Fast negative helium ions produced by double electron capture in single He^+-He collisions

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Fast (0.35–1.2-MeV) He^- ions produced by double electron capture have been observed in He^+-He collisions. The thickness of the gas-jet target ($4\times10^{13}-10^{15}$ atoms/cm^2) has been calculated without any separate calibration. Single-collision conditions have allowed a direct measurement of the total cross section of the double electron capture. These data can be related to the development of theoretical studies on double electron capture.

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I. INTRODUCTION

In recent years, double electron capture (DEC) by a multiple charged ion colliding with a many-electron atom or molecule has been studied in several laboratories. The DEC of light ions is of great theoretical interest since it can establish or test scaling rules that enable us to predict the behavior of more complex collision partners. One of the fundamental reactions in this field is the He^+-He($1s^2$) collision leading to the production of He^- ions. The He^- ion has been the subject of a number of investigations dealing with either formation [1,2], destruction (electron detachment [3], photodetachment [4]), or lifetime and fine structure [5] of this ion, which exists in metastable, doubly excited states only. Measurements [6] on the decay of He^- ions have shown that all the fine-structure components are long-lived metastable ($\tau > 1$ μs). The lifetime studies have shown that two quite distinct lifetime components of 10 and 350 μs are present. These lifetimes have been associated with the $J=\frac{1}{2}$, $\frac{3}{2}$, and $J=\frac{5}{2}$ fine-structure components of the lowest doubly excited quartet state $(1s2s2p)\,^4P^o$, which is known to be bound and metastable against autoionization. More recently, it has been suggested [7] that the lifetime component of 10 μs should be associated with the lowest doubly excited doublet state $(1s2p^2)\,^2P^e$, which is metastable against autoionization. Up to now, the He^- ions have been mainly produced by collision involving one-electron capture by fast metastable helium atoms.

In this paper, we report on measurements of fast He^- ions produced by direct double electron capture in a single He^+-He collision via a gas-jet target.

II. EXPERIMENTAL PROCEDURE

A. Experimental setup

The experiment was performed at the 2.5-MV Van de Graaff accelerator of the Institut de Physique Nucléaire de Lyon with He^+ ion beams accelerated to energies ranging from 0.35 to 1.2 MeV. Figure 1 shows the experimental setup. After momentum analysis by a magnetic field, the incident beam was defined by two collimating apertures (D_1,D_2), which ensure an angular dispersion of ±0.5 mrad. Before reaching the gas jet, the beam was intercepted by an electrically isolated fast rotating chopper that measured the incident beam current (about 1 nA).

The gaseous target was a jet formed by expansion of helium in vacuum through a thin tube (length 350 mm, diameter 2 mm). The pressure in a gas reservoir at the inlet of the capillary was maintained constant by an electromagnetic valve associated to a pressure-flow controller. The input pressure P_0, varying from 0.7 to 5.5 Torr, was measured with a Baratron gauge giving a precision better than 2 mTorr. Very-high-purity helium gas was used; gas impurities (mainly H_2O, Ne, and N_2) represented less than 1 ppm, which leads to a negligible contribution to the He^- production. The gas jet was mounted on a micrometric goniometer that allows all possible translations (X,Y,Z) of the jet by steps of 50 μm. The capillary was set in order to get the maximum beam-jet overlap.

The low pressure in the collision chamber was ensured by a 2000 liter/s oil diffusion pump. During the experiments, the vacuum in the collision chamber varied from 2×10^{-6} to 7×10^{-5} Torr, depending on the gas-jet inlet pressure. A differential pumping system using two diffusion pumps (Fig. 1) on both sides of the collision chamber was able to maintain a residual gas pressure of 5×10^{-7} Torr in the region closed to the gas jet. Three other diffusion pumps (Fig. 1) placed along the beam line ensured high vacuum conditions with a mean residual pressure of 2×10^{-7} Torr. We have checked that the pressure in the detector chamber and along the beam line was not significantly affected by the admission of the target gas.

