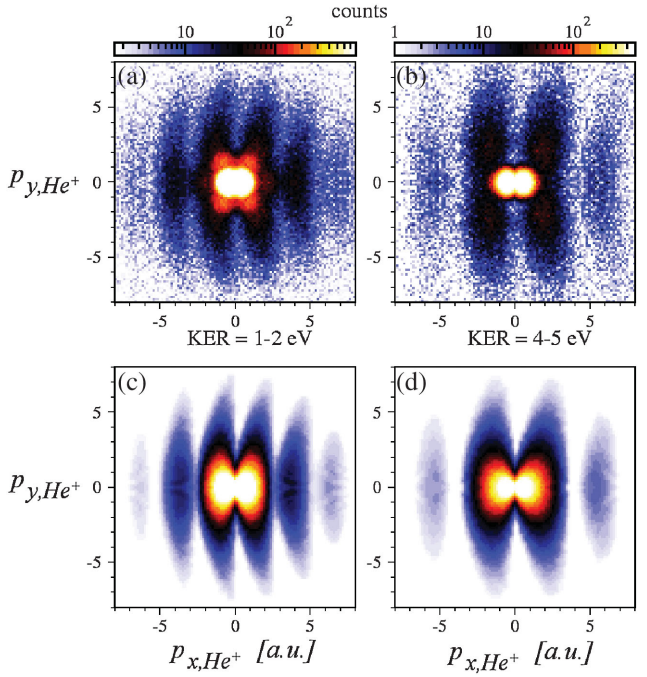

FIG. 2 (color). Two-dimensional distribution of the momentum transfer to He scattered at HD^+ and leading to dissociative electron transfer $\mathrm{He} + \mathrm{HD}^+(1s\sigma_g) \rightarrow \mathrm{He}^+ + \mathrm{HD}(1s\sigma_g, 2p\sigma_u) \rightarrow \mathrm{He}^+ + \mathrm{H}(1s) + \mathrm{D}(1s)$. The experimental distributions shown in (a) and (b) consist of events where the break-up direction of the molecule is measured between 85° to 95° with respect to the direction of impact. The internuclear vector at the classical dissociation limit and pointing from the H to the D defines the x axis. (c) and (d) Predictions from a quantum mechanical treatment of the slit dynamics. Two different regions of KER have been selected: (a) and (c): 1 eV < KER < 2 eV, (b) and (d): 4 eV < KER < 5 eV. Momenta are given in atomic units ($m_e = e = \hbar = 4\pi\varepsilon_0 = 1$).

equivalent to a spatial interference pattern because the helium is propagated to macroscopic distances during the process of measurement. The horizontal axis of the coordinate frame is defined by the fragmentation direction of the molecule with the H directed to the right. We select two different regions of KER, which correspond to distinct internuclear distances (slit separations). As stated above, the interference pattern is inverted compared to the optical case. More importantly, the vertical interference stripes are bent, which was not observed for H_2 molecules [5] where the additional final state symmetry arising for identical molecular fragments enforces the reflection symmetry of the diffraction pattern about the vertical axis and therefore prohibits the pattern bending to a specific side. As we will show, this bending is a result of the coherent rotation of the molecular axis initiated by the momentum kick [15].

A macroscopic double pinhole, as shown in Fig. 1(b), has a well-defined orientation and slit separation. If we consider the dynamics of our microscopic molecular double slit classically and neglect the momentum transfer by the diffracted He, the excited HD would dissociate along the bond axis and the fragment direction would coincide with the slit axis. In this case one would expect the maxima and minima in Figs. 2(a) and 2(b) to lie along vertical lines, in clear contradiction to the observed curved pattern which shows that the momentum transfer is relevant in our case. The observed final H or D direction does not coincide with the internuclear axis at the time when the scattering occurred.

