Coincidence electron spectroscopy of electron-impact multiple ionization of argon

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Measurements of the partial double differential ionization cross sections of argon atoms have been made by performing coincidences between recoil ions and energy- and angle-selected slow electrons produced in 12–24 keV e^--Ar collisions. Results show that the Ar^{3+} ions are formed mainly by a two-step-one process via electron shake-off. These measurements have enabled determination of the shake-off probability $S = 0.14 \pm 0.01$ for $L_{2,3}$ subshell, which is in good agreement with a theoretical predication. Additionally, Ar^{2+} and Ar^+ ions are found to arise respectively from the filling of *L*-shell vacancy by an auger transition and from the direct ionization of *M*-shell of the argon atom.

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I. INTRODUCTION

A detailed understanding of the dynamics of multiple ionization processes of the atomic systems under impact of energetic charged particles is one of the central goals in atomic collision physics. Such an approach of studying ionizing collisions of atoms has proved to be useful in studying the properties of atoms [1]. However, the most basic few-body break-up processes, such as electron-impact single ionization of hydrogen and double ionization of helium have been studied recently [2–4], whereas the transition to true many-body systems still remains a challenge. Theoretically, the case of a single ionization of atomic targets by impact of high velocity electrons can be explained satisfactorily by employing the Born approximation calculations; however, the case of a multiple ionization becomes a complex process due to involvement of many-electron problems [5]. On the experimental side, tests of these processes need detailed descriptions of the outgoing continuum and of the characteristic electrons and those of the produced ions from collision.

The collision of energetic electrons with a many-electron atom, such as argon, may create an inner-shell vacancy. Such a vacancy is subsequently filled by a cascade removing one or more electrons. This situation leads to the multiple ionization of the target involving a complex process. Investigation of such processes is the subject of our present interest. When a charged particle of intermediate to high velocity collides with a many-electron atom, the valid perturbation approximations categorize different kinds of multiple electronic transitions. Firstly, at a very high impact velocity, the multiple electronic transitions are caused by a two-step-one (TS1) process wherein a single first-order perturbation involving an active electron-projectile interaction takes place. This process further combines with one or more second-order electron-electron interactions called a shake-off process [6]. Secondly, at the intermediate velocities, the electron transitions are caused by independent sequential interactions between the incident charged particle and the promoted electrons; the involved interactions are assumed to be independent first-order processes. Such a mechanism producing the multiple ionizations is called a two-step-two (TS2) process. In case of a multiple ionization of an atom, the emitted electrons are considered to be the fingerprints of the transitions through which the final charge states of the target atom are produced and can be separated by measuring the coincidences between the produced ions and the electrons ejected from different shells or subshells of the atom during collision. The study of double ionization of helium by charged particle impact has received a considerable interest in the recent past and the mechanisms governing its double ionization are known more accurately [7]. However, in the case of a many-electron atom, such as argon, the multiple ionization mechanism becomes much more complex owing to the initial vacancy relaxing to a final multiple charged state of the atom.

Theoretically, a complete picture of shake-off and two-step processes involved in production of multiple ionization of atoms by the charged particle impact has been developed by McGuire [5]. The ionization of argon has mainly concentrated on the calculations of total ionization cross sections [8,9]. The experimental differential study of multiple ionization of argon by electron impact was made by Santos *et al.* [10] using coincidences between the scattered electrons with a known energy-loss and the produced ions. In general, a detailed mechanism for producing the multiple ionization of the target cannot be understood through such experiments. The first partial double-differential cross sections (PDDCS) of ionization of argon by electrons at impact velocity $v = 4.66–24.07$ a.u. were measured by Hippler *et al.* [11] using a slow/fast coincidence technique between the ejected electrons of known energies and the produced ions. However, the results obtained in their experiment were found to be inconsistent with the available theoretical predictions. The origin of the inconsistency is suspected to lie in the full extraction of the produced ions from the collision zone.

In this paper, we report on measurements of the PDDCS of argon ions produced under impact of 12–24 keV electrons with the argon atoms in order to provide a detailed insight into the dynamics of multiple ionization of the target. The present velocity range of the incident electrons ($v = 29.47–41.68$ a.u.) was chosen to encompass the region

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where the multiple ionization dominantly takes place by a shake-off process [12]. Making use of the present coincidence technique, our data corroborate that the double ionization of argon takes place dominantly through the *L*-shell vacancy creation in the target, which is also suggested by other workers, for example, Carlson *et al.* [13] and DuBois and Manson [14]. We show further, from the velocity dependence of PDDCS, that the shake-off process is mainly responsible for formation of a triply ionized argon atom under the present collision conditions and it has enabled us to determine the value of shake-off probability *S*.