The emergent beam was magnetically analyzed in a detection chamber. The negative and positive ions were

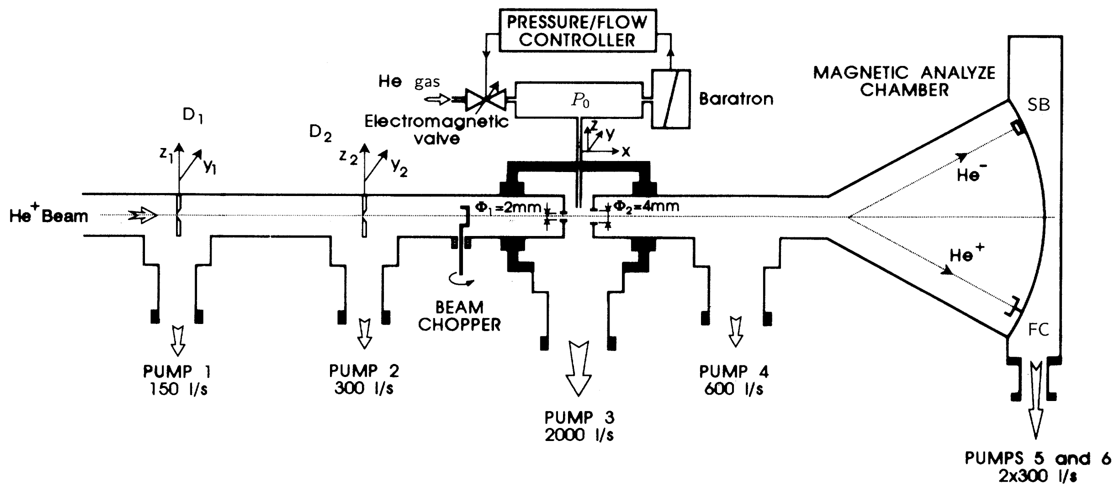

FIG. 1. Experimental setup (see text for details). SB denotes a surface-barrier detector and FC a Faraday cup.

simultaneously detected by two movable detectors, a surface-barrier detector (SB) and a Faraday cup (FC), respectively. The Faraday cup was negatively biased at the entrance aperture to present the escape of secondary electrons. Care was taken to ensure complete collection of particles into their respective detector. For all the energies studied, the time of flight from the gas target to the detection system (0.25 μs for the lowest energy) is small compared to the lifetime of the two main metastable states of the He^- ion (10 and 350 μs).

B. Absolute determination of the cross sections

To deduce absolute cross sections from the data, it is necessary to extract the target thickness. The thickness of the gas-jet target depends on two parameters, the inlet pressure P_0 which was measured with a precision better than 0.3%, and the position of the jet capillary with respect to the incident ion beam, which was measured with a precision better than 4% due to the micrometric goniometer. For the directivity of a beam formed by a gas flow in a capillary, one distinguishes several flow regimes according to the ratio value d/L_{eff}, where d is the diameter of the capillary and $L_{\text{eff}}=(2L\lambda_0)^{1/2}$, λ_0 being the mean free path of atoms and L the length of the capillary. In our experiment,

$$d/L_{\text{eff}} \geq 1 \ . \tag{1}$$

In these conditions, we obtain a viscous flow quantitatively well described by the kinetic theory. The target density can be calculated without requiring a separate calibration from a known cross section. The jet profile has been determined by using the theoretical expressions given by Troitskii [8]. Preliminary experiments [9] were performed to test these calculations, in particular by scanning the jet with a thin capillary connected to a mass spectrometer. The angular distributions of the particle flow were measured from 0.7 to 5.5 Torr input pressure (P_0). The normalized angular distributions were verified to keep the same shape in good agreement with the law predicted by Troitskii [8].

$$\left[\frac{dN}{d\Omega\, dt}\right]_\Theta = \left[\frac{dN}{d\Omega\, dt}\right]_{\Theta=0} \cos^{5/2}\theta \ . \tag{2}$$

On the other hand, we have found that the total particle flow through the jet capillary followed a P_0^2 dependence in accordance with the Poiseuille formula. The target thickness was given by the integral of the profile performed on the ion-beam–gas-target overlap.

