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Double capture with simultaneous ionization in He²⁺ on Ar collisions

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We have investigated double electron capture with simultaneous target ionization in collisions of 25-keV/amu $^3\text{He}^{2+}$ projectiles on argon. Doubly differential cross sections for electrons emitted in angles 0° , 20° , 45° , 90° , 128° , 175° , in coincidence with the neutralized projectiles were obtained. To establish the relative importance of the double capture channel in the electron emission at the present intermediate impact velocity, differential cross sections for total electron emission were also measured. A narrow symmetric cusp-shaped structure for electrons with velocity close to the ion velocity, associated with the neutral emerging projectile, is observed. Binary encounter electrons, masked by other processes in total emission, are clearly observed for the double capture plus ionization channel.

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I. INTRODUCTION

In the last decade, interest on impact ionization processes that involve more than one active electron has increased. The simplest system to study these phenomena is the collision of bare ions with a He target, which has been extensively investigated to understand the role of electron correlation in electron emission processes [1]. Different approximations have been proposed in order to model these many-particle mechanisms. An independent particle model, for instance, assumes that each electron interacts with the projectile independently of the other electrons [2,3]. However, in order to remove some discrepancies with experimental results, models including electron correlation in the initial and final states [4], and during the collision [5], have been proposed.

Comparison between experimental data and theoretical models showed that the relative importance of electron-electron interaction compared with the projectile-electron interaction depends strongly on the incident energy, which determines the characteristic time during which each interaction is effective [4]. In this sense, it seems to be necessary to investigate multiple electron processes, distinguishing each different final state of the collision system. This kind of information is blurred in total cross section, thus differential cross sections become essential.

At low and intermediate impact energies, electron emission with simultaneous capture of target electrons becomes dominant in comparison with direct multiple ionization channels [6]. Therefore, channels involving electron capture enhance the production of highly charged recoil ions. In an

experiment related to the present one, Moretto-Capelle *et al.* [7] studied He²⁺ on Ar collisions by measuring the emitted electrons at 35° in coincidence with recoil ions. For their low energy (12.5 keV/amu), potential energy curves of the quasimolecular states give clues for evaluating the relative importance of the different collision channels involving ionization. They concluded that the main contribution to ionization when multicharged recoil ions are produced is associated with capture of target electrons by the projectile, in agreement with previous measurements performed with a different experimental setup [8]. Even though the experiment of Moretto-Capelle *et al.* can be used as a reference for our present work, we have to consider that already, differences in the results for ionization have been found between 12.5 and 25 keV/amu.

Here we report measurements of doubly differential cross sections of ionization, as a function of angle and energy of the emitted electron, simultaneous with double electron capture by the projectile (DCI). The system under study was 25 keV/amu³ He²⁺ impinging on Ar atoms. The doubly differential cross section for total (noncoincident) electron emission (TEE) is also measured. Available data for the DCI process mainly consisted of total cross sections [9], and only recently, new techniques allowed us to get additional information about the momentum [10] and the charge state of the outgoing particles [8].

The present measurements for DCI confronted us with some difficulties due to possible artifact effects of contamination from other collisional processes, either produced by undesirable components of the primary beam or by double collisions with the atomic target. Care was taken to assess these contributions, and to perform the measurements accordingly. Details of the technique are explained in the next section.

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TABLE I. Main processes that contribute to our measurements. (1) is the process of interest. (2) and (3) are contributions of double collisions. (4) and (5), collisions due to contamination of the beam. All cross-section data are from [9,15], except $\sigma_I(He^0)$, from [16].

	Beam	Collision processes		Cross section ($\times 10^{-16}$ cm ²)	
		First collision	Second collision	First collision	Second collision
1 2 3	³ He ²⁺	$He^{0} + Ar^{3+} + e$ $He^{+} + Ar^{2+} + e$ $He^{2+} + Ar^{+} + e$	$He^{0} + Ar^{+}$ $He^{0} + Ar^{2+}$	$\sigma_{DCI}(\text{He}^{2+}) = 1.04$ $\sigma_{TI}(\text{He}^{2+}) = 2.15$ $\sigma_{I}(\text{He}^{2+}) = 1.9$	$\sigma_{SC}(\text{He}^+) = 6.02$ $\sigma_{DC}(\text{He}^{2+}) = 2.37$
4	³ He ⁺	$He^0 + Ar^+ + e$		$\sigma_{TI}(\mathrm{He}^+) = 2.21$	
5	$^{3}\mathrm{He^{0}}$	$He^0 + Ar^+ + e$		$\sigma_I(\mathrm{He}^0) = 13.6$	