II. EXPERIMENTAL

The present investigations were carried out on a crossed-beam type experimental facility developed for studying the slow electron-recoil ion coincidences [15,16]. The electrons ejected from the interaction region were energy analyzed by a 45° parallel plate electrostatic analyzer [full width at half maximum (FWHM)=12%] with a narrow solid angle (0.01 sr) and were detected by a channel electron multiplier (CEM) operating in a pulse counting mode. As the detection efficiency of CEM for low energy electrons is relatively low, the electric potential at the CEM mouth was maintained at 250 V for obtaining their maximum detection efficiency. The reduction of Earth's magnetic field near the collision zone and inside the analyzer was necessary in order to minimize the deflection of low energy electrons reaching the detector. This was accomplished by using a 0.5 mm thick antimagnetic μ-metallic shielding inside the wall of the reaction chamber and outer walls of the analyzer. The multiply charged ions produced in the interaction region were extracted by an electric field (180 V/cm) in the vertical direction and were detected by a channeltron; the applied field was perpendicular to both the direction of ejected electrons entering the electrostatic analyzer and to the incident electron beam direction. The extraction field was optimized for a full extraction of Ar^+ ions (slowest ions) and for their full transmission in the time-of-flight (TOF) spectrometer. The details of optimization procedures for full extraction and transmission probabilities of collisionally induced multiply charged ions are given elsewhere [15–17]. The ejected electrons viewed by the electrostatic analyzer were not affected by the presence of the extraction field. This was confirmed by observation that no noticeable change took place in the measured energy distributions of the ejected electrons "with" and "without" applied extraction fields.

III. RESULTS AND DISCUSSION

The partial double-differential cross sections of argon ions were measured by detecting coincidences between produced recoil ions and electrons of different energies ejected at angle θ_δ=90° to the incident beam direction. This angle of ejection was chosen due to a well-defined interaction zone which is viewed by the analyzer compared to the case at forward or backward ejection angles. A typical TOF spectrum of different argon ions in coincidence with the ejected electrons of energy E_δ=190 eV is shown in Fig. 1 for 12 keV incident

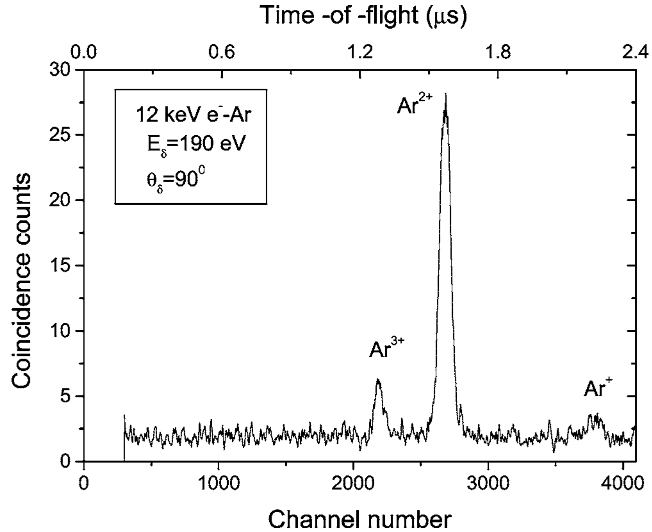

FIG. 1. Time of flight spectrum of argon ions produced from coincidences between the secondary electrons of 190 eV ejected at 90° and the recoil ions in collisions of 12 keV electrons with argon atoms.

electrons on argon in a single collision condition. A typical acquisition time for obtaining such spectra with a less than 2% counting statistics for Ar^{2+} ion was about 36 h. Figure 1 shows the doubly charged argon ions to be the most abundant while the singly charged ions are much less in intensity compared to both Ar^{2+} and Ar^{3+}. This observation clearly suggests that the Ar^+ ions are produced due to directly ejected electrons of energy 190 eV and that they are produced with a very small probability [18]; however, the *L*-shell vacancy production with subsequent emission of *L-MM* auger electrons seems to contribute maximum in producing the multiple ionization of argon atoms in the considered collisions.

