Young-Type Experiment Using a Single-Electron Source and an Independent Atomic-Size Two-Center Interferometer

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Evidence is given for Young-type interferences caused by a single electron acting on a given double-center scatterer analogous to an atomic-size double-slit system. The electron is provided by autoionization of a doubly excited He atom following the capture of the electrons of H_2 by a He^{2+} incoming ion. The autoionizing projectile is a single-electron source, independent of the interferometer provided by the two H^+ centers of the fully ionized H_2 molecule. This experiment resembles the famous thought experiment imagined by Feynman in 1963, in which the quantum nature of the electron is illustrated from a Young-like double-slit experiment. Well-defined oscillations are visible in the angular distribution of the scattered electrons, showing that each electron interferes with itself.

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The wave nature of the electron has been demonstrated in several experiments, by observing diffraction or interference patterns resulting in electron scattering on matter [1–6]. The most famous example is given by the Davisson-Germer experiment [1]. In this experiment, electrons from a heated filament were accelerated and scattered on the surface of a nickel metal. Structures in the intensity of scattered electrons with increasing accelerating voltage were observed, showing the wave behavior of the electrons. In other experiments [2–4], the authors tried to set up the well-known Young double-slit experiment using electrons instead of light, to show that each electron interferes only with itself. As pointed out by Feynman in 1963, the difficulty in realizing this kind of experiment lies in the fact that, because of the short wavelength of the electron, the distance between the two slits has to be of atomic-scale size, otherwise the spacing of the interference fringes would be too narrow to discern the interference pattern [7]. Moreover, to show that each electron interferes only with itself, another challenging condition is to prevent any chance of finding two or more electrons in the double-slit apparatus at the same time (single-electron condition).

In 1961, Jönsson performed for the first time an actual double-slit experiment with electrons [2]. The electron beam was produced by a 50-kV electron source of common variety [2]. Jönsson succeeded in showing an electron-interference pattern by using very narrow slits (0.5 μm wide and $\sim$0.1 μm apart) in a copper foil, and large distances between the slits and the observation screen. Several years later, Merli *et al.* [3], and then Tonomura *et al.* [4], performed another fascinating electron-interference experiment, making the attempt to reach the single-electron regime in order to demonstrate that each electron interferes with itself. Their method consisted in attenuating the intensity of the electron beam produced in an electron microscope equipped with an electron biprism and a position-sensitive detector. The biprism, invented in 1956 by Möllenstedt and Dücker [8], consists of two parallel grounded plates with a fine filament between them, at a positive potential. This apparatus is akin to the biprism proposed by Fresnel and to the ''slip of card'' interferometer used by Young in an early demonstration [9]. More recently, interference effects in the ionization of H_2 or D_2 molecules by energetic ionized particle impact [10–12] were observed. These effects were considered to be linked to Young's experiment. However, in these latter experiments, the interfering electron originates in the two-center scatterer itself, while in Young's historical experiment the light was emitted by an independent source [13].

Here, we report on the successful realization of a novel atomic-scale Young-type experiment, in which one single electron scatters on a double-center system, analogous to a double-slit apparatus. This experiment was been proposed and theoretically studied two years ago [14]. An incoming He^{2+} ion captures both electrons from a H_2 molecular target. After this primary process, the fully ionized H_2^{2+} target dissociates into two H^+ fragments. The two electrons are captured either on nonautoionizing $1snl'$ ($n \geq 2$) configurations (l' is the angular momentum of the electron) or on doubly excited $2lnl'$ ($n \geq 2$) configurations [15], with nearly equal probabilities. In the latter case, the outgoing He projectile may autoionize (with probability close to unity), emitting an electron of well-defined energy in any direction. Hence, when emission takes place in the backward direction, the autoionized electron scatters on the two H^+ centers which play the role of an atomic-size double-slit apparatus. The dimension of this double-slit system ($\sim$1 nm) is of the same order of magnitude as that of the interfering-electron wavelength ($\sim$0.2 nm). In this collision event, the doubly excited He projectile is a

source of a single electron, which can be coherently scattered on the two H^+ fragments. An elementary single-electron interference experiment is thus performed in this collision event, where the electron emitter and the interferometer are independent from each other and distinctly separated. The repetition of such collision events leads to a succession of independent elementary experiments in which the interferometer is destroyed after a single electron has passed through, thus providing an unquestionable method to fulfill the single-electron condition [14]. The present experiment thus resembles the famous ''thought experiment'' imagined by Feynman in 1963 [7], in which the quantum nature of the electron is illustrated from a Young-like double-slit experiment. Here, similarly to the case of Young's experiment with light [16], the interference manifests itself in periodic fringes in the electron yield.

