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Fast oscillating structures in electron spectra following $He^{q+}+He$ collisions ($q=1,2$) at low projectile energies

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Distributions of ejected electrons following collisions of slow He^+ and He^{2+} ions and a He target were measured for projectile energies of 20 and 40 keV, respectively. The electrons were detected at angles of 30° and 90° with respect to the incident beam direction. Superimposed on a continuous background originating from target ionization, small amplitude, high-frequency oscillations are revealed. The frequency of these oscillations is found to be nearly independent of the projectile charge and observation angle. In view of recent experiments and calculations, the origin of such oscillations is discussed. Processes such as autoionization following the production of highly excited states, Fermi-shuttle ionization, or coherent electron emission caused by interference between the target centered and projectile centered amplitudes, are considered.

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During the last five years, many experimental [1–4] and theoretical works [5–8] have been devoted to the study of interferences resulting from the coherent emission of electrons from H_2 or D_2, following the impact of various projectiles such as ions or electrons. These (first-order) interference effects manifest themselves as oscillations in the electron spectra as a function of the ejected electron velocity [1,3,4]. In addition, oscillations occurring with about 2 or 3 times the frequency of the main oscillatory structure were found [3,9]. These oscillations were interpreted as a second-order effect, where the electron wave emitted at one center interferes with the wave backscattered at the other center. The frequency and the phase shift associated with these oscillations were shown to be nearly independent of the observation angle [3,9] and the projectile velocity [3]. In contrast with the results obtained for a molecular target, no first- or second-order oscillations were revealed in collisions involving ions and an *atomic* target such as He [1,2], confirming that the observed interferences reflect the molecular nature of the target.

Very recently, evidence for significantly higher frequency oscillations was reported in the electron emission spectra of H_2 by fast H^+ impact [3]. Although no conclusion was reached, it was suggested that these oscillations might be due to interference between direct electron emission and autoionization involving continuum doubly excited H_2 states giving rise to so-called free-free transitions. Another explanation, considered not likely for the relatively fast H^++H_2 collisions of Ref. [3], involved coherent electron emission from the transient molecule formed by the passing ion with one (or both) of the target H_2 centers. In other words, the projectile H^+ acts, during the collision, as one of the centers from which the electron is coherently "ejected."

Within the framework of this latter hypothesis, such interference effects would also be expected in collisions of ions with *atomic* targets where the number of centers of the transient molecule is reduced to two. Such interference effects are likely to be more pronounced when the projectile and target centers are the same. As an example, a theoretical study of H^++H collisions at 20 keV has been developed [10]. In this study, interferences between the target-centered and projectile-centered amplitudes were added using finite Hilbert basis-set calculations to describe the angular distribution of the ejected electrons. With the inclusion of the interferences, the theoretical results were found to be in excellent agreement with experimental differential cross sections [11] for detection angles smaller than 90°. Thus, Ref. [10] shows that, even using an *atomic* target, the interferences resulting from coherent electron emission may be revealed.

In the present paper, we report the experimental observation of small but well-defined periodic structures in the velocity distributions of ejected electrons following collisions of 20 keV $^3He^+$ and 40 keV $^3He^{2+}$ with ^4He atoms. The emitted electrons have been observed at detection angles of 30° and 90° with respect to the incident beam direction. In the following, the origin of these structures is discussed. Different processes are proposed, and their consequences are compared with the present results. These results are also compared with those found in fast e^-+He collisions.

The experimental setup has been described in detail in many papers (see, for example, Ref. [12]), so that only a brief description is given here. The experiments were conducted at the 14.5 GHz electron cyclotron resonance (ECR) ion source of the LIMBE (Ligne d'Ions Multichargés de Basse Energie) facility, at the Grand Accélérateur National d'Ions Lourds (GANIL) in Caen. The He^{q+} ions were extracted at a voltage of 20 kV. We used ^3He in the ECR source in order to prevent contamination from H^+ and H_2^+. The ions were collimated to a diameter of ~2 mm before entering the collision chamber. Typical currents of about 50 nA were obtained and collected in a Faraday cup, and were used to normalize the measured electron spectra. Because of the He beam spread, even at the projectile energies used here, care was taken to reduce spurious electrons originating from sur-