For further investigation, we have analyzed the data collected for the relative *n*-fold double-differential cross sections, that is, DDCS $(n+)$ of argon ions from the TOF spectra. These spectra were obtained by performing coincidences between the recoil ions and the secondary electrons of kinetic energies ranging from 70 eV to 270 eV ejected at 90° with respect to the incident beam direction. The relative DDCS $(n+)$ were obtained by normalizing the true total coincidence counts of the *n*-fold ionized argon atoms with the measured total number of incident electrons and with the detection efficiency of CEM for *n*-fold charge state of the ions. The relative detection efficiencies for Ar^+, Ar^{2+}, and Ar^{3+} are estimated to be 67%, 92%, and 100% respectively. Figure 2 shows the variation of the relative partial DDCS $(n+)$ of argon ions with charge states $n+$ $(n=1-3)$ as a function of the energy of secondary electrons ejected from collisions of 12 keV electrons with argon. The relative uncertainties in partial double-differential cross-sections for Ar^+, Ar^{2+}, and Ar^{3+} ions are found to be 14%, 2%, and 6% respectively which are mostly within the size of symbols shown in the figure except for Ar^+. The TOF spectra correlating to low energy secondary electrons exhibit the singly charged argon ions to be most abundant and the relative partial DDCS of

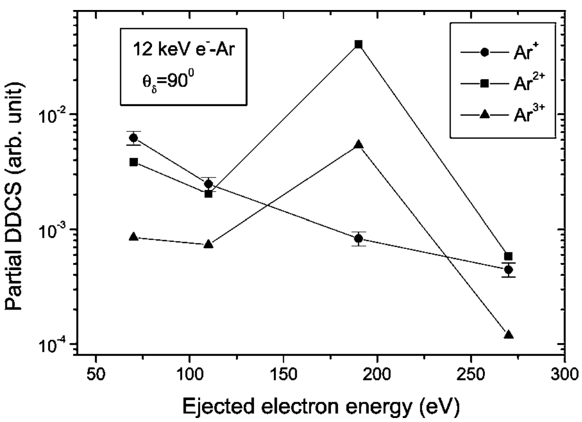

FIG. 2. Partial double differential cross-sections (PDDCS) for multiple ionization vs energy of the secondary electrons for 12 keV electron-argon collisions. The ejection angle θ_δ is 90°. Data are obtained for secondary electrons having energy 70–270 eV in coincidence with ● Ar^+, ■ Ar^{2+}, and ▲ Ar^{3+} ions. The lines connecting the data points are to guide eyes.

these ions are seen to decrease as the secondary electron energy increases to its largest value. This trend changes for electrons in the vicinity of argon *L-MM* auger transition at around secondary electron energy of 190 eV. There, the doubly and the triply charged ions are found to be more abundant than the singly charged ions.

The experimental data presented in Figs. 1 and 2 suggest that there is another channel through which the multiple ionization of the argon atom is produced. This channel is found to be responsible for the creation of an inner *L*-shell vacancy in the initial stage and then causing the vacancy filled subsequently through the *L-MM* auger transitions under a two-step-one (TS1) process. Hence, the TS1 process becomes more important for producing the multiple ionization of the target atom compared to the direct single and multiple ionizations caused by a TS2 process (as discussed in the Introduction) in the energetic collisions of present consideration.