The present experiment was performed at GANIL (Grand Accélérateur National d'Ions Lourds), in Caen (France). Details of the scattering chamber and the electron spectrometer have been described previously [17], and so only a brief description is given here. A $^3He^{2+}$ ion beam with a velocity of 1.4×10^6 m s^{-1}, corresponding to a kinetic energy of 30 keV, collided with a gas jet of H_2 molecules leading to a uniform H_2 pressure in the collision region. The densities of the gas target and of the projectile beam were $\sim 2 \times 10^{11}$ cm^{-3} and 10^5 cm^{-3}, respectively. Thus, the average distance between two neighboring He^{2+} projectiles is thus larger than $l_{\text{proj}} \sim 100$ μm.

Let us now consider the single-electron condition stated above in more detail. Since the double-capture probability p_{DC} is $\sim 10^{-6}$–10^{-7} and since each autoionizing projectile cannot emit more than one electron, it can be stated that the average distance between two neighboring single-electron emitters is of the order of $l_{\text{far}} \sim 1$ cm (which is much larger than l_{proj}). In comparison, the distance l_{near} between a given He^{**} single-electron source and its $H^+ + H^+$ target partner as well as the distance between the two H^+ scatterers are generally smaller than 10 nm when the Auger electron is emitted. Under these conditions, we can estimate the ratio between the probability p_2 that two electrons scatter on a given two-center system and the square of the probability p_1 that only one electron reaches this scatterer, as given by $p_2/p_1^2 \sim (l_{\text{near}}/l_{\text{far}})^2 \sim 10^{-12}$–$10^{-13}$. This result ensures the condition $p_2/p_1^2 \ll 1$ to be fulfilled in a single-electron regime, as it was shown in quantum optics for the similar case of single-photon interference experiments (see [18] and references therein).

To show the realization of the single-electron condition, another complementary approach consists in considering the different characteristic times involved in the collision. The flux of He^{2+} projectiles passing through the target gas did not exceed $\eta \sim 5 \times 10^{12}$ ions per second. Since each autoionizing projectile (produced in a double-capture event) emits exactly one electron, the average time delay between two consecutive single-electron emission events is larger than $\Delta t_e \sim 1/(\eta \cdot p_{\text{DC}}) \sim 200$ ns (when assuming $p_{\text{DC}} = 10^{-6}$). On the other hand, the time duration t_c of the collision process (including double electron capture, autoionization, and electron scattering at the two-center H^+-H^+ system) is of a few femtoseconds. Since the collision processes have an extremely small duration t_c compared to their repetition time Δt_e, each electron scattering event can be considered to be well separated and independent from others. Hence, the present characteristic distances and times lead to the feature that elementary single-electron interference experiments are performed in individual collision processes well separated from each other both in time and space. Since these individual scattering processes are repeated with similar initial conditions many times, what is actually measured here is the ensemble probability of the diffraction of just one single electron by one single two-center scatterer.

A spectrometer, which consists of an electrostatic parallel-plate analyzer, was used to detect the emitted electrons as a function of their energy and emission angle [17]. A typical energy distribution, recorded at an observation angle of 150° with respect to the incident beam direction, is shown in Fig. 1. Two distinct contributions are seen: (i) a monotonically decreasing part originating from direct ionization of the target molecule and, (ii) a group of peaks in the range ~10–18 eV, due to autoionization of the projectile after double electron capture into $2lnl'$ ($n \geq 2$)

