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# Linewidth oscillations in a nanometer-size double-slit interference experiment with single electrons

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In this article we provide experimental evidence of an interference phenomenon that, to the best of our knowledge, has so far not been observed with either matter or light. In a nanometer-sized version of Feynman's famous two-slit "thought" experiment with single electrons, we managed to observe that the width of a quasi-monochromatic line oscillates with the detection angle. Furthermore, we find that it occurs in counterphase with the line intensity. We discuss the underlying mechanism that produces this unexpected result.

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In 1963 Feynman *et al.* proposed a "gedanken" experiment [1] that, by reproducing the 1807 Young's two-slit demonstration [2] with electrons instead of light, was intended to exemplify the dual-wave-particle nature of matter, one of the main cornerstones of modern physics. In 1923, simultaneously with the confirmation provided by the Compton experiment [3] of Einstein's "energy quantum" of light [4], de Broglie advanced the idea of the wave-like nature of a moving matter particle [5]. Since then, this wave-particle duality and the concomitant analogy between light and matter rank as paradigms of our basic understanding of nature. Even though Feynman himself warned that nobody should try to set up the electron interference experiment since "the apparatus would have to be made on an impossibly small scale," [1] its viability was recently demonstrated and tested [6,7]. The complementary experiment with single photons was pioneered by Taylor in 1909 [8] and demonstrated by Grangier *et al.* in 1986 [9]. In all these pedagogically clean experiments, some of which have become standard in modern physics courses, each single electron or photon hits the screen like a particle, but traverses the slits like a wave. Thus, over many repetitions, an interference pattern builds up as oscillations of the intensity with the observation angle.

Let us recall that in these experiments, and even with a quasi-monochromatic source, the actual wavelength distribution $dI/d\lambda$ at a given observation angle $\theta_d$ would be characterized by a well-defined width at half maximum $\Delta\lambda$. As we explained previously, interferences usually manifest themselves as oscillations of the total intensity $I(\theta_d) = \int dI/d\lambda\, d\lambda$. The question arises whether it would be possible to observe similar oscillations in other quantities, as, for instance, the linewidth $\Delta\lambda$ itself. Note that this question is not as trivial as it might look at first glance. No such oscillations are expected to occur in the standard two-slit experiment with photons.

In general, it is straightforward to demonstrate that for a quasi-monochromatic line with a very narrow wavelength range, the dependencies of $dI/d\lambda$ on $\theta_d$ and $\lambda$ would not be significantly intertwined, and therefore $\Delta\lambda$ would not oscillate or would only present a very mild dependence on $\theta_d$ at the most. On the contrary, a strong oscillation of $\Delta\lambda$ with $\theta_d$ would mean that a different and yet undiscovered mechanism is at work.

Thus, our goal is to produce a nanometer-size setup that would not only demonstrate Young's interference of single electrons as proposed by Feynman *et al.* [1] but would also do this in such a way that the linewidth would oscillate with an amplitude comparable to that of the intensity. One advantage of this analysis over more standard procedures is that the measurement of the linewidth would not require a normalization of the spectra in intensity, as was the case in our previous analysis [7].

But let us point out that any experiment that—as is this one—is set up to resemble Feynman's thought demonstration must meet the following basic but challenging conditions [10].

1. The apparatus has to be nanometer sized, otherwise the spacing between the interference fringes would be too narrow to be discerned.
2. The interferometer has to consist of two slits or centers, so as to mimic the demonstration proposed by Young in 1807 [2].
3. The source and the two-center scatterer have to be distinctly separated [7,10]. In addition, to provide evidence that one electron interferes with itself, a fourth condition has to be fulfilled:
4. As for the case of photons [11], no more than one single electron must be in the double-slit apparatus at any given time.

In a recent article [7], we studied the process

$$He^{2+} + H_2(1\sigma_g) \rightarrow He^{**}(2\ell n\ell', n \geqslant 2) + H^+ + H^+ \quad (1a)$$
$$\rightarrow He^+ + H^+ + H^+ + e^- \quad (1b)$$

as a nanometer-sized realization of Feynman's experiment, where the outgoing autoionizing helium atom plays the role of the source of a single electron emitted with a wavelength $\lambda$ of the order of a few angstroms, while the two residual protons provide the double-center interferometer. In the lapse of time between the double-electron capture (1a) and the electron emission process (1b), as characterized by the autoionization mean lifetime, the two protons reach a nanometer separation $d$. This Coulomb explosion represents an essential piece of the studied process, since otherwise the interference fringes would be separated by more than 180° and therefore would not be visible [6].