II. EXPERIMENTAL PROCEDURE

The experimental setup is described in detail elsewhere [11,12]. In the experiment reported here, a ${}^{3}\text{He}^{2+}$ beam of 25 keV/amu interacted with an effusive Ar gas target provided by a hypodermic needle of 0.25 mm diameter. The point of interaction is also the object focus of a cylindrical mirror analyzer that selects the energy of the electrons produced in the collision process. The angle of emission of the electrons is chosen by rotation of the analyzer. The half-angle of acceptance cone was 2° and the resolution in energy was 6%. The beam was collimated to 0.6×0.6 mm² by means of two sets of four-jaws slits located at 0.5 m and 1.2 m before the target.

Projectiles leaving the collision chamber were charge-state selected by an electrostatic field provided by parallel plates, and then detected through two secondary-electron converters equipped with high-count rate channeltrons. The pressure in the transport line was 2×10^{-7} Torr, and it did not change when the target gas was used, due to the location of high impedance apertures at the entrance and exit of the collision chamber. The base pressure in the collision chamber was 4×10^{-8} Torr. Spurious magnetic fields were reduced to less than 5 mG in the collision region by means of three pairs of Helmholtz coils.

Standard coincidence techniques were used to measure electrons in coincidence with emerging neutralized He projectiles. The total noncoincident number of electrons (start pulses) and projectiles (stop pulses) were also recorded and used for normalization.

Doubly differential cross sections of total electron emission were obtained by a standard electron spectroscopy technique, in which the electron counts for a given angle and energy were normalized to a selected beam charge, collected in a Faraday cup.

In our experiment we detected electrons in coincidence with neutralized He projectiles in order to measure ionization simultaneously with double capture process. However, there are other nondesirable collision processes that contribute with the same final products to the measured coincidence spectra. They are associated with double collision processes and collisions of projectiles with other charge states that contaminate the primary He²⁺ beam. We estimated the effect of these contaminations using reported values of total cross sec-

tions for the different reactions (see, for example, [13]).

Consider, in the first place, double collision processes in which an electron is emitted by the target and a He²⁺ projectile is neutralized in its path up to the projectile detector. Such a double-collision contamination could be important due to the large cross section involved in comparison with the DCI one. In case of a dilute target, the probability of double collisions is a quadratic function of the target thickness, while single events depend linearly. Then, by decreasing the target thickness, this contamination can be reduced to a negligible amount. We found that the importance of such undesirable processes is strongly dependent on the energy and the angle of emission of the electrons. Thus, in our experiment we chose the working pressure of 1×10^{-6} Torr in the collision chamber, which limited the contamination to less than 10% for electron energies up to 10 eV. For energies greater than 25 eV, we could increase the pressure to 5 $\times 10^{-6}$ Torr, keeping the contamination in less than 5%. These values were estimated with the help of measured coincidence rates as a function of chamber pressure [13,14] and are well within the expected values calculated by using total cross sections for the processes listed in Table I. Next we consider the effect due to other charge states of the projectiles arriving at the target. Taking into account the values of total cross section for charge exchange, we estimated the fractions of single charged and neutral He projectiles in the main He^{2+} beam to be $f(He^{+}) = 2\%$ and $f(He^{0}) = 0.5\%$, respectively. Since these values are small, only singlecollision processes are important in order to evaluate the contribution to the coincidence counting. We found, using total cross section data, that the more important contributions of undesirable coincidences are transfer ionization by He⁺, which amounts to 2.3%, and single ionization by He⁰, which is about 5.4% (see Table I).