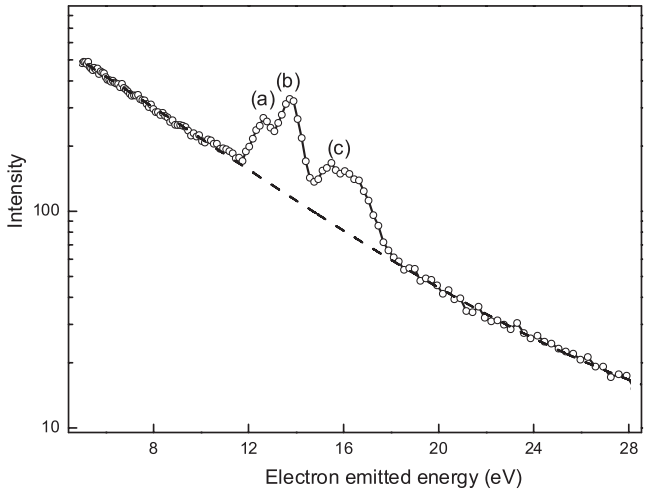

FIG. 1. Energy distribution for electron emission in 30 keV $^3He^{2+} + H_2$ collisions, at a detection angle of 150°, with respect to the incident beam direction. The intensity, as well as the electron energies, are given in the laboratory frame. The continuously decreasing contribution originates from the direct ionization of H_2. The superimposed structures are related to autoionization of He following the production of doubly excited states: peak (a), He($2s^2\,^1S$); peak (b), He($2s2p\,^1P$) and He($2p^2\,^1D$); group of peaks (c), He($2lnl'$) with $n \geq 3$. The dashed curve is the result of a polynomial fit (order 3 in the lin-log scale) of the direct ionization contribution.

doubly excited configurations. The peaks (a), (b), and (c) are associated with $2s^2\ {}^1S$, $2l2l'$ ($2s2p\ {}^1P$ and $2p^2\ {}^1D$) and $2lnl'$ ($n \geq 3$) states, respectively. Since the fluctuations of the beam intensity and the target density were less than ~0.3%, only the statistical uncertainties were taken into account. Whatever the detection angle, they were found to be less than 2%, except at electron energies larger than those found for autoionization, where they can reach ~3%.

It should be noted that at the present collision velocity, the observed autoionization peaks are not significantly affected by interferences with the direct ionization term, which becomes significant at larger projectile velocities. In fact, previous experiments on the $H^+ + He$ system demonstrated that below $2 \times 10^6\ m\,s^{-1}$ impact velocity, interferences among autoionization lines and with the direct ionization term are small compared to the pure autoionization contribution [19].

To single out the autoionization contribution, the direct ionization part was fitted with a polynomial function in the lin-log scale (dashed curve in Fig. 1), and subtracted from the experimental data. The degree (2 or 3) of the fitting polynomial did not affect the final result, within less than 1%. Thus, the relative autoionization intensity was determined with a total uncertainty of ~3% at maximum. Finally, to obtain the angular dependence of autoionization, the resulting spectra were integrated over the emitted electron energy.

The integrated intensity is presented in Fig. 2(a) (full circles) as a function of the detection angle θ_d in the range 95°–160°. At first sight, the intensity is found to increase with increasing θ_d. This increase is partly due to an instrumental effect since the collision length as viewed by the spectrometer increases significantly with increasing θ_d between 90° and 160°. Nevertheless, a careful inspection of the data reported in Fig. 2(a) shows the existence of at least three oscillations superimposed on the main dependency. From the intensity, the autoionization cross section was determined by taking into account the aforementioned collision length. As shown in previous experiments [20], this length increases according to $l = l_o/\sin\theta_d$ (l_o is the length determined at 90°) with increasing detection angle θ_d between 90° and 160°. The result for the cross section is presented in Fig. 2(b). Well-defined oscillations with pronounced maxima and minima are visible in the angular distribution of the electron emission cross section, providing clear evidence for the interference pattern. To further analyze the latter periodic structure, one may refer to previous theoretical research [14]. In a single-scattering approximation, the detector can be reached by the autoionizing electron through three different trajectories, either directly or after being scattered by the target nuclei. The interference between the direct and scattered waves is known to produce very small effects that are hardly distinguishable from those due to adjacent overlapping resonances [21]. This is partially due to the fact that the phase shift depends on the distance between the He^+ outgoing projectile of velocity $\vec{v}_P$ and the center of mass of the molecule. This distance varies with time and is not the same for every event. Therefore the oscillations do not share a common frequency and are almost completely destroyed in the coherent sum. On the other hand, the phase shift between the waves scattered by the two nuclei reads $\varphi \approx \vec{s} \cdot \vec{d}/\hbar$ [14]. Here, $\vec{s} \approx m(\vec{v} - \vec{v}_o)$ is the momentum transferred to the electron of mass m, where $\vec{v}_o$ and $\vec{v}$ are its velocity in the laboratory frame before and after the scattering by the nuclei, respectively. The phase depends on the relative position $\vec{d}$ of the H^+ centers, which also varies from event to event, both in magnitude and orientation. However, the interference pattern is not washed out as in the former case. This is due, in part, to the very slow evolution of the Coulomb explosion in