From the quantum mechanical perspective, our molecular-slit ensemble of HD^+ molecules comprises an incoherent superposition of vibrational and rotational excited states. We restrict our modeling to molecules initially in the rotational ground state of HD^+ which describes a coherent superposition of all possible orientations in three dimensions; no classical analog exists for such a state. Nevertheless, prominent interference effects can still be observed if specific momenta (i.e., break-up directions and KER) of the dissociation fragments are selected. This is because the collision process causes rotational excitation which is essential to generate orientational effects.

To interpret our experimental results we compare them with calculations derived from semiclassical and quantum-mechanical theoretical approaches. In all approaches the diffracted wave is described by the superposition of contributions from the respective nuclei of the molecular double slit. The key difference is the way in which momentum is transferred from the projectile to the molecular slit.

Our first approach employs a classical treatment of fragment trajectories. Momentum transfer is assumed to occur at either the H or the D atom. Such a localization of momentum transfer to a particular point in space was envisaged in the Bohr-Einstein debate inspired thought experiment. Thus, while the wave scatters simultaneously from both atoms (delocalization of the projectile wave function), the momentum transfer occurs at only one, causing the intermolecular axis to rotate in a clockwise or in an anticlockwise direction. The degree and sign of rotation, which depends on the magnitude and direction of the relative momentum kick [16] and on whether the momentum transfer occurs at the H or D atom, is calculated within classical mechanics, by numerically solving Newton's equation for the relative motion of the molecular fragments. The orientation of the molecular slit at the time of scattering can then be inferred and interference patterns calculated.

Two-dimensional distributions for the momentum transferred between the helium and the H or D atom are shown in Fig. 3, panel (a) or (b), respectively. As scattering from both is expected to occur with equal probability, it is appropriate to compare the average of distributions (a) and (b) [presented in Fig. 3(c)] with the results of measurement [Fig. 2(a)]. As is clearly seen, the model achieves unsatisfactory agreement with the experimental results, exhibiting a more complex structure of lesser contrast than observed in measurement.

Our second approach pursues the strategy suggested in Bohr's reply to Einstein, namely, that the slits themselves be treated quantum mechanically. This results in a system

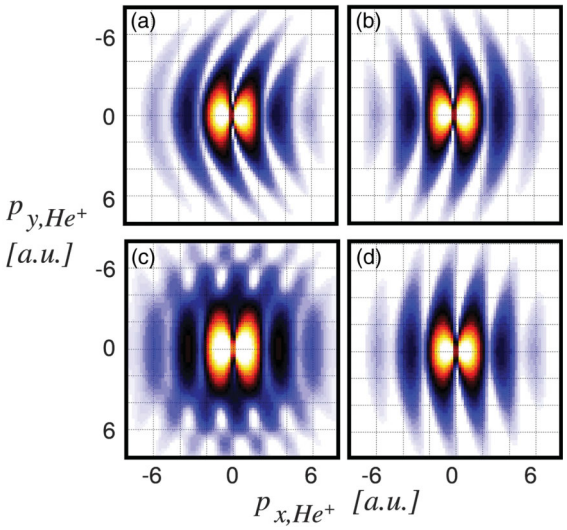

FIG. 3 (color). Two-dimensional distribution of the momentum transfer to He scattered at HD^+ and leading to dissociative electron transfer. Semiclassical calculations for HD internuclear distances resulting in 1 eV $<$ KER $<$ 2 eV. The x axis is defined by the internuclear vector at the classical dissociation limit pointing from the H to the D. The dissociation is started with a relative motion of the fragments resulting from momentum transfer at either (a) the H or (b) the D atom from the scattered helium. These two cases are classical analogs to the two translation factors used to modify the initial state wave function of the quantum mechanical model. (c) Average of (a) and (b). (d) The transferred momentum is shared equally between H and D atoms.

of three correlated particles, H, D, and the scattered helium, which can only be solved by applying approximations.

Since the total momentum of the closed system comprising these three particles is conserved, the motion of the center of mass of the double slits equals the momentum change of the scattered particle. Therefore, its quantum mechanical description will not provide new insights. In contrast, additional information is gained from the quantum mechanical description of the relative motion of the two scattering centers which involves a coherent superposition of double-slit orientations and slit distances and the effect of rotational and vibrational excitation during the collision process.

