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L-Shell Contributions to Multiple Ionization of Ar^{i+} Ions (i=1,2,3) by Electron Impact

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With the use of crossed beams of electrons and multiply charged argon ions, dominant contributions (above 90%) of L-shell ionization to multiple ionization of Ar^{i+} ions (i =1, 2, 3) at electron energies below 800 eV are identified. The observed contributions are explained in terms of the electron rearrangement processes following L-shell ionization.

In this Letter we report on direct measurements of absolute cross sections for multiple ionization of argon ions in charge states 1, 2, and 3 and explicitly demonstrate the dominance of a two-step process which involves the ionization of the L-shell and the subsequent emission of Auger and shakeoff electrons. To our knowledge this is the first experiment where multiple ionization of multiply charged ions has been quantitatively investigated. The obtained data provide, in particular, information about the relative strengths of multiple versus single electron interactions. The present results are directly relevant to the charge-state distributions of ions in very thin plasmas (like the interstellar medium) where multiple ionization by high-energy particle or photon bombardment may dominate.

It has been shown previously that the single ionization of ions can be enhanced by the excitation of inner-shell electrons to discrete states followed by autoionization processes.[1] Measured cross sections for C^{3+}, N^{4+}, and O^{5+} by Crandall *et al.*[2] show an increase of this enhancement with increasing charge state of the target ions. Calculations of Hahn[3] predict the dominance of the two-step process for the single ionization of molybdenum ions in charge states not less than +24. For multiple ionization, however, the dominance of a two-step process with ionization of inner shells is shown already in the present work for the removal of three electrons from Ar^{1+} in a single electron-ion collision.

The experimental investigation of such processes has been made accessible by use of an electron gun of high perveance, $P = 9.5\ \mu\mathrm{A\ V}^{-3/2}$, which provides a dense electron beam of about 180 mA/cm^2 at 1 kV and an interaction length of 60 mm combined with uniform electric potential in the region of the traversing ion beam. Thus the obtained signal rates are much higher than in earlier experiments of the crossed-beam type. Moreover, beams of multiply charged ions can be produced in an ion source with an ion flux sufficiently high to enable the detection of multiply ionized product ions by a single-particle detector.

The experimental procedure used was the same as that described in detail recently.[4] The obtained results for multiple ionization of Ar^{1+} ions are represented in Fig. 1. The cross sections $\sigma_{1,3}$ for double ionization and $\sigma_{1,4}$ for triple ionization show appearance potentials which coincide with the sum of the ionization potentials of the two, respectively, three outermost electrons. While $\sigma_{1,3}$ is a smooth function within the present experimental accuracy, the cross section $\sigma_{1,4}$ abruptly increases at about 250-eV electron energy which is the ionization threshold of the $L_{2,3}$ shell of Ar^{1+}. Furthermore, the observed cross section $\sigma_{1,5}$ seems to have an appearance potential which is not given by the sum of the ionization potentials of the four outermost electrons of Ar^{1+} but again by the ionization threshold for the $L_{2,3}$ shell. From these data it can be concluded that inner-shell–vacancy production with subsequent emission of electrons contributes to multiple ionization of Ar^{1+} ions.

For further investigation cross sections $\sigma_{2,4}$, $\sigma_{3,5}$, and $\sigma_{2,5}$ for double and triple ionization of Ar^{2+} and Ar^{3+} ions, respectively, have beem measured (see Fig. 2). For these ions the cross sections for double ionization already give evidence for L-shell contributions by the abrupt increase of $\sigma_{2,4}$ and $\sigma_{3,5}$ at the respective threshold energies for the ionization of the $L_{2,3}$ shell. In the case of $\sigma_{2,5}$ this effect is less obvious.

In summary, the experimental data shown in Figs. 1 and 2 provide strong evidence for an additional channel of multiple ionization of ions via L-shell ionization followed by electron emission. Compared with the direct ejection of two or more electrons the contribution of this two-