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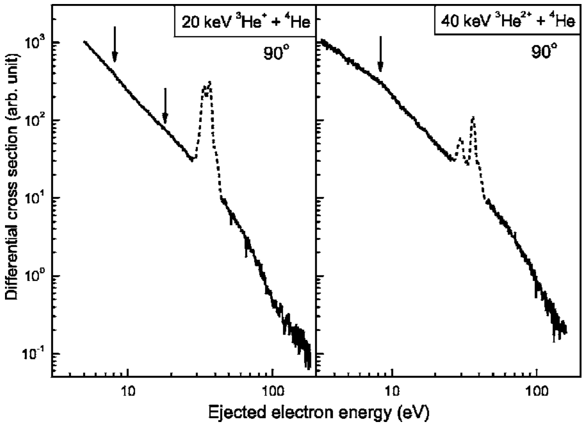

FIG. 1. Doubly differential cross sections associated with the production of electrons in 20 keV He$^+$+He and 40 keV He^{2+}+He collisions, at an observation angle of 90°. Full curves: single ionization; dashed curves: autoinoization following double excitation, double capture, and transfer excitation (see the text). The arrows indicate the presence of broad low-intensity structures.

faces. In the center of the chamber, the He projectiles crossed an effusive gas jet of He. The residual pressure inside the chamber was kept below $\sim 3 \times 10^{-5}$ mbar in order to ensure single collision conditions. The electrons produced in the collision were detected at angles of 30° and 90°, using a single-stage spectrometer consisting of an electrostatic parallel-plate analyzer. The geometric resolution, defined by $R = \Delta\varepsilon/\varepsilon$, where ε is the electron energy and $\Delta\varepsilon$ the full width at half maximum, is constant and of the order of 5%. The doubly differential cross sections were obtained for both collision systems by subtracting the residual background from the spectra, obtained without He and with He, respectively, and then by dividing the result by the electron energy in order to take into account the geometric resolution of the spectrometer.

Figure 1 shows doubly differential cross sections (solid lines) at a detection angle of 90° for 20 keV He$^+$+He (left-side) and 40 keV He^{2+}+He (right-side) collisions. The experimental data, which can be attributed essentially to the single ionization (SI) process, exhibit a monotonic decrease of about four orders of magnitude as the emission energy increases.

Superimposed on the ionization spectra, two structures are observed. The structure centered at ~ 36 eV is due to autoinization following double excitation (DE) of the target, giving rise to $2l2l'$ configurations, while the structures centered at ~ 33 eV (left side of Fig. 1) and ~ 29 eV (right side of Fig. 1) are due to projectile $2l2l'$ configurations caused by transfer excitation and double capture, respectively, which also decay by means of Auger transitions.

Since the Auger electrons partly originate from moving emitters, they are influenced by kinematics. Kinematics effects on the electron energy and intensity are not accounted for in the data of Fig. 1 (dashed lines). Nevertheless, this has no consequence on the forthcoming discussion since we only focus on the ionization contribution of the spectra.

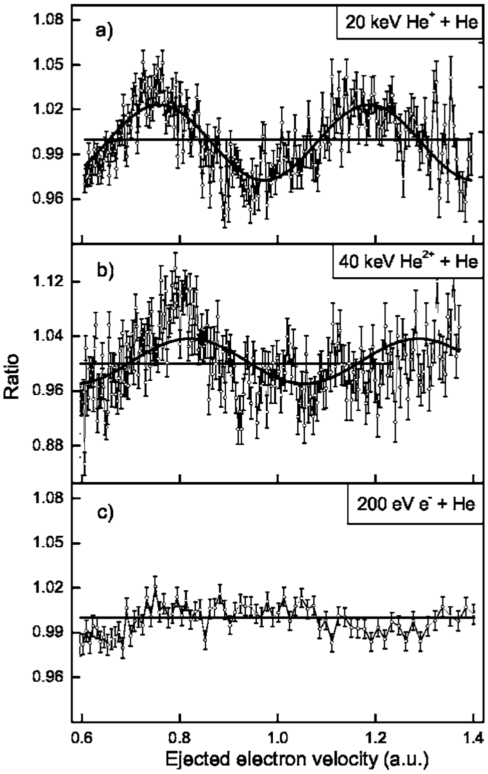

FIG. 2. Ratios (open circles) between experimental doubly differential cross sections and simulated ionization cross sections using polynomial functions, for the collision systems He$^+$+He (a) and He$^+$+He (b) as a function of the emitted electron velocity, at a detection angle of 90°. The ratios are fitted (full curves) using a sinusoidal function. The ratio for 200 eV e$^+$+He (c) collisions is also shown.