In contrast to other previous experiments [12–16] the one studied by us and reported in Ref. [7] fulfills all four aforementioned conditions stated for a Feynman-type

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demonstration. In particular, with a degree of second-order coherence of the order of $10^{-12}$ [7], it unquestionably ensures the single-electron condition, in an *ensemble* of independent elementary experiments in which the two-proton interferometer is destroyed after being traversed by the single electron emitted by the helium source [6].

In our previous article [7], the intensity $I(\theta_d)$ was measured for different detection angles $\theta_d$ in a range from 95° to 160° with respect to the incident beam direction. More recently, this measurement was extended with an independent experimental setup, all the way into the backward direction (around 180°) [17,18]. The intensity presented very well-defined oscillations superimposed on the main angular dependency. These oscillations could be fitted with a Bessel function of zero order [10,19,20],

$$j_o(\delta) = \frac{\sin\delta}{\delta}, \tag{2}$$

with $\delta = \frac{4\pi d}{\lambda}\cos(\theta_d/2)$, where $\lambda$ is the electron wavelength in the laboratory rest frame. Note that in analogy with double-slit interferences with photons, two parameters play a fundamental role in this expression: the distance $d$ and the wavelength $\lambda$ [21]. Due to the Coulomb explosion of the $H_2$ molecule and the Doppler shift originating from the motion of the helium atom with velocity $v_p$ in the laboratory reference frame, they depend on quantities such as the velocity $v_p$ and the detection angle $\theta_d$, and so does the interference pattern. The average distance $d$ between the protons when the electron is emitted was found to be $0.86 \pm 0.02$ nm, which is very close to the theoretical estimate for the given experimental conditions [6,7].

Unfortunately, this previous experiment was not suited for the study of the linewidth since (1) the data correspond to undiscerned $2lnl'$ ($n \geqslant 2$) lines and (2) the resolution was not enough to separate them. But now, by using spectra obtained at a resolution higher than that of those previously reported in Ref. [7], we could focus on a single Auger line originating from the deexcitation of the $2s^2\,{}^1S$ configuration and determine its linewidth at observation angles ranging from 120° up to 160°.

The experiment was carried out at the ARIBE (Accélérateur pour la Recherche avec les Ions de Basse Energie) facility at GANIL (Grand Accélérateur National d'Ions Lourds) in Caen, France. Details of the scattering chamber and the electron spectrometer have been explained previously [22], so only a brief description is given here. A beam of ${}^3He^{2+}$ ions with a typical current of ~200 nA was extracted from a 14.5-GHz electron cyclotron resonance source, accelerated at 30 keV, magnetically analyzed to avoid contamination by undesirable ions, focused, directed into a scattering chamber, and finally collected into a Faraday cup. At the center of the scattering chamber, the ion beam crossed an effusive gas jet of $H_2$ molecules with a density of $\sim 10^{11}$ cm$^{-3}$. The emitted electrons were detected and analyzed by means of an electrostatic parallel-plate spectrometer which could be positioned at angles from 20° to 160°, with respect to the incident beam direction. The high-resolution spectra were obtained by decelerating the emitted electrons at the entrance of the spectrometer.

Typical Auger electron spectra obtained at high resolution are presented in Fig. 1, as a function of the electron energy $E$ in the laboratory frame, at detection angles of 120° and 160°. As shown previously [7], each spectrum