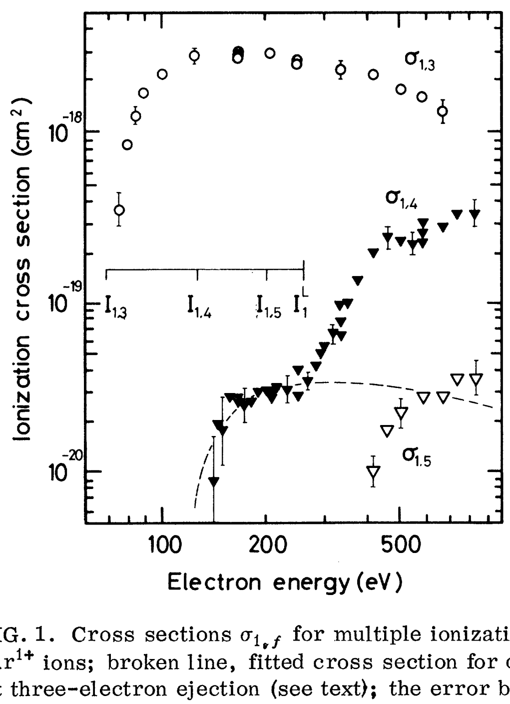

FIG. 1. Cross sections $\sigma_{1,f}$ for multiple ionization of Ar^{1+} ions; broken line, fitted cross section for direct three-electron ejection (see text); the error bars shown are determined from the square root of the quadrature sum of all contributing experimental uncertainties. The ionization thresholds $I_{1,f}$ of the f outermost electrons and I_1^L (Ref. 11) of the $L_{2,3}$ shell of Ar^{1+} are indicated.

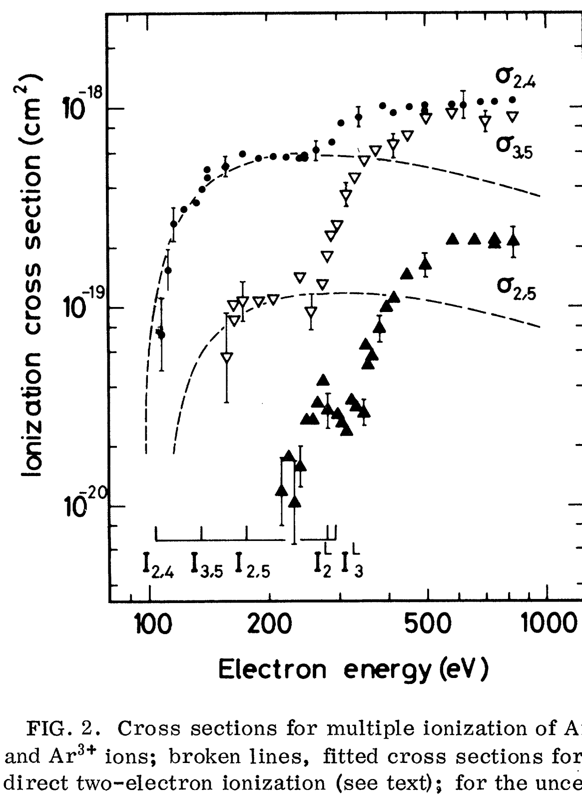

FIG. 2. Cross sections for multiple ionization of Ar^{2+} and Ar^{3+} ions; broken lines, fitted cross sections for direct two-electron ionization (see text); for the uncertainties see Fig. 1; the ionization potentials $I_{2,4}$, $I_{3,5}$, and $I_{2,5}$ for the outermost electrons and I_2^L, and I_3^L (Ref. 11) of the $L_{2,3}$ shell of Ar^{2+} and Ar^{3+}, respectively, are indicated.

step process [which is denoted by $\sigma_{i,f}(L)$ in the following, where i is the initial and f the final charge state of the ions] becomes more and more important for increasing charge state i and increasing number $(f-i)$ of released electrons. Although a (relatively small) effect could be expected, in principle, also for $\sigma_{1,3}$ near 250 eV the present measurements do not give evidence for a contribution $\sigma_{1,3}(L)$. This seems to be due to the presently limited experimental accuracy and the moderate line density of data points $\sigma_{1,3}$.

The inner-shell contribution $\sigma_{i,f}(L)$ can be determined by subtracting the partial cross section for the removal of the outermost electrons from the measured total cross sections. The energy dependence of the partial cross sections is approximated by fitting the data below the L threshold with the function $\sigma = (A/E_i E_e)\ln(E_e/E_i)$ with variable parameter A. E_e denotes the electron energy, and E_i the sum of the ionization potentials of the respective outermost electrons. The obtained fit functions are represented in Figs. 1 and 2 by the broken lines. Although the extrapolation of these curves to electron energies above 300 eV seems arbitrary the uncertainty resulting from the subtraction of the extrapolated cross sections from the measured total cross section is within some few percent since the observed contributions $\sigma_{1,f}(L)$ from the L shell are by far dominating beyond 300 eV. Only in the threshold region larger errors may occur.