A closer inspection of the spectra of Fig. 1 reveals small broad structures centered at ~ 8 eV for both systems and ~ 15 eV for He$^+$+He. These structures are systematically observed at both investigated detection angles. To make more noticeable the eventual presence of oscillations, the experimental cross section of Fig. 1 were divided by a fit function $f(\varepsilon)$, which reproduces the ionization contribution without interferences, in the energy range from 5 to 25 eV. The energy range limitation for the fit is due first to the Auger peaks, and second to the lack of statistics at electron energies larger than 50 eV (Fig. 1). Since the ionization cross sections decrease exponentially, an exponential function was chosen for $f(\varepsilon)$. For a logarithmic scale, this is equivalent to a polynomial function [13], whose degree was varied from 2 to a maximum of 4. Care was taken, by calculating successive derivatives of $f(\varepsilon)$, to generate no extrema in order to avoid nonphysical oscillations.

To verify the validity of the fitting procedure, the same technique was also applied to the collision system e^-+H at 2.4 keV [4]. For ejected electron velocities ranging from 0.4 to 1.4 a.u., corresponding to energies ranging from 2 to 27 eV, respectively, the theoretically determined cross sections shown in Fig. 1 of Ref. [4] were fitted with polynomial functions of order 2. Within the precision of the calculations, the ratios between the theoretical cross sections and the fitting functions were found equal to unity with a maximum deviation of 10^{-3}. In addition, no spurious oscillation was observed, showing that the use of polynomial functions of

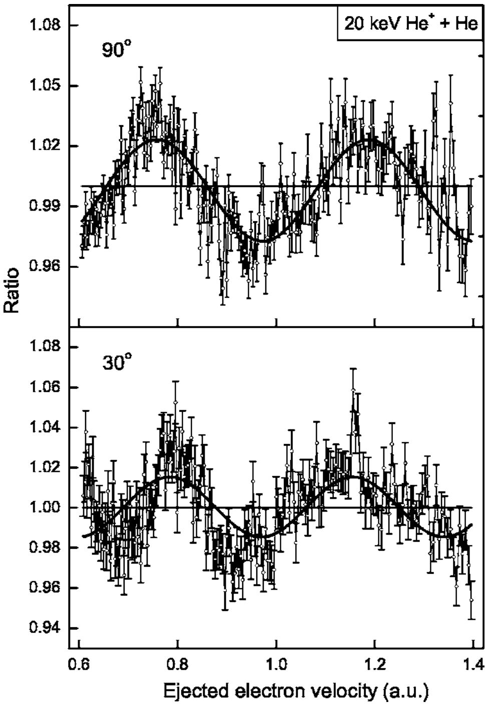

FIG. 3. Ratios (open circles) between experimental doubly differential cross sections and simulated ionization cross sections using polynomial functions, for the collision system He$^+$+He as a function of the emitted electron velocity, at detection angles of 30° and 90°.

low order is justified to describe single ionization without interference.

The ratios between experimental cross sections and simulated ionization cross sections using polynomial functions are shown in Fig. 2 (open circles) for He$^+$+He [Fig. 2(a)] and He^{2+}+He [Fig. 2(b)] at a detection angle of 90°. Two oscillations are clearly visible in the case of the He$^+$+He collision, with maxima at $\sim$0.75 and 1.2 a.u. To better visualize these oscillations, the ratios were fitted with a sinusoidal function (solid curves) of the form $a_o + a_m \sin(2\pi N v_e + \varphi)$, where v_e is the ejected electron velocity, and a_o, a_m, N, and φ are fitting parameters. The error bars, deduced from statistical uncertainties, are found to be less than 1% (the counting rate is larger than 10^4), which is smaller than the amplitude a_m of the oscillations. For the collision system He^{2+}+He, the oscillations are still present in the electron velocity range from 0.6 to 1 a.u., but the lack of statistics at higher velocities does not permit a conclusion concerning the presence of oscillations at velocities larger than 1 a.u.

In the bottom part of Fig. 2, the ratio is also plotted for the collision system e^-+He, for which no oscillation is expected, at a projectile energy of 200 eV (corresponding to a velocity of $\sim$4 a.u.) and a detection angle of 90°. No significant structure appears, supporting the conclusion that the oscillations found for He$^+$ and He^{2+} are not an experimental artifact. Consequently, the projectile plays a determinant role in the emergence of the observed oscillating structures.

In Fig. 3, we plot the ratio for the collision system He$^+$+He at the detection angles of 30° and 90°. The oscillations are clearly visible and reproducible at each angle, further indicating that they are not fortuitous. The amplitude and the frequency of the oscillations are found to be independent of the observation angle and the projectile charge, within their uncertainties. In addition, the frequency is of the same order of magnitude as that found in the case of H$^+$ +H$_2$ collisions [3]. It is interesting to note that the period of the oscillation for He$^+$ ($\sim$0.45 a.u.) is close to the corresponding projectile velocity (0.52 a.u.).