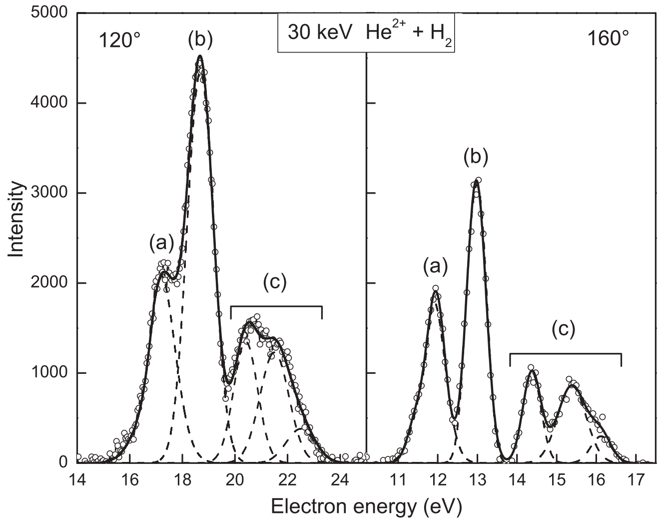


FIG. 1. Autoionization contribution in 30-keV $He^{2+} + H_2$ collisions, at detection angles of 120° (left side) and 160° (right side), obtained by subtracting the direct ionization part from the total intensity. The autoionization structures are fitted in order to separate the different configurations. The peaks (a) and (b) are due to the deexcitation of configurations $2s^2\,{}^1S$ and $(2s2p\,{}^1P\text{–}2p^2\,{}^1D)$, respectively, while the structures (c) originate from the deexcitation of the $2lnl'$ ($n \geqslant 3$) configurations. The dashed curves fit the different Auger structures (see text).

consists of structures originating from Auger deexcitation following double-electron capture. The peaks (a) and (b), centered at the lowest energies, are due to the deexcitation of configurations $2s^2\,{}^1S$ and $(2s2p\,{}^1P\text{–}2p^2\,{}^1D)$, respectively, while the structures (c) originate from the deexcitation of the $2lnl'$ ($n \geqslant 3$) configurations. Due to kinematics effects, the structures (a) and (b) partly overlap, especially at angles close to 90°. This explains why the present analysis is restricted to angles larger than 120°. In addition, since the autoionization process occurs in the Coulomb field of the two protons, the profile of the Auger structures is modified by postcollisional effects [23]. Thus, to separate each structure (dashed curves in Fig. 1), Gaussian curves distorted by a function of the type $\exp\{a_o \arctan[b_o/(E - E_o)]\}$ were used [24]. The quantity $E_o$ is the energy at the maximum of the structure, while $a_o$ and $b_o$ are fitting parameters.

The experimental linewidths are presented in Fig. 2 (open circles), as a function of the detection angle in the range 120°–160°. In addition to the enhancement of the natural width due to the resolution of the spectrometer, which for the present case is $\Delta E_{\text{res}} \sim 0.4$ eV, other broadening effects can increase the widths [25]. They are mainly due to the variations of the polar and azimuthal detection angles and the Barker and Berry effect [26]. These four contributions are represented in Fig. 2 (dashed-dotted curves). The corresponding total width $\Delta E_{\text{tot}}$ (dashed curve) is in excellent agreement with experiment. Furthermore, it shows a decrease from 1.3 eV down to 0.7 eV for increasing angles that is consistent with the data. However, within the uncertainties, which are smaller than 0.02 eV, well-defined oscillations (solid curve in Fig. 2) are clearly visible, superimposed on the main dependency.

In Fig. 3 we compare the angular dependence of the width and maximum of the $2s^2\,{}^1S$ line. The oscillations were singled