The spectra of partial DDCS of argon ions in the characteristic auger transition region (around 190 eV) are interesting to examine more closely where the charge states Ar^{2+} and Ar^{3+} are found to be mostly produced by *L*-shell vacancy creation. A detailed analysis of the DDCS $(n+)$ spectra corresponding to electrons ejected due to *L-MM* auger transition may provide information about the L_1 and $L_{2,3}$ contributions to the multiple ionization, including the shake-off process. This information can be obtained by knowing the L_1 subshell $[E_i(2s)=320\text{ eV}]$ and $L_{2,3}$ subshell $[E_i(2p)=246.25\text{ eV}]$ contributions to double and triple ionization of argon atom due to transitions at 190 eV, where E_i is the binding energy of the corresponding subshell. The *L*-shell electronic transitions causing the emission of secondary electrons of 190 eV and contributing to the production of doubly and triply ionized argon ions are understood as follows: The double ionization of the atom is produced due to a direct ionization of the $L_{2,3}$ subshell by incident electrons, which subsequently gets filled by $L_{2,3}$-*MM* single auger process. Due to this transition, the $L_{2,3}$ shell contribution to the double ionization of argon is shown to be $0.89\sigma_{2p}$ [19], where, σ_{2p} is the $L_{2,3}$ shell ionization cross section obtained by using Lotz's formula [20]. And the triple ionization is produced due to a pure ionization of the $L_{2,3}$ subshell followed by a shake-off and a single auger process ($L_{2,3}M$-*MMM*). Due to these transitions, the $L_{2,3}$ subshell contribution for producing the triple ionization of argon is found to be $0.92\sigma_{2p}S$ [21], where S is a shake-off probability. Furthermore, the pure ionization of the L_1 subshell followed by L_1–$L_{2,3}M$ Coster-Kronig auger transitions (35–55 eV) provides the $L_{2,3}M$ vacancies which contribute to the production of triple ionization through $L_{2,3}M$-*MMM* auger transition. The contribution of the L_1 subshell for producing $L_{2,3}M$-*MMM* transition through $L_{2,3}M$ vacancies is only 5% [22]. Hence, the L_1 subshell's contribution to triple ionization of argon is not very significant compared to that of $L_{2,3}$ subshell. Therefore, the contribution of L subshells to produce Ar^{2+} and Ar^{3+} corresponding to the secondary electrons having energy 190 eV, i.e., DDCS $(2+)$ and DDCS $(3+)$ respectively can be written as:

$$\text{DDCS}(2+) = 0.89\sigma_{2p}, \tag{1}$$

$$\text{DDCS}(3+) = 0.92\sigma_{2p}S. \tag{2}$$

Hence, by combining the above relations with the experimental ratio of DDCS $(3+)$ to DDCS $(2+)$, the shake-off probability S can be readily estimated.

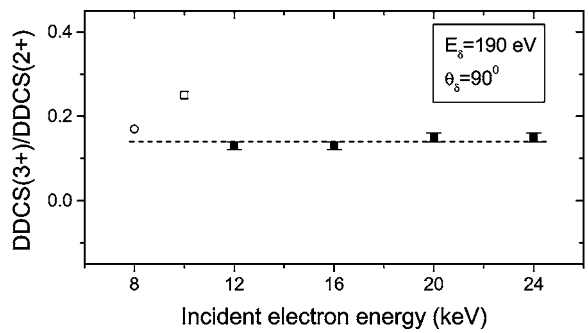

FIG. 3. Cross-section ratio DDCS $(3+)/\text{DDCS}(2+)$ for production of triply and doubly charged argon ions corresponding to an ejected electron energy of 190 eV at $\theta_\delta=90°$ vs incident electron energy. Present measurements: (■); dashed line is the best fit to our data points; Hippler *et al.* (Ref. 11), (○) and Van der Wiel and Wiebes (Ref. 22), (□).

Figure 3 displays the variation of the experimentally obtained ratios of DDCS $(3+)$ to DDCS $(2+)$ as a function of the incident energy ranging from 12 to 24 keV when argon ions were detected in coincidence with the secondary electrons of energy 190 eV ejected at 90°. The variation is found to be almost constant within the experimental uncertainty of about 7%. From this observation, we conclude that Ar^{3+} ions are formed mainly due to the shake-off process and their strength of formation is independent of the incident electron energy. Thus it may be stated that the triple ionization of argon occurs mainly due to the vacancy creation in the $L_{2,3}$ subshell by first-order interaction between a projectile and the active target electrons followed by second-order interactions among the target electrons. This second-order interaction being a part of the two-step-one (TS1) process for pro-