Finally, we have to recall that, even though we have taken into account only single-electron emission processes in the previous discussion, our present measurements include multiple ionization with simultaneous double capture. However, if we consider that double-capture processes with simultaneous ionization of two or more electrons have a reported cross section $\sigma = 0.151 \times 10^{-16}$ cm² [9], and that for DCI $\sigma_{\rm DCI} = 1.04 \times 10^{-16}$ cm² [9], then multiple ionization

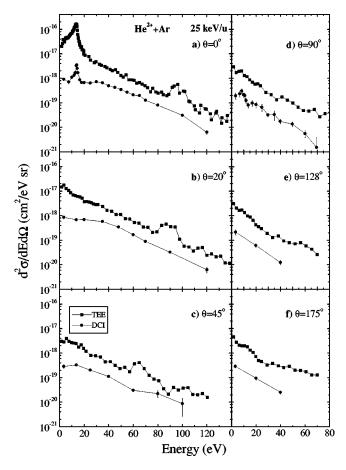


FIG. 1. Doubly differential cross sections: Squares, total electron emission (TEE); circles, ionization with simultaneous double capture (DCI), for 25-keV/amu 3 He²⁺ + Ar, with electron emission angles of (a) θ =0°, (b) 20°, (c) 45°, (d) θ =90°, (e) 128°, and (f) 175°. Lines are to guide the eyes.

processes are expected to contribute with not more than 15% to the present measurements.

III. RESULTS AND DISCUSSION

The experimental results for the doubly differential cross sections of TEE and DCI are shown in Fig. 1. Absolute cross-section values for the electron emission were obtained by integrating the measured differential cross section and normalizing to the total cross section reported by DuBois [9]. In the case of the coincidence data, absolute values for the DCI process were obtained by normalizing the total electron counts (start pulses) in the coincidence measurement with the total electron emission differential cross section, for each angle and energy of emission. This normalization procedure includes a correction factor due to the efficiency of the coincidences system that was estimated as 88% [12].

In the forward direction spectra, at zero degree, the total emission as well as the coincidence spectrum of DCI show, as the main structure, a peak centered at the electron equivalent energy $T=v^2/2=13.6$ eV, where v=1 a.u. is the projectile velocity. Collision channels with all charge states of the emerging projectile contribute to the cusp observed for the total emission. For He²⁺ outgoing projectiles, electrons

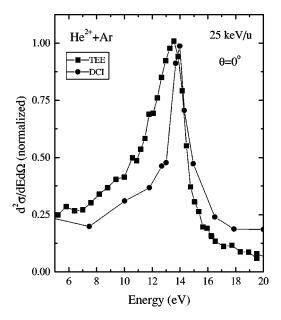


FIG. 2. Cusp emission spectra in the forward direction normalized to the maximum. Squares, total electron emission (TEE); circles, ionization with simultaneous double capture (DCI).

come from pure ionization processes, which are mainly single ionization. On the other hand, when the final projectile is He⁺, at least a two-electron transfer ionization process (TI) is required. In both cases, the appearance of a cusp in the electronic distribution has been interpreted as due to the long-range final-state interaction between the electron and the charged projectile (He²⁺ and He⁺, respectively) [17–19]. However, the shape of this cusp might be dependent on the particular formation mechanism of each outgoing state. In the case of TI, some experimental evidence of an independent mechanism of bound and continuum two-electron capture was obtained [20], but the effect of electron correlation cannot be disregarded.

A different situation is found for the cusp emission in the case of the DCI process, which results in a neutral outgoing He projectile. Taking into account the lack of long-range interaction between the electron and the neutral atom, it is very noticeable that a cusp structure is present in this collision channel. A mechanism to produce this final state in DCI has not been identified yet. In a related collision system, with neutral He as projectile, a cusp in the electron emission has also been found [21,14]. In this case, an interpretation was given that attributes the cusp to a resonance process associated to a metastable state of the He atom emerging from the collision [19].

In Fig. 2, we show the cusp corresponding to the total emission and to DCI, normalized at the maximum. There is an asymmetry toward low electron energies in the case of total emission. This asymmetry is attributed to the simultaneous interaction of the cusp electron with both projectile and residual target [22,23]. On the other hand, a narrow, more symmetric cusp is observed for DCI process, a fact that also is in accordance with the results obtained for the capture to the continuum by neutral He projectiles [21], and for TI with He⁺ as projectile [24]. Following the resonance phe-

nomenon description [19], a peak narrower than that resulting from a pure Coulomb interaction in the final state is predicted for the later case. This kind of cusp, but more symmetric, is observed in different systems when the projectile or the target is neutral after the collision [12,13,21,24]. A small shift in the position of the cusp maxima is observed in Fig. 2. In principle, a divergence centered at the projectile velocity is expected. However, as it has been previously noted [25,26], the measured cusp with a finite energy resolution of the electron spectrometer presents a shift of the maximum in the case of an asymmetric cusp, as seen in TEE in Fig. 2.