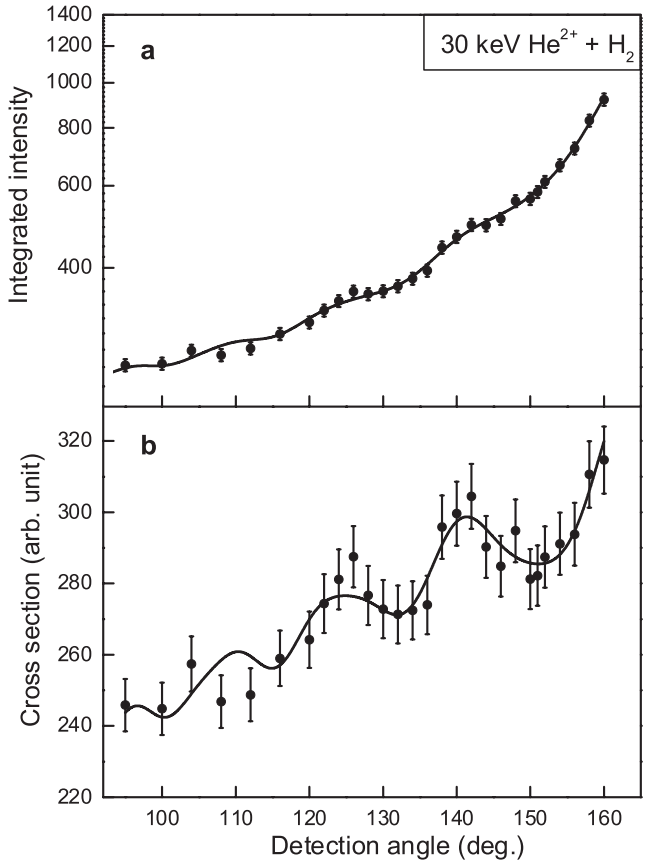

FIG. 2. Total intensity (a) for autoionization following double electron capture in 30 keV $^3He^{2+} + H_2$ collisions, as a function of the detection angle (solid circles). The autoionization intensity is given in the frame of the autoionizing He^{**} doubly excited atom. Superimposed to a main increase of the intensity, at least three oscillations are visible. The oscillations are well reproduced (full curve) by using, in the fitting procedure, the interference Debye-Ehrenfest term (see text). In (b) differential cross section for autoionization, derived from the intensity, reveals the significance of the interference pattern.

comparison with the recession of the projectile and the sudden passage of the autoionizing electron [14]. Furthermore, averaging the Young interference term $\cos\varphi$ over the orientation of the molecule yields $\langle\cos\varphi\rangle = \frac{1}{2}\times \int_{-1}^{1}\cos(\delta\cos\xi)d\cos\xi = \sin\delta/\delta$, where we have defined $\delta = sd/\hbar$. A simple calculation shows that the momentum transfer can be written as $s = 2h\cos(\theta_d/2)/\lambda$, where $\lambda = h/mv$ is the de Broglie wavelength of the electron [14]. This term $\sin\delta/\delta$ with $\delta = 4\pi\cos(\theta_d/2)d/\lambda$ was first derived by Debye [22] and independently by Ehrenfest [23] in 1915, but applied to the diffraction of x rays by molecules. It shows that the average in the molecule orientation only vanishes when δ is a nonzero multiple of π, thus preserving an oscillatory structure in the electron detection angle θ_d [10,14]. This Debye-Ehrenfest term was fitted to the oscillatory structure, as shown in Fig. 2(b) to get $d \approx 8.6 \pm 0.2$ Å, in good agreement with the theoretical estimate of the order of $d \approx 8.7$ Å obtained for the $2s^2\ ^1S$ line by means of the model [14] while evaluating the average internuclear separation d at the time when the electron reaches the molecule. This result leads to a pseudoperiod of $\sim 17°$ in the interference pattern [Fig. 2(b)].

The excellent fitting provided by the Debye-Ehrenfest term [Fig. 2(b)] is a clear demonstration that an atomic-scale Young-type interference experiment with each single electron acting on a single two-center system has been successfully realized in the present work. It is shown here that each electron interferes with itself. This experiment, with its new distinct features, might not only serve as a clear illustration of an elementary quantum effect, but also help to gain new insight into a current and important trend of research in atomic, molecular, and optical physics [10–15,18]. For example, the fact that the average distance d between the slits depends on the projectile velocity v_p and on the autoionization rates Γ_i of the projectile [13,14] leads to the striking feature that the average size of the atomic interferometer could be controlled by modifying v_p and/or the projectile and target species. We believe that investigating the interferometer-size dependence of the interference pattern would be an outstanding and complementary manner to further illustrate the elementary quantum effect studied here. Furthermore, in view of the theoretical predictions [14] and the present experimental results, it is likely that the use of a coincidence technique would allow for significantly enhancing the visibility of the interference pattern by selecting the orientation of the molecular target.

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