The results of the procedure described above, i.e., the extracted cross sections $\sigma_{2,4}(L)$ and $\sigma_{3,5}(L)$ for twofold ionization, and $\sigma_{1,4}(L)$ for threefold ionization of Ar^{2+}, Ar^{3+}, and Ar^{1+} ions, respectively, are shown in Fig. 3. Also represented in this figure are cross sections $\sigma_{2,4}(L)$ and $\sigma_{1,5}(L)$. However, the estimate of the latter contributions is rather crude since there are not distinct well-isolated step increases in the total-ionization data which would allow the above subtraction procedure. An upper limit of the error bars including the cross sections $\sigma_{2,5}(L)$ has been estimated by the assumption that $\sigma_{2,5}(L)$ equals the measured cross section $\sigma_{2,5}$ and a lower limit is given by the procedure described above starting from the weakly indicated step in the $\sigma_{2,5}$ function at $E_e \approx 300$ eV. For $\sigma_{1,5}(L)$, only the upper limit is given with the assumption $\sigma_{1,5} = \sigma_{1,5}(L)$ which seems to be reasonable regarding the trends of the cross sections for direct multiple ionization.

For further comparison cross sections $\sigma_{0,2}(L_{2,3})$ for the single Auger process $L_{2,3}$-MM including

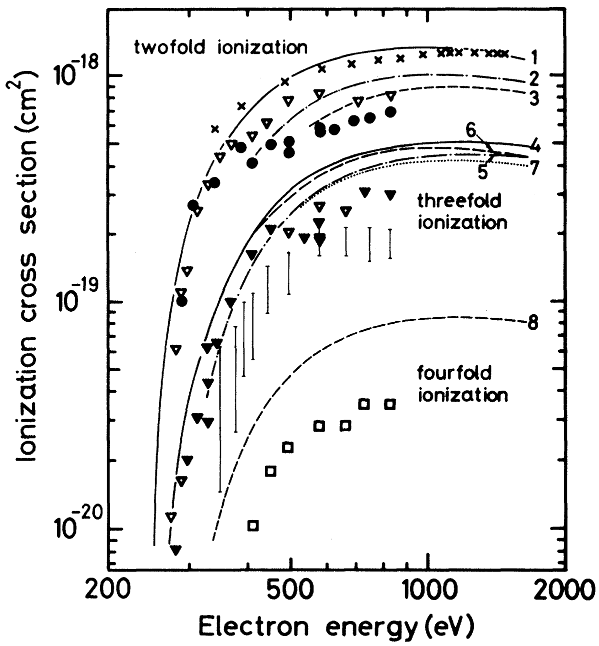

FIG. 3. Experimental cross-sections: $\sigma_{0,2}(L_{2,3})$ (Ref. 5), crosses; $\sigma_{2,4}(L)$, closed circles; $\sigma_{3,5}(L)$, inverted open triangle for twofold ionization; $\sigma_{1,4}(L)$, inverted closed triangle; $\sigma_{2,5}(L)$, data points enclosed in error bars for threefold ionization; and $\sigma_{1,5}(L)$, open square for fourfold ionization via L-shell ionization of Ar ions (see text). The lines 1–5 represent cross sections calculated from the Gryziński theory (Ref. 10) and the branching ratios for electron emission processes following L-shell ionization (see text): 1, $\sigma_{0,2}(L)$; 2, $\sigma_{2,4}(L)$; 3, $\sigma_{3,5}(L)$; 4, $\sigma_{1,4}(L)$; 5, $\sigma_{2,5}(L)$. The lines 6, 7, and 8 have been obtained with use of the relative abundances of ions generated after L-shell ionization of Ar atoms (Ref. 12): 6, $\sigma_{1,4}(L)$; 7, $\sigma_{2,5}(L)$; 8, $\sigma_{1,5}(L)$.

shakeup transitions during the Auger process, i.e., $L_{2,3}$-MMM^*, initiated by electron impact are included in Fig. 3, which have been determined from Auger-spectroscopy measurements[5] with neutral Ar. Figure 3 exhibits three separate groups of L-shell contributions to the double, threefold, and fourfold ionization of argon in different charge states. The cross sections in each group, i.e., for different charge states i of the parent ion, do not differ very much from each other. This seems to be easily intelligible if only the binding energies of Ar^{i+} $L_{2,3}$ electrons are considered which remain within 270± 20 eV for charge states i between 0 and 3. On the other hand, for a complete understanding of the observed phenomena the deexcitation processes subsequent to the production of a vacancy in the L shell have to be taken into account also.