It can be assumed that close scattering at one of the molecular centers involves a localized momentum transfer at this nuclear site. This implies a definite correlation between the momentum transfer and the internuclear wave function describing the molecule. The outgoing wave of the scattered particle is a coherent superposition of contributions from the two scattering centers. Correspondingly, the internuclear wave function after scattering describes the molecule getting stretched or compressed and getting clockwise or anticlockwise rotationally excited, depending on the pathway of the scattered particle. Our quantum mechanical approach is based on the assumption that the description of the internal degrees of freedom of the molecule is sufficient to characterize the complete system. To further reduce the dimensionality of the problem, we treat only the plane perpendicular to the beam axis.

The collision process leads to an internuclear wave function Ψ' which is derived from the initial state wave function Ψ by multiplying it with a coherent superposition of two translation factors, namely,

$$\Psi'(\mathbf{R}) = \Psi(\mathbf{R}) \left[\exp\left(i\mathbf{k}\mathbf{R} \frac{m_D}{m_D + m_H} \right) - \exp\left(-i\mathbf{k}\mathbf{R} \frac{m_H}{m_D + m_H} \right) \right].$$

The evolution of this kicked HD nuclear wave function [15] is calculated on the same repulsive excited HD potential used in the semiclassical modeling. A multiconfigurational time-dependent Hartree approach (MCTDH) [17] is used to propagate the wave function until the dissociation limit is reached. The asymptotic wave function is then analyzed to extract the distribution of the KER and the angle φ between the final internuclear vector $\mathbf{R}$ and the momentum-transfer vector $\mathbf{k}$. Quantum mechanical and experimental results in terms of KER and φ are presented in Fig. 4.

We perform these calculations for a large number of absolute values of $\mathbf{k}$. Because the momentum transfer to the scattered particle is a measured value we can add up these independent KER-φ distributions to finally compose the diffraction pattern of the scattered particle shown in Figs. 2(c) and 2(d). In contrast to the semiclassical predictions, excellent agreement is now achieved with the experimental results in Figs. 2(a) and 2(b).

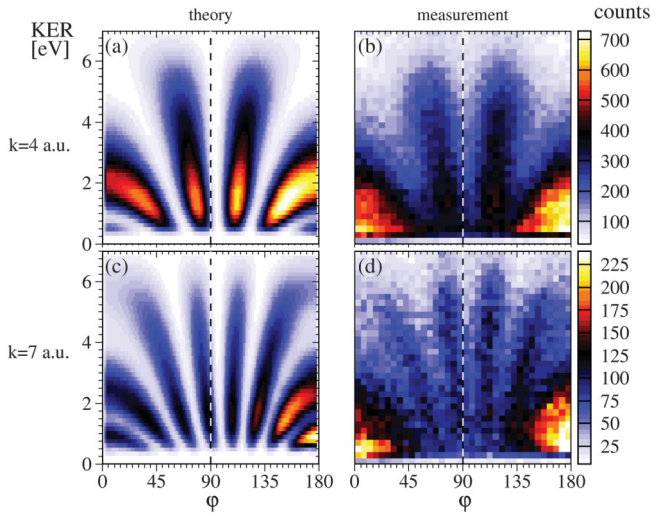

FIG. 4 (color). Final state of dissociated HD molecules for different magnitudes of momentum transfer $p = \hbar k$ to the molecule. The final state is parameterized by the KER and the angle φ between the vector $\mathbf{k} = \mathbf{p}_{He^+}/\hbar$ and the inter nuclear vector $\mathbf{R}$ at the dissociation limit or the measured breakup direction. (a) and (c) show time dependent quantum mechanical calculations for $|\mathbf{k}| = 4$ a.u. and 7 a.u. at the transversal plane. The results of calculations for different vibrational states have been added incoherently. The right panels show experimental results for (b) 3.5 a.u. $< |\mathbf{k}| < 4.5$ a.u. and (d) 6.5 a.u. $< |\mathbf{k}| < 7.5$ a.u. for an angle θ between the direction of impact and dissociation of between 80° and 100°.