Another distinctive feature observed in the DCI spectra, when compared to that of the total emission, is an enhanced contribution due to the collision mechanism known as binary encounter (BE). In this process, the projectile interacts directly with a target electron in a close collision, with the residual recoiling target as spectator. The predicted electron velocity distribution corresponds to a sphere given by $|\mathbf{v}_{e}|$ $-\mathbf{v} = v\sqrt{1+\varepsilon_i/T}$, where T is the equivalent energy defined previously, \mathbf{v}_e is the electron velocity, and ε_i is the binding energy of the electron in the initial state. Its shape should reflect the initial momentum distribution of the electron. Conservation of energy and momentum show that BE can occur only if the velocity of the projectile is greater than $\sqrt{2|\varepsilon_i|}$ and, for low impact energies, the momentum of the recoil ion has to be considered. The measurements of electron emission without coincidence techniques (TEE) do not show any distinguishable structure related to this simple process for low projectile impact energy. On the other hand, at emission angles $\theta = 0^{\circ}$ and 20° and electron energy about 30 eV, the broad structure seen in the DCI spectra may be attributed to the BE collision. This interpretation is consistent with the picture of double capture as a collisional process involving a small impact parameter, where a strong interaction between the projectile and the target electrons is ex-

In the total emission differential cross sections shown in Figs. 1(a–c), a structure, dependent on the angle of emission, is also present. At 0° electron emission angle the structure peaks at around 100 eV, shifting to lower energies for higher observation angles. At 45° it is found around 65 eV in the spectrum. This structure corresponds to autoionization processes of the He projectile [27], affected in energy and angle by the Doppler shift. As the projectile finally becomes singly charged after autoionization, the structure is only observed in the total emission where the autoionization processes contribute to the TI collision channel.

At 90° , we measured a more detailed spectrum, looking for an indication of a possible contribution of a Thomas-like mechanism to the DCI process [28], which was found at high impact energies [29,30]. We could not detect any evidence of such a mechanism for DCI processes at the projectile energy of this work. Only a structure at low electron energies is observed, in a region where Ar and He autoionization peaks are expected [27].

Doubly differential measurements shown in Fig. 1 give information about the contribution of the DCI process to the electron production as a function of the angle and energy of

the emitted electrons. At the lower electron energy measured for DCI, at 3 eV, we obtained a contribution of only 2% to the total electron emission at $\theta = 0^{\circ}$, that rises to 5% at 20° . For higher angles it is about a 9% and almost independent of the angle. For $\theta = 0^{\circ}$, 20° , and 45° , the contribution of DCI increases with the electron energy up to about 20%, in accordance with the above-mentioned relative enhancement of binary encounter emission associated with double capture. The small contribution of DCI at low energies could be understood based on the argument of the small impact parameter collisions required for this process. Low-energy electron emission is usually associated with soft collisions where large impact parameters prevail. At $\theta = 90^{\circ}$ an almost constant contribution of about 12% is obtained in the covered energy range, while at 128° and 175°, with only three measured energies, it is about 10%.

IV. CONCLUSIONS

By means of electron-projectile coincidence techniques, doubly differential cross sections of ionization simultaneously with double capture have been measured for 25-keV He²⁺ on Ar. These data allow a more detailed study of processes with three or more active electrons. In particular, as it was already observed for the same system [7,8], those collision processes involving capture of target electrons are relevant in the enhanced production of highly charged recoil ions at low and intermediate impact energies.

A cusp in the emission of electrons with velocity close to that of the projectile is observed associated with neutralized outgoing He. This cusp is narrower than the one observed in the total emission, which is dominated by capture to the continuum by He²⁺ and transfer ionization, that is, processes with charged emerging projectiles. A similar feature observed in the case of capture to the continuum of neutral He projectiles [21] gives a stronger divergence than that due to a pure Coulomb interaction between the electron and the projectile [19]. With a similar interpretation, cusp electrons in DCI may be associated with a capture processes resulting in formation of metastable state of the emerging He atom.

As the differential emission for total electron production was also measured, we could discuss the relative contribution of the DCI process to the electron emission. Two features were observed in DCI, a relative enhanced contribution of electrons from binary encounter collisions and a low contribution at small electron energies. Both could be related to the importance of small impact parameter collisions for DCI, compared to the other processes producing electron emission.

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