The dominant errors in the cross-section measurements are systematic errors. Main contributions arise from the uncertainty in the target thickness ($\pm 8\%$). Small additional statistical errors come from the measurement of the charge fractions. The current from the beam chopper and counts from the surface barrier are integrated and subject to errors of $\pm 2\%$ arising directly from beam intensity fluctuations.

III. RESULTS

Usually, in gas-target–ion-beam collisions, the cross sections are obtained by the well-known growth rate method. Individual cross sections can be obtained if a beam consisting of only one charge state i is sent through a collisional chamber. The charge states j in the emergent beam are then observed for various target thicknesses. Let x represents the target thickness (number of atoms per cm^2) and $F_{ij}(x)$ the fraction of ions emerging in the charge state j (i.e., the number of emergent ions in the charge state j divided by the number of incident ions). For small target thicknesses,

$$[F_{ij}(x)-F_{ij}(0)]/x=\sigma_{ij}+Bx+\cdots \ , \tag{3}$$

where $F_{ij}(0)$ is the fraction of the charge state j measured without target. In the single-collision condition related to small target thicknesses, the fraction $F_{ij}(x)$ is linear with respect to x, the slope being equal to the cross section σ_{ij} (in cm^2). The parameter B in Eq. (3) describes the contribution of double collisions.

In order to test the apparatus, we sent H^+ ions through the He gas jet and measured the fraction of emergent H^- ions with respect to the target thickness. Figure 2 represents the typical curve obtained with 0.3-MeV H^+ colliding with helium gas. Figure 2 shows also the ratio $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ with respect to the target thickness x. We observe that this ratio is constant below the thickness 4×10^{14} atoms/cm^2, which means that for these thicknesses ($<4\times10^{14}$ atoms/cm^2) the double collision is negligible. This ratio corresponds to the cross section $\sigma_{1\text{-}1}$, which is found to be equal to $(5.9\pm0.6)\times10^{-23}$ cm,^{2} a value in agreement with previous results [10(a)] [$(7.0\pm1.1)\times10^{-23}$ cm^2]. One notes that the production of H^- by direct double electron capture is weakly disturbed by the double-collision processes; for a thickness equal to 10^{15} atoms/cm^2, $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ is 15% higher than $\sigma_{1\text{-}1}$. Assuming a three-component system [11,12] with charge $-1,0,1$, and a purely positive charged incident beam, the growth of the negative beam with respect to the target thickness can be easily calculated using the corresponding six charge-exchange cross sections. Taking from the literature the following cross sections σ_{10} (6.5×10^{-19} cm^2) (Ref. [11]), $\sigma_{0\text{-}1}$ (7.8×10^{-20} cm^2) (Ref. [10(a)]), σ_{01} (5.4×10^{-17} cm^2) (Ref. [10(a)]), σ_{-11} (3×10^{-18} cm^2) (Ref. [12]), σ_{-10} (1.10^{-16} cm^2) (Ref. [3]), and our result for $\sigma_{1\text{-}1}$ (5.9×10^{-23} cm^2), we obtained an excellent agreement with our data (see the calculated curve on Fig. 2). We point out that other tests of our system have been done with several processes to compare our results with previous results. As an example, we have studied the transmission of fast $H_3{}^+$ (0.47 MeV) through the helium gas jet. The dissociation cross section measured [$(1.4\pm0.14)\times10^{-16}$ cm^2] is in very good agreement with the value obtained by Jalbert, Coelho, and de Castro Faria [13] [$(1.37\pm0.21)\times10^{-16}$ cm^2)]. We have also measured the cross section for single electron loss for 0.35-MeV He^+ colliding with He. The obtained value [$(2.2\pm0.2)\times10^{-16}$ cm^2] is in good agreement with the one obtained by Rudd *et al.* [14] [$(2.\pm0.2)\times10^{-16}$ cm^2].