The origin of such oscillations is now discussed. Without the support of precise theoretical calculations, it is impossible to conclude unambiguously that the structures originate from interferences in coherent electron emission from the transient target and projectile centers. Other processes, such as autoionization, Fermi-shuttle process, or possibly transfer ionization (TI), may, in principle, be invoked to explain the creation of these structures.

If an autoionization process is involved, it follows necessarily that configurations of the type $nln'l'$, with n and n' larger than 2 are populated, since the structures are centered at energies lower than those associated with the $2l2l'$ configurations. In addition, the independence of the oscillatory structure on the detection angle shows that any shift due to the Doppler effect is negligible, indicating that the structures originate from deexcitation of the target. However, for the configurations $3l3l'$ and $3l4l'$, simple calculations using the COWAN code [14] give rise to ejected electron velocities of 0.6 and 0.65 a.u., respectively, providing an entirely different structure from that of the two measured maxima (0.75 and 1.2 a.u.). Thus, the production of highly excited states is unlikely to explain these structures.

In previous works, multiple scattering processes have been studied (see [15] and references therein), involving sequences of backscatterings of an emitted electron between an incoming ion and an atomic target. Introducing the shorthand P and T to denote the electron-projectile and electron-target scatterings, respectively, long sequences such as P-T-P and P-T-P-T were observed. From simple kinematics, formulas for the ejected electron velocities have been derived. It was found, for example, that the maxima of the structures are independent of the observation angles for P^n-T^n sequences, and located at $2nv_p$, where v_p is the projectile velocity. Starting with projectile ionization, the T^n-P^n sequences give maxima at $(2n-1)v_p$. In contrast, for P^n-T^m or T^n-P^m sequences where $n \neq m$, the positions of the maxima vary with the observation angle. In our work, since the maxima are located at the same position, only the P^n-T^n sequences (and T^n-P^n sequences for He$^+$ impact) have to be considered. The He$^+$ and He^{2+} projectile velocities are 0.52 and 0.73 a.u., respectively. For He$^+$ impact, P-T, T-P, and P-T-P-T sequences are expected to give rise to electrons with velocities centered at 1.04, 1.56, and 2.08 a.u., respectively. In the case of a He^{2+} projectile, the expected maxima are located at 1.46 and 2.92 a.u. From our measurements and analysis (Figs. 2 and 3), it appears that, though the velocity separation of the maxima is close to the expected value for He$^+$ impact, the observed peak positions (at 0.75 and 1.2 a.u.) do not support the Fermi-shuttle ionization mechanism to explain the observed structures.

Finally, it is mentioned that transfer ionization (TI) might contribute to the measured ejected electron spectra. Various mechanisms can give rise to TI [16], including autoionization as already discussed above, but quantitative results on these different contributions are not available. Furthermore, the fact that the observed oscillatory behavior for He^+ and He^{2+} is essentially the same suggests a common origin for these oscillations not due to transfer ionization, which would likely give rise to different electron emission structures for these projectiles of different charge. In any event, experimental studies capable of isolating the TI contributions, combined with a detailed theoretical treatment of the ejected electron spectra, will likely be required in order to evaluate the possible effect of transfer ionization.

In summary, we observed well-defined oscillatory structures in single-ionization electron spectra following collisions between He^+ and He^{2+} projectiles and He target atoms at low impact velocities. The emitted electrons were detected at angles of 30° and 90°. The experimental cross sections were fitted using polynomial functions, which were used to simulate the decreasing noninterfering ionization cross sections. For both collision systems, oscillations were observed with maxima and frequencies independent of the observation angle. Since no available theoretical calculation predicts these small high-frequency oscillations, their origin remains open for discussion. It seems that neither autoionization processes following the production of highly excited states of the target, such $3lnl'$ $(n \geqslant 3)$, nor Fermi-shuttle ionization, are likely to explain the emergence of the observed structures. However, it is recalled that these oscillations have frequencies of the same order as those found for the system $H^+ + H_2$ [3]. In addition, the collision systems used here are similar to the system $H^+ + H$ explored theoretically [10], for which interferences in ionization from both projectile and target centers have been shown to play a role. Hence, a precise theoretical investigation is needed to complement the present study and to provide a satisfactory explanation for the experimental observations.

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