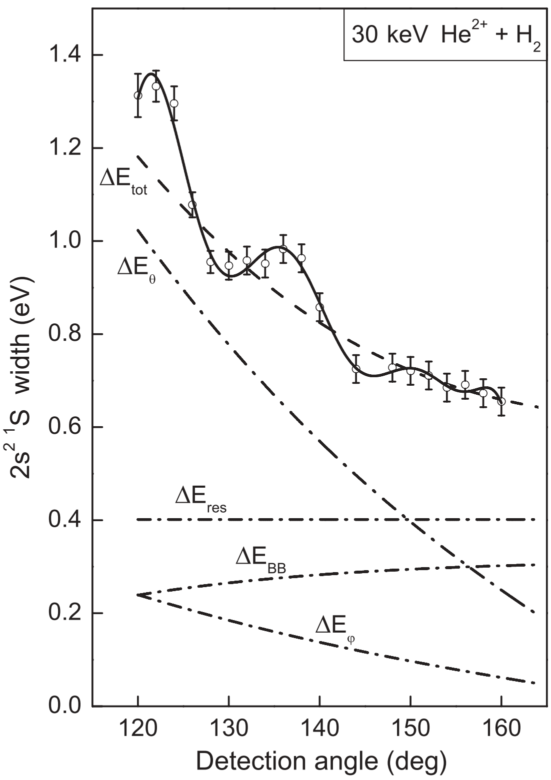


FIG. 2. Experimental width of the $2s^{2}\,{}^{1}S$ line (open circles) in 30-keV $He^{2+} + H_2$ collisions, as a function of the detection angle. The solid curve fits the experimental data, using Eq. (3). The contribution $\Delta E_{\rm res}$ originates from the resolution of the spectrometer. The broadenings $\Delta E_\theta$ and $\Delta E_\varphi$ are due to the variations $\Delta\theta$ and $\Delta\varphi$ of the polar and azimuthal angles of the electron, respectively, whereas $\Delta E_{BB}$ is caused by the Barker and Berry effect. The corresponding total width $\Delta E_{\rm tot}$ is also reported (dashed curve).

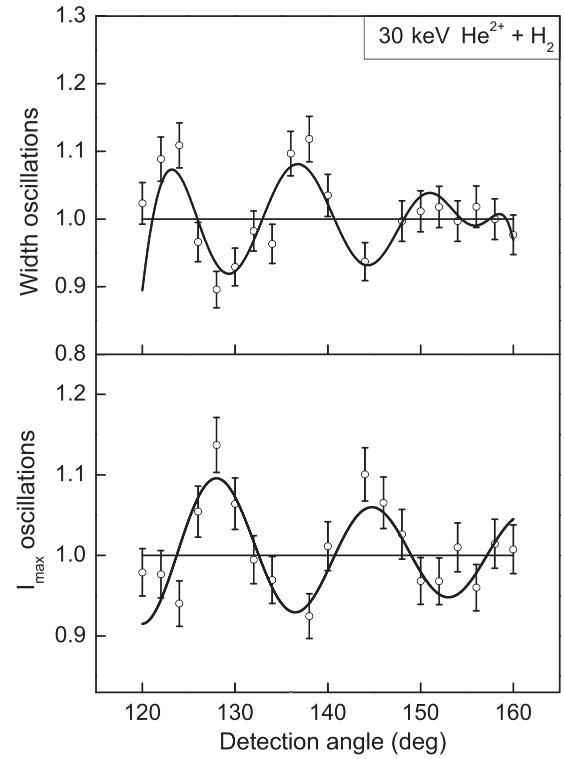


FIG. 3. Oscillating terms in the experimental width and maximum intensity of the $2s^{2}\,{}^{1}S$ line in 30-keV $He^{2+} + H_2$ collisions as a function of the detection angle. The oscillating terms are obtained in both curves by dividing the experimental data by a monotonous background obtained by fitting the data with a second-order polynomial function. The solid curves fit the experimental results, using Bessel functions of zero order and first order.

out by dividing both curves by the main monotonous dependence. We see in Fig. 3 that both the width and the maximum intensity of the $2s^{2}\,{}^{1}S$ line oscillate with about the same period, which precisely coincides with the one obtained for the total yield in Ref. [7]. This is not entirely surprising taking into account that the same Young-type interference mechanism is the source of all the oscillations reported. However, there are two other results that immediately strike the eye. One is that the relative amplitude of the linewidth oscillations is comparable to that of the maximum intensity. The second is entirely unexpected and seems to indicate that the width and the maximum intensity oscillate in counterphase. In what follows we provide a simple interpretation of these findings.

Let us write the amplitude in the intensity $I(E,\theta_d) = |A|^2$ as a sum of three terms $A = A_o + A_+ + A_-$, where $A_\pm$ and $A_o$ are related to whether the electron has or has not been rescattered by the protons, respectively [27]. Besides the direct autoionizing intensity $|A_o|^2$ and a cross term, $2\,\mathrm{Re}(A_oA_+^* + A_oA_-^*)$, that describes the path interference between the direct and scattered autoionization amplitudes, the remaining contribution $|A_+ + A_-|^2$ is responsible for the Young-type interference itself between the waves scattered on both protons. Of course, the path interference between the direct and scattered amplitudes in $2\,\mathrm{Re}(A_oA_+^* + A_oA_-^*)$ can also produce oscillations, but they are significant only in the close vicinity of the backward direction ($\theta_d > 160^\circ$), outside the angular range of the presently reported data [28,29]. Furthermore, as shown by Moretto-Capelle *et al.* [30] the interference between overlapping resonances can be neglected. Thus, the oscillations observed in the width and maximum of the $2s^{2}\,{}^{1}S$ line for $He^{2+} + H_2$ collisions can be unambiguously associated with a Young-type double-slit interference mechanism.