After these conclusive tests on the apparatus, we have studied the He^- ion production in the collision He^+-He in the 0.35–1.2-MeV energy range. Figure 3 shows the typical curve obtained for the fraction of He^- per He^+ incident ion at the energy of 0.35 MeV with respect to the target thickness. We report also the ratio $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ with respect to the target thickness. We observe that this ratio is constant for target thicknesses below 10^{14} atoms/cm^2. Spurious He^0 atoms can be produced by one-electron capture of the incident He^+ ions from multielectron atoms of the residual gas between the first analyzing magnet and the gas target (2 m), and then can capture an additional electron in the He target. This double-collision process contributes to the He^- production with a linear dependence on the target thickness. From the cross sections of the involved processes, one can estimate this contribution to the He^- production by calculating the ratio

$$I^-_{0\text{-}1}/I^-=(\sigma_{10}\Lambda\sigma_{0\text{-}1}x)/[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]$$
$$=(\sigma_{10}\sigma_{0\text{-}1}\Lambda)/\{[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x\}$$

for target thicknesses (x) below 10^{14} atoms/cm^2. $I^-_{0\text{-}1}$ denotes the intensity of the negative-ion beam generated from the spurious He^0 by one-electron capture ($\sigma_{0\text{-}1}$) and I^- the total intensity of the negative-ion beam produced in the He target. Λ represents the thickness of the residual gas, about 2×10^{12} atoms/cm^2 [$(2-5)\times10^{-7}$ Torr)]. To estimate the ratio $I^-_{0\text{-}1}/I^-$, we have taken from literature the cross sections of the involved processes as follows:

(i) for the lowest energy (0.35 MeV)—$\sigma_{10}=7\times10^{-17}$ cm^2, Ref. [10(b)] (He-air) and $\sigma_{0\text{-}1}\approx\sigma^*_{0\text{-}1}$ (mainly the metastable He^{0*} participate [1(b)]) with $\sigma^*_{0\text{-}1}\approx2\times10^{-19}$ cm^2. $\sigma^*_{0\text{-}1}$ has been extrapolated by a velocity dependence in v^{-7} from the data of Ref. [1(a)] in agreement with the typical velocity dependence for the one-electron-capture cross section in the energy range studied here and for

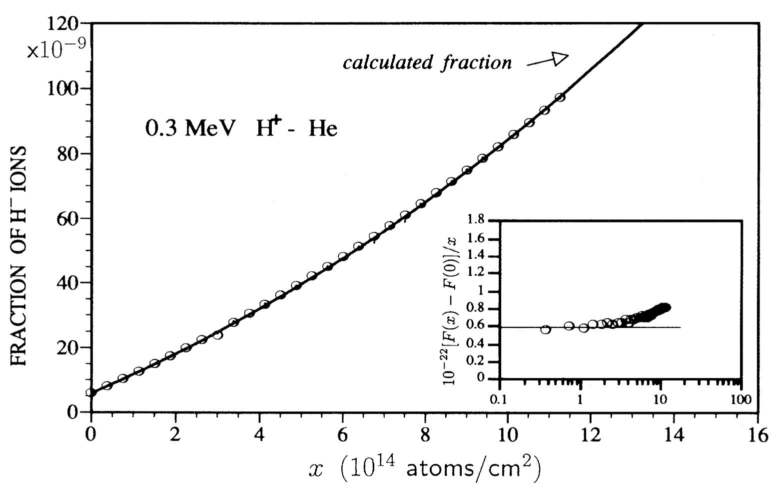

FIG. 2. Fraction of H^- ions as a function of x, the target thickness, for 0.3-MeV H^+ colliding with He. Inset: The ratio $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ as a function of x (logarithmic scale). The line corresponds to the cross section $\sigma_{1\text{-}1}$ (cm^2).