It is known from earlier work[6] that the $L_{2,3}$ fluorescence yields for the $2p^{-1}\,3p^{-n}$ configurations of Ar are between 1.48×10^{-4} for $n=0$ and 3.78×10^{-3} for $n=3$. The situation is similar for the L_1 shell. Therefore deexcitation after L-shell ionization practically takes place via electron-emission processes. In the case of $n=0$ the branching ratios of the different possible processes are known. If we differentiate only between L_1 shell $[E_i(2s)=326.5$ eV$]$ and $L_{2,3}$ shell $[E_i(2p)=249.2$ eV$]$,[7] contributions to double and triple ionization of neutral Ar have to be considered (the spectroscopic notation for each process is given in parentheses together with the pertinent cross section), and they are given below.

The L-shell processes contributing to double ionization are as follows: 1(α) pure ionization of the $L_{2,3}$ shell (cross section σ_{2p}) followed by a single Auger process[8] ($L_{2,3}$-MM; $\sigma^{1\alpha}=0.89\sigma_{2p}$); 1($\beta$) pure ionization of the $L_{2,3}$ shell followed by an Auger process combined with a shakeup transition ($L_{2,3}$-MMM^*; $\sigma^{1\beta}=0.053\sigma^{1\alpha}=0.047\sigma_{2p}$) (Ref. 5); 1($\gamma$) pure ionization of the L_1 shell (cross section σ_{2s}) followed by a single Auger process (L_1-MM; $\sigma^{1\gamma}=0.052\sigma_{2s}$) (Ref. 7).

The L-shell processes contributing to triple ionization are as follows: 2(α) pure ionization of the $L_{2,3}$ shell followed by a double Auger process ($L_{2,3}$-MMM; $\sigma^{2\alpha}=0.12\sigma^{1\alpha}=0.11\sigma_{2p}$) (Ref. 8); 2($\beta$) ionization of the $L_{2,3}$ shell with shakeoff[9] ($L_{2,3}M$; $0.15\sigma_{2p}$) followed by a single Auger process ($L_{2,3}M$-MMM; $\sigma^{2\beta}=0.89\times0.15\sigma_{2p}=0.13\sigma_{2p}$) (Refs. 8 and 9); 2($\gamma$) pure ionization of the L_1 shell followed by a Coster-Kronig transition (L_1-$L_{2,3}M$; $0.95\sigma_{2s}$) (Ref. 7) which is then followed by a single Auger process ($L_{2,3}M$-MMM; $\sigma^{2\gamma}=0.89\times0.95\sigma_{2s}=0.84\sigma_{2s}$) (Ref. 8). Higher-order processes which contribute less significantly have been omitted.

Since, as Fig. 3 indicates, the branching ratios given above do not strongly depend on the charge state of the parent Ar, it is possible to calculate $\sigma_{i,i+2}(L)$ and $\sigma_{i,i+3}(L)$ for Ar^{i+} with $i=0,\ldots,3$ if the pertinent cross sections σ_{2s} and σ_{2p} are known. The latter can be evaluated from the theory of Gryziński[10] by use of the respective ionization potentials of L electrons in Ar^{i+} ions.[11] From the very good agreement between this theory and measured cross-sections σ_{2p} for $i=0$ (Ref. 5) reasonable results can be expected in the other cases, too. The cross sections $\sigma_{0,2}(L)$, $\sigma_{2,4}(L)$, and $\sigma_{3,5}(L)$ for double ionization and $\sigma_{1,4}(L)$ and $\sigma_{2,5}(L)$ for triple ionization calculated on the basis of constant branching ratios for the electron-emission processes are also shown in Fig. 3.

A second method to determine these cross sections is provided by an earlier measurement of the relative abundances of ions formed as a result of L-shell vacancies in Ar atoms.[12] The determined fractions Ar^{1+}:Ar^{2+}:Ar^{3+}:Ar^{4+} are 0:2:72:24 for an initial L_1 vacancy and 0:74:24:2 for an initial $L_{2,3}$ vacancy. With the assumption of constant relative abundances for different parent ion charge states the cross sections $\sigma_{1,4}(L)$, $\sigma_{2,5}(L)$, and $\sigma_{1,5}(L)$ have been calculated using the Gryziński theory and are also shown in Fig. 3.

Although the calculations have only considered the branching ratios for neutral Ar the agreement with the data obtained for ions is surprisingly good. It might be inferred at most from the results represented in Fig. 3 that with increasing ion-charge state the probability for the emission of two or more electrons following L-shell ionization slightly decreases and the single Auger process more and more prevails.

In conclusion, we have shown that dominant contributions of the L shell occur for multiple ionization of Ar ions.

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