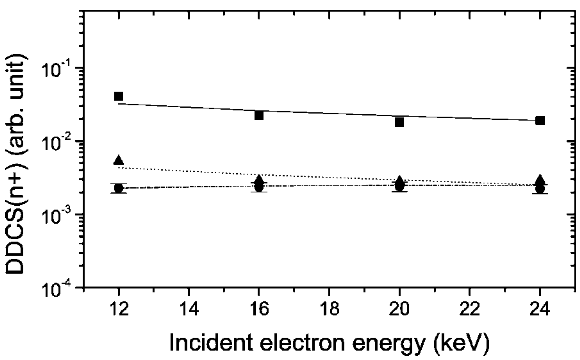

FIG. 4. n fold double differential cross sections DDCS $(n+)$ of argon ions corresponding to ejected electron energy of 190 eV at θ_δ=90° vs incident electron energy. Present measurements: ● Ar^+, ■ Ar^{2+}, and ▲ Ar^{3+}. Dotted and solid lines correspond to the calculations with $S=0.14$ for Ar^{3+} and Ar^{2+}, respectively. These curves have been normalized to experiment at impact energies of 20 keV and 24 keV, respectively. The dashed-dotted line represents the theory for a singly differential cross section of M-shell ionization (see text).

duction of triple ionization of argon atom is found to be independent of the velocity of the projectile and, therefore, the ratio reaches a constant value in the considered region of impact energy.

Further, in Fig. 3, we have shown two independently measured data points: one obtained at incident electron energy of 8 keV by Hippler *et al.* [11] and the other of 10 keV by Van der Wiel and Wiebes [22]. The data point of Hippler *et al.* is slightly higher than the present data whereas the data point of Van der Wiel and Wiebes is deviated by a large value. The deviation in the former case may be associated with the extraction and detection of ions while in the latter case, it arises due to the fact that the data point was obtained by performing an energy-loss experiment. Such measurements do not provide accurate ionization cross sections for $L_{2,3}$-MM transition yielding the secondary electrons of 190 eV.

Using the ratio of DDCS $(3+)$ to DDCS $(2+)$ as mentioned above [see Eqs. (1) and (2)], the shake-off probability for production of Ar^{3+} ions due to vacancy creation in the $L_{2,3}$ subshell is obtained by combining the DDCS $(2+)$ and DDCS $(3+)$ relations with the experimental ratio. The estimated value of the shake-off probability is found to be $S=0.14\pm0.01$. It is interesting to compare the shake-off probability S predicted by theoretical calculation $S=0.15$ by Carlson and Nestor [21] and $S=0.16$ by Van der Wiel and Wiebes [22] with its estimated value from our experiment $S=0.14$. The values are found to be in a very good agreement. In Fig. 4, the relative DDCS $(n+)$ for different charge states $n+$ $(n=1-3)$ of argon ions are plotted as a function of the incident electron energy. A satisfactory agreement between experiment and theoretical relations using $S=0.14$ for estimation of DDCS $(2+)$ and DDCS $(3+)$ as mentioned earlier [see Eqs. (1) and (2)] is obtained when we normalize our data with the theory, for example, at incident energy of 24 keV for Ar^{2+} (see Fig. 4). Furthermore, we have calculated the M-shell ionization cross sections in the plane-wave Born approximation using the analytical M-shell form factors given by Johnson *et al.* [23] and have compared with the experimental DDCS $(1+)$ values obtained for different impact energies. This comparison is done under the assumption that for a particular angle of secondary electron emission, the variation of corresponding single ionization yields by impact of electrons of various energies remains the same. A good agreement is observed between the experimental DDCS $(1+)$ values and the theoretically calculated M-shell ionization cross sections (see Fig. 4). The above comparisons lead to the conclusion that the singly charged argon ions are chiefly produced due to the direct outer M-shell ionization in the argon atom. However, such ions may also be produced by an autoionization process, the identification of which is not possible in the present experiment.

IV. CONCLUSIONS

We have investigated the details of multiple ionization process of argon atom under impact of 12–24 keV electrons. The examination of velocity dependence of partial double-differential cross sections shows that the electron correlation contributes substantially following the two-step-one process in the production of Ar^{3+} through electron shake-off with a probability $S=0.14\pm0.01$. This value is found to be in a very good agreement with the theoretical values given by Carlson and Nestor as well as Van der Wiel and Wiebes. From the energy spectrum of ejected electrons correlated to the produced multiply charged ions, it is found that the Ar^{2+} ions are dominantly produced by the vacancy creation in the L-shell of the atom followed by L-MM auger transition. In the considered collisions, the direct ionization of M-shell electrons produced by first-order interaction with the incident electrons is found to be the principal cause for formation of Ar^+ ions. The results obtained in the present work are expected to enhance our understanding about the multiple ionization and fragmentation processes of many electron atoms and molecules under impact of charged particles or photons.

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