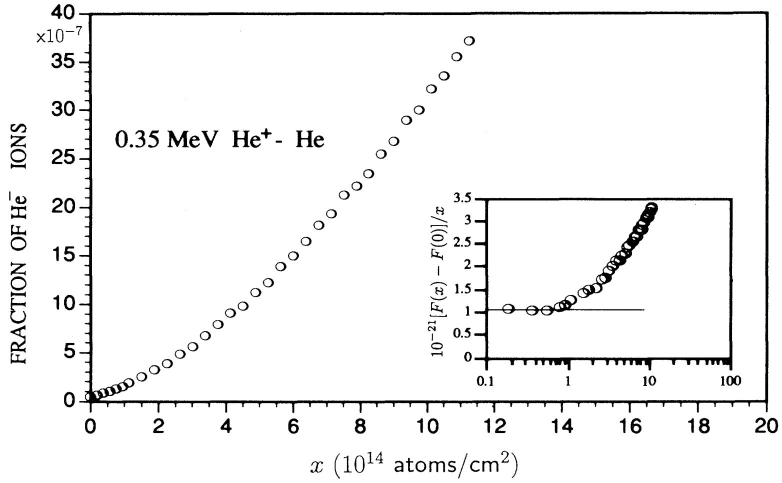

FIG. 3. Fraction of He^- ions as a function of x, the target thickness, for 0.35-MeV He^+ colliding with He. Inset: The ratio $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ as a function of x (logarithmic scale). The line corresponds to the cross section $\sigma_{1\text{-}1}$ (cm^2).

light projectiles (see, for example, Ref. [12] and references therein).

(ii) for a highest energy (0.9 MeV)—$\sigma_{10} \approx 6\times10^{-18}$ cm^2, Ref. [15] ($He\text{-}O_2$) and $\sigma_{0\text{-}1} \approx \sigma^*_{0\text{-}1}$ with $\sigma^*_{0\text{-}1} \approx 10^{-20}$ cm^2. $\sigma^*_{0\text{-}1}$ has been extrapolated as above.

That leads to a He^- production due to spurious He^0 smaller than 3% of the total He^- production in the energy range studied here. This value is probably overestimated since all the spurious He^0 ions produced in the residual gas are not in metastable states. So, in Fig. 3, the region where the ratio $[F_{1\text{-}1}(x)-F_{1\text{-}1}(0)]/x$ is constant, corresponding to target thicknesses below 10^{14} atoms/cm^2, is a single-collision region where He^- ions are produced by double electron capture. The importance of the He^- production by double collision with respect to the one by single collision, for target thickness greater than 10^{14} atoms/cm^2, explains the fact that the DEC process with He^+ incident ions could not be easily directly observed. Compared to the H^- production in the same thickness range, the He^- produced by the DEC are strongly disturbed by double-collision processes. In the He^- case, the ratio $[F_{1\text{-}1)}(x)-F_{1\text{-}1}(0)]/x$ for x equal to 10^{15} atoms/cm^2 is 3.5 greater than the cross section $\sigma_{1\text{-}1}$.

The DEC cross sections measured in the present work are given in Table I. They are small but only about a factor of 20 smaller than the cross section of the DEC by H^+ ions colliding with He at the same velocity. In the limited energy range investigated, the DEC cross section has a velocity (v) dependence in v^{-10}. When comparing the two-electron capture cross sections σ_{20} (taken from the literature [16]) and $\sigma_{1\text{-}1}$, we observe that, as expected, σ_{20} is much greater than $\sigma_{1\text{-}1}$ (about 10^4 at a given velocity). Moreover, in the same energy range, the DEC cross sections for the He^{2+}-He process have a velocity dependence in v^{-11} (Ref. [16]).

TABLE I. Double electron capture cross sections $\sigma_{1\text{-}1}$ for various energies.

Energy (MeV)	$\sigma_{1\text{-}1}$ (cm^2)
0.350	$1.0\pm0.1\times10^{-21}$
0.400	$6.8\pm0.7\times10^{-22}$
0.625	$7.0\pm0.7\times10^{-23}$
0.900	$9.5\pm0.9\times10^{-24}$
1.200	$2.8\pm0.3\times10^{-24}$

IV. CONCLUSION

Despite the great number of experimental results, the theoretical studies of the double electron transfer process using different quantum or semiclassical approximations are limited due to the complexity involved. The He^{2+}-He collision leading to He production is the most studied system. This work represents measurements of double-electron capture cross sections in He^+-He collisions. Most of the theoretical calculations treat the process of double electron capture in the independent-particle approximation [17–20]. However, the effect of the electronic correlation is still discussed. The results presented in this work could be very useful to test these methods since, among the negative ions resulting from a double electron capture, the He^- ion is the simplest open-shell system. Nevertheless, we point out that no calculation is yet available in the literature concerning the total cross sections of the double electron capture by He^+ in helium at intermediate velocities.

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