Finally, it is interesting to reflect on whether it is possible to incorporate the concept of a coherent momentum transfer into classical dynamics. To investigate this hypothesis we modify our semi-classical model by assuming that, in each collision, the momentum transfer is divided equally between both nuclei. The result presented in Fig. 3(d) shows excellent agreement with both the experimental results [Fig. 2(a)] and those from the fully quantum mechanical treatment of the slit dynamics [Fig. 2(c)]. This shows that the process of coherent momentum transfer indeed possesses a physical analog in the classical world: the momentum transfer is shared by the scattering centers even though the forces along the classical trajectories involve only a single scattering center.

In conclusion, we have observed Young-type interferences behind a free-floating isotope-labeled molecular double pinhole and measured the momentum transfer. Consistent with Bohr's arguments, a quantum mechanical description of the molecular slit dynamics is appropriate to describe the observed interference phenomena. Moreover, it is sufficient to completely define the system dynamics; no additional treatment of the scattered projectile is necessary to describe the interference phenomena. Momentum transfer from the projectile to the slit is shown to modify

the interference features in full agreement with predictions from quantum modeling the kicked-molecule slit dynamics. As an alternative to a quantum mechanical description of the slits, our results show that a classical description of the slits according to Einstein's original viewpoint of the thought experiment is still possible. In that case one has, however, to assume a delocalized nonclassical interaction. Interestingly, for the specific pathway-symmetric thought experiment of Fig. 1(a) this net interaction would not lead to a recoiling of the slits.

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[1] N. Bohr, in *Albert Einstein: Philosopher Scientist* (Cambridge University Press, Cambridge, England, 1949), p. 201.
[2] Y. Shi, Ann. Phys. (N.Y.) **9**, 637 (2000).
[3] W. K. Wootters and W. H. Zurek, Phys. Rev. D **19**, 473 (1979).
[4] P. Bertet, S. Osnaghi, A. Rauschenbeutel, G. Nogues, A. Auffeves, M. Brune, J. M. Raimond, and S. Haroche, Nature (London) **411**, 166 (2001).
[5] L. Ph. H. Schmidt, S. Schössler, F. Afaneh, M. Schöffler, K. Stiebing, H. Schmidt-Böcking, and R. Dörner, Phys. Rev. Lett. **101**, 173202 (2008).
[6] R. N. Zare, Mol. Photochem. **4**, 1 (1972).
[7] R. N. Zare, J. Chem. Phys. **47**, 204 (1967).
[8] L. Ph. H. Schmidt, T. Jahnke, A. Czasch, M. Schöffler, H. Schmidt-Böcking, and R. Dörner, Phys. Rev. Lett. **108**, 073202 (2012).
[9] S. Chelkowski, P. B. Corkum, and A. D. Bandrauk, Phys. Rev. Lett. **82**, 3416 (1999).
[10] Z. Amitay *et al.*, Phys. Rev. A **60**, 3769 (1999).
[11] T. F. Tuan and E. Gerjuoy, Phys. Rev. **117**, 756 (1960).
[12] O. Jagutzki, V. Mergel, K. Ullmann-Pfleger, L. Spielberger, U. Spillmann, R. Dörner, and H. Schmidt-Böcking, Nucl. Instrum. Methods Phys. Res., Sect. A **477**, 244 (2002).
[13] R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshammer, and H. Schmidt-Böcking, Phys. Rep. **330**, 95 (2000).
[14] J. Ullrich, R. Moshammer, A. Dorn, R. Dörner, L. Ph. H. Schmidt, and H. Schmidt-Böcking, Rep. Prog. Phys. **66**, 1463 (2003).
[15] W. Domcke and L. S. Cederbaum, J. Electron Spectrosc. Relat. Phenom. **13**, 161 (1978).
[16] E. Kukk *et al.*, Phys. Rev. Lett. **95**, 133001 (2005).
[17] M. H. Beck, A. Jäckle, G. A. Worth, and H.-D. Meyer, Phys. Rep. **324**, 1 (2000).