Averaging on the molecule orientations, the dependence of $|A_+ + A_-|^2$ on the separation $d$ between the protons is fully ascribed to the $j_0(\delta)$ factor in Eq. (2) [6] and is responsible for the oscillations of the maximum intensity observed in Fig. 3 [7]. However, it is clear that since this multiplicative factor affects the $|A_+ + A_-|^2$ term alone, it does not suffice by itself to fully explain the width's oscillations. However, we have to carefully consider the additive contribution of $|A_o|^2$ and $2\mathrm{Re}(A_oA_+^* + A_oA_-^*)$. Thus, we write $I(E,\theta_d) = g(E,\theta_d) + f(E,\theta_d)j_o(\delta)$, where $g(E,\theta_d) = |A_o|^2 + 2\mathrm{Re}(A_oA_+^* + A_oA_-^*)$ and $f(E,\theta_d)j_o(\delta) = |A_+ + A_-|^2$, and determine the linewidth as the sum of the half widths $\Delta E_\pm$ on both sides of the maximum $E_o$ by using the relation $I(E_o,\theta_d)/2 = I(E_o \pm \Delta E_\pm,\theta_d)$. By performing a Taylor expansion of $j_o(\delta)$ and the auxiliary functions $g(E,\theta_d)$ and $f(E,\theta_d)$ to first order in energy at the autoionization line, we obtain

$$\Delta E \approx \Delta E_o - a(\theta_d)j_o(\delta) + b(\theta_d)j_1(\delta), \qquad (3)$$

where $j_1(\delta) = -dj_o/d\delta$ is the Bessel function of first order, $\Delta E_o$ is the total width in the absence of any oscillation, and $a(\theta_d)$ and $b(\theta_d)$ are nonoscillating and positive functions of $\theta_d$. Note that if $a(\theta_d)$ were negative, then the presence of the first-order Bessel function in expression (3) would induce a phase shift of only $\pi/4$ with the intensity oscillations, that is, two times smaller than observed experimentally. In contrast, a positive value of $a(\theta_d)$ gives rise to a phase shift of $\pi/2$, and so the linewidth should be in counterphase with the maximum intensity, as experimentally observed.

The experimental width distribution (Fig. 3) was fitted using relation (3). Polynomial functions of order 2 were used for $a(\theta_d)$ and $b(\theta_d)$. The factor $a(\theta_d)$ was found to be positive when $\theta_d$ was smaller than 155°, which is consistent with the predictions of the simple model. So, both the experiment and the model agree on the fact that the linewidth oscillates in counterphase with the maximum intensity.

In this article, an interesting aspect of electron interferences is evidenced. Instead of investigating the total intensity of undiscerned $2lnl'$ ($n \geqslant 2$) autoionization configurations, we focused on a single $2s^{2\,1}S$ line and determined its maximum and linewidth at angles ranging from 120° to 160°, where interferences are expected to occur. This detailed analysis revealed well-defined oscillations (Fig. 3) in the angular dependence of both the maximum intensity and the linewidth. The maximum oscillates in phase with the total intensity on the observed angular range, showing that the oscillations of the total intensity are mainly due to the variations of the line maximum. More surprisingly, the $2s^{2\,1}S$ linewidth was found to strongly oscillate (Fig. 2) in counterphase with the maximum (Fig. 3), a fact that can be explained by means of simple theoretical arguments. These results not only provide a Feynman-type demonstration of the presence of a nanoscale Young-type interference of single electrons but also complete and reinforce the analysis performed previously on the undiscerned $2lnl'$ ($n \geqslant 2$) configurations [7].

Finally, the counterphase linewidth oscillations reported in this article might be prone to be found in similar configurations with matter particles and photons. Thus, the present results open the way to valuable research in both atomic and molecular collisions and across field boundaries in optical physics. Presently, atomic collisions and laser experiments are under way at CIMAP in order that comparisons may be made with the present results.

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