

Investigation of Inelastic Processes Occurring During Collisions between K^+ Ions and Ar Atoms

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Submitted July 23, 1971

Zh. Eksp. Teor. Fiz. **62**, 61–69 (January, 1972)

Excitation and ionization processes and also their relative roles in slow ion-atom collisions are studied. For this purpose the differential scattering cross sections, ion inelastic energy loss spectra, and ultraviolet emission spectra are studied for collisions between K^+ ions and Ar atoms. The initial ion energy is 2.0 keV. Simultaneous use of the differential scattering and spectroscopic techniques made it possible to study both excitation of the energy levels and decay of the respective excited states. Investigation of excitation processes showed that in K^+ -Ar collisions, $3p^54p$ and $3p^53d$ levels in Ar are excited with highest probability. The cross section for direct excitation of $3p^54s$ Ar levels, notwithstanding that they are the lowest-lying levels, does not comprise more than 5 to 10% of the total cross section for excitation of Ar levels ($7 \cdot 10^{-17}$ cm²). In contrast to Ar, on excitation of the K^+ ion the $3p^54s$ levels are excited with highest probability. The contribution of cascade transitions to Ar resonance-line excitation is determined; the contribution is significant and is 70–90%. Estimates are made of the contributions to the total ionization cross section of ionization processes, ionization processes involving excitation of the ion, and autoionization processes. The contributions of the direct ionization process and ionization processes with ion excitation do not exceed 5 and 25%, respectively. Autoionization processes are of decisive importance in Ar ionization.

I. INTRODUCTION

IN ion-atom collisions, the fraction of the kinetic energy of the relative motion of the colliding particles that is consumed by inelastic electronic transitions (inelastic energy loss) is differently distributed among the different inelastic channels. The experimental determination of the relative roles of different channels encounters considerable difficulties. These difficulties are connected mainly with the large number and with the variety of the inelastic processes that are realized even when the collision energy is on the order of a kiloelectron volt.

A convenient approach to the solution of the problem is to perform simultaneous measurements of the spectra of the inelastic energy losses of the incoming particle at different values of the scattering angle and of the emission spectra or energy spectra of the electrons released in the course of the collision. The method wherein the inelastic scattering is analyzed, unlike most other methods, makes it possible to determine directly the cross sections for the excitation of the energy levels from the ground level, regardless of the transition energy and regardless of the manner in which the corresponding excited states decay subsequently, with release of electrons or with emission of photons. Consequently, it becomes possible to determine in a single experiment the cross sections of processes that are customarily investigated by different methods, such as the excitation of the target atom and of the incident ion, single and multiple ionization of the colliding particles, ionization with excitation and autoionization of the target atoms. In those cases when the excited state decays via several channels as a result of subsequent realignment of the electron shell, the spectra of the inelastic losses make it possible to determine the total cross section of the corresponding processes. The relative cross section of each of the processes can be deter-

mined by comparing the spectra of the inelastic losses with the emission spectra.

In the present study we determined the spectra of the ion inelastic energy losses in the region from 0 to 50 eV for scattering through angles from 1 to 10°, and the emission spectra in the wavelength range from 1100 to 500 Å.

II. MEASUREMENT PROCEDURE AND RESULTS

1. Spectra of Inelastic Energy Losses

The procedure for obtaining the inelastic-loss spectra and a brief description of the experimental setup are given in^[1,2]. A collimated ion beam entered a collision chamber filled with the investigated gas. A rotating collimator was used to separate the ions scattered through fixed angles in the course of single collisions. These ions were energy-analyzed by an electrostatic analyzer with parallel plates and detected by a secondary-electron multiplier. The inelastic energy loss spectra were recorded automatically. The relative energy resolution of the setup was 1300, and the angular resolution 20'; the relative error in the determination of the inelastic energy losses was 1–2%.

Typical inelastic energy-loss spectra are shown in Fig. 1. It is seen from the figure that the spectra have a discrete structure. The first peak in the loss spectrum corresponds to inelastically scattered ions. From the shape and the width of this peak one can estimate the apparatus function of the setup. The remaining peaks in the loss spectra were identified using data on the energy levels of the incoming ion and the target atom^[3,4]. The second peaks in the spectra corresponds to excitation of 3p electrons and single ionization of the Ar atom, the third to excitation of 3p electrons of the K^+ ions, and the fourth to excitation of autoionization states of the Ar atom, connected with excitation of one 3s or two 3p electrons and to ionization with excitation. An investi-

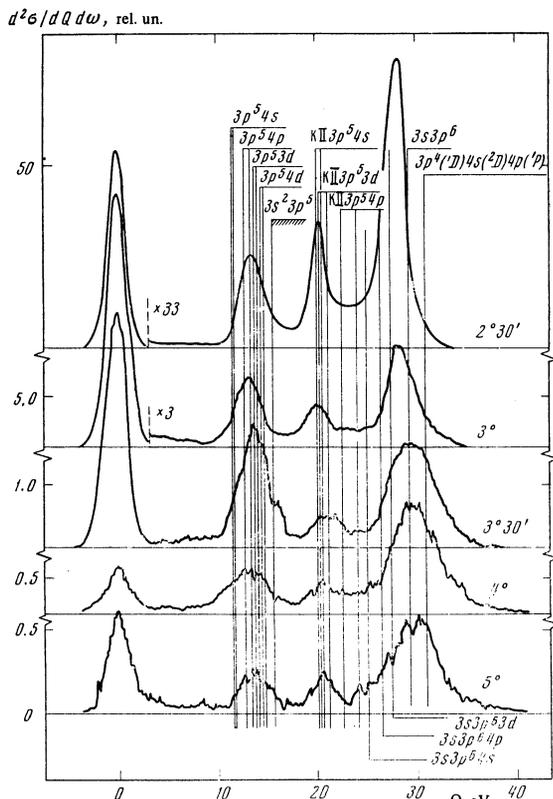


FIG. 1. Inelastic energy loss spectra of K^+ ions scattered through different fixed angles by Ar atoms. Initial energy 2.0 keV. The vertical lines indicate the positions of the energy levels of the isolated Ar and K^+ .

gation of the dependence of the area of each of the peaks on the scattering angle of the incident ions has shown that the excitation cross sections of the investigated transitions have a threshold character. As the scattering angle is increased, the cross section of each inelastic transition remains small up to a certain threshold value of the scattering angle. When the threshold angle is reached, the cross section of the transition increases sharply. With further increase of the scattering angle, the cross sections of the inelastic transitions decrease, experiencing oscillations that are pronounced to a greater or lesser degree. The threshold character of the angular dependences of the differential cross sections of the inelastic transitions makes it possible to integrate these cross sections with respect to the scattering angle and to determine in this manner the relative contributions of the different inelastic transitions corresponding to the structure in the loss spectrum. Among the different processes that lead to ionization of the target atom, a decisive role may be played, as shown in^[2], by the processes of autoionization and of ionization with excitation. These processes correspond to the fourth peaks in the loss spectra. Direct ionization (direct removal of an electron to the continuous spectrum without formation of an intermediate autoionization state) and multiple ionization, as shown in^[2], make no noticeable contribution to the effective cross section of the target-atom ionization.

It can thus be concluded that the integral of the area of the fourth peak with respect to the scattering angle corresponds, accurate to several percent, to the effective

cross section for the ionization of the target atom (the cross section of the electron emission σ_-). This correspondence makes it possible to estimate, by making independent measurements of the effective ionization cross section of the target atoms, the absolute values of the differential scattering cross sections obtained in the present study. Such an estimate of the differential scattering cross sections was obtained by comparing our relative measurements at an ion energy 2.0 keV with the ionization cross section obtained in^[5], equal to $1.8 \times 10^{-16} \text{ cm}^{-2}$ at this energy. The error in the determination of the absolute value of this cross section, in accordance with the estimate made in^[5], is 15%. The reproducibility of our relative measurements of the cross sections is 10%. In addition, the use of calibration in the determination of the absolute values of the differential cross sections leads to an additional error of approximately 15%.

2. Emission Spectra

The experimental setup and the procedure used to investigate the emission in the region of the vacuum ultraviolet are described in^[6,7]. The emission was analyzed with the aid of a vacuum monochromator with a diffraction-grating radius $R = 0.5 \text{ m}$ (1200 lines/mm) and a linear dispersion 17 \AA/mm . The radiation was registered with an open electron multiplier of the Venetian-blind type, the first dynodes of which were gold plated.

The spectrum of the emission produced in the ultraviolet region when K^+ ions collide with Ar atoms is shown in Fig. 2. The part of the spectrum shown in the figure contains all the intense emission lines located in the range 1100–500 \AA .

The lines were identified with the aid of the spectral-line tables^[8]. Lines 1–5 (see the caption of Fig. 2) were observed in first order of the spectral resolution, and lines 6–13 in the second order.

Of great importance in the determination of the contribution of the processes of excitation and ionization with excitation to the inelastic energy loss cross sections is the determination of the relative intensities of these lines. In the present study, the transmission coefficient of the spectral instrument and the efficiency of the detector were not specially investigated, so that a comparison of the relative intensities is possible only for spectral lines that are close in wavelength and have furthermore been obtained in the same order of spectral resolution. Such a comparison presupposes that the transmission of the instrument and the registration efficiency do not experience abrupt changes in a spectral region of 10–20 \AA , and that the polarizations of the compared lines do not differ significantly. For a qualitative determination of the dependence of the transmission coefficient on the wavelength, we used measurements of the efficiency of the diffraction grating^[9] used in the present experiment. Since the efficiency measurements were performed only in the wavelength range 950–2400 \AA , these measurements were extrapolated to 860 \AA in order to estimate the total intensity of the 879.9, 869.7, and 866.8 \AA lines of Ar I.

The radiation-registration efficiency was determined mainly by the photoemission coefficient of the first

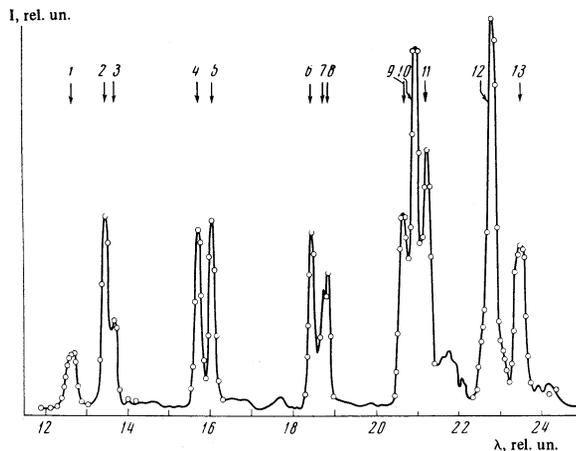


FIG. 2. Emission spectrum in the 500–1100 Å range for K^+ –Ar collisions. Initial ion energy 2.0 keV. Line identification:

- 1 — Lines of Ar I: 879.9 Å ($3p^4 1S - 5s [1/2]^o + 869.8$ Å ($3p^4 1S - 5s' [1/2]^o$) + 866.8 Å ($3p^4 1S - 3d' [1/2]^o$),
 2, 3 — Lines of Ar II: 919.8 Å ($3p^3 2P^o - 3p^3 2S$), 932.0 Å ($3p^3 2P^o - 3p^3 2S$),
 4, 5 — Lines of Ar I: 1048.2 Å ($3p^4 1S - 4s' [1/2]^o$), 1066.7 Å ($3p^4 1S - 4s [1/2]^o$),
 6–8 — Lines of K II: 600.8 Å ($3p^4 1S - 4s' [1/2]^o$), 607.9 Å ($3p^4 1S - 3d [1/2]^o$),
 612.6 Å ($3p^4 1S - 4s [1/2]^o$),
 9–11 — Lines of Ar II: 666.0 Å ($3p^3 2P^o - 3 d^2 P$) + 664.6 Å ($3p^3 2P^o - 3 d^2 D$),
 672.8 Å ($3p^3 2P^o - 4s^2 D$) + 671.8 Å ($3p^3 2P^o - 4s^2 D$) + 670.9 Å ($3p^3 2P^o - 4 s^2 D$),
 679.4 Å ($3p^3 2P^o - 4s^2 D$) + 679.2 Å ($3p^3 2P^o - 3 d^1 P$) + 678.0 Å ($3p^3 2P^o - 3 d^1 P$),
 12–13 — Lines of Ar II: 725.6 Å ($3p^3 2P^o - 4s^2 P$) + 723.4 Å ($3p^3 2P^o - 4s^2 P$),
 745.3 Å ($3p^3 2P^o - 4s^2 P$) + 744.9 Å ($3p^3 2P^o - 4s^2 P$) + 740.3 Å ($3p^3 2P^o - 4s^2 P$)

dynode of the secondary-electron multiplier. The dynode was gold plated and the data on the photoemission of gold^[10] were therefore used to estimate the registration efficiency as a function of the wavelength.

Estimates of the transmission coefficient and of the registration efficiency in the wavelength range 850–850–1050 Å have shown that the total intensities of lines 1 (Ar I), 2 and 3 (Ar III), and 4 and 5 (Ar I) have a ratio 1 : 1.8 : 1.7. The error in this ratio apparently does not exceed a factor of two. In spite of the relatively large error, these data, as will be shown later, are useful for the determination of the relative roles of the processes of excitation, ionization with excitation, and autoionization of the target atom. The relative intensities within the limits of a group of two neighboring lines (2 and 3, 4 and 5, 6 and 7, 8) can be determined directly from Fig. 2.

III. DISCUSSION OF RESULTS

The data on the cross sections for inelastic losses and the cross sections for the excitation of the spectral lines in the ultraviolet region of the spectrum were used to estimate the cross sections of a number of inelastic processes and for the determination of the relative roles of these processes in the dissipation of the inelastic energy loss for each of the partners of the K^+ –Ar collision.

1. Relative Role of the Excitation and Direct Ionization of Ar

The energy-loss spectra make it possible to determine (from the shape of the peak and from the position of its maximum) the relative probability for the excitation of individual levels of Ar. It is seen from Fig. 1 that the maximum of the second peak in ion scattering through angles making the decisive contribution to the

excitation cross section of Ar ($2^\circ 30' - 3^\circ 30'$), does not go outside the energy-loss region 13.3–13.8 eV. It follows therefore that the levels excited with the largest probability in the course of the collision are those with the configurations $3p^5 4p$ and $3p^3 3d$ (and possibly levels with configuration $3p^5 5s$, whose energy is close to the energy of the levels of the configuration $3p^5 3d$). The slight dependence of the position of the maximum of the second peak on the scattering angle is apparently due to the change in the relative probability of the excitation of the levels with configurations $3p^5 4p$ and $3p^5 3d$ ($3p^5 5s$) when the distances between the colliding particles decrease to different values.

The second peaks in the energy-loss spectra (Fig. 1) correspond, as already noted above, not only to excitation processes, but also to direct ionization of the Ar atom. Integration of the area of this peak with respect to the scattering angle yielded a value 7×10^{-17} cm² for the sum of the cross sections of the indicated processes. In the figure, the direct ionization process corresponds to that part of the second-peak area which lies beyond the boundary of the continuous spectrum, i.e., to the right of the $3s^2 3p^5$ line. Estimates of this part of the peak area, performed for all scattering angles with allowance for the influence of the apparatus function of the setup on the excitation line contours, have shown that the cross section for the process of direct ionization does not exceed 5% of the total cross section for the excitation of the Ar atom.

From an analysis of the inelastic loss spectra it also follows that the cross section for the excitation of the levels with configuration $3p^5 4s$ from the ground level is much smaller than the cross section for the excitation of the levels $3p^5 4p$, even though these levels lie at lower energies, and amounts to 5–10% of the total cross section for the excitation of the Ar atom, i.e., $(3-7) \times 10^{-18}$ cm².

Thus, in view of the smallness of the direct-ionization cross section it can be assumed that the cross section value given above (7×10^{-17} cm²) corresponds mainly to the total cross section for the excitation of the Ar atom, and that a decisive contribution to this cross section is made by excitation of the energy levels with configurations $3p^5 4p$ and $3p^5 3d$ ($3p^5 5s$).

2. Role of Cascade Transitions in the Excitation of the Ar Resonance Lines

In the investigation of the excitation function of the spectral lines in optical experiments, considerable interest attaches to a clarification of the relative role of the processes that lead to excitation of the levels for which these lines originate. Decisive among these processes, if only single collisions are involved, are direct excitation from the ground-state level and transitions from higher levels.

As noted earlier, the energy-loss spectra make it possible to determine directly the cross section for direct excitation of the levels. Therefore a comparison of the loss spectra with spectroscopic measurements of the emission cross sections of the lines for which the investigated levels are the upper ones is a convenient method of revealing the role of cascade transitions in the population of these levels. We note that the deter-

mination of the effective direct-excitation cross sections by spectroscopic methods encounters great difficulties, connected with the fact that the wavelength range of the spectral lines whose cross sections must be determined for this purpose can extend from the short ultraviolet to the infrared.

It is seen from Fig. 2 that the atomic lines of Ar most intensely excited in the ultraviolet region of the spectrum are the resonance lines $3p^6\ ^1S - 4s'[1/2]^0$ and $3p^6\ ^1S - 4s[1/2]^0$, and also the group of lines corresponding to the transitions from the levels $5s[1/2]^0$, $5s'[1/2]^0$, and $3d[1/2]^0$ to the ground-state level. At the same time, the cross section for direct excitation of the levels with configuration 4s, which are the upper ones for the resonance lines, is only 5–10% of the total cross section for the excitation of Ar (see Sec. III, Item 1). It can therefore be concluded that the levels with configuration 4s are populated mainly via cascade transitions from the upper levels.

For a quantitative determination of the role of the cascade transitions in this case, let us estimate the total cross section for the excitation of the 4s levels. There are four Ar energy levels corresponding to the configuration $3p^54s$, two resonant and two metastable. Since all the levels of these configurations are populated mainly via cascade transitions, and since the intensities of the resonance lines turned out to be equal, as seen from Fig. 2, we can assume that the number of transitions to each of the levels is approximately the same. Recognizing that the summary intensities of the resonance lines and of the lines whose upper levels are $3d'$, $5s$, and $5s'$ have a ratio 1.7 : 1, we find that when cascade transitions are taken into account, the ratio of the cross section for excitation of levels with configuration 4s (including the metastable levels) to the cross section for the excitation of the levels $3d'$, $5s$, and $5s'$ is 3.4 : 1. This yields for the total excitation cross section of the 4s levels an approximate value 5×10^{-17} cm². Comparing this cross section with the estimate of the cross section for direct excitation of the 4s levels ($3 \times 10^{-18} - 7 \times 10^{-18}$ cm²) we find that in collisions between 2.0-keV K⁺ ions with Ar atoms the contribution of the cascade transitions to the excitation of the Ar resonance lines is quite large and amounts to 70–90%. Some uncertainty in this value is due to the fact that we still do not know what fraction of the cross section obtained by us for the direct excitation of the 4s levels is connected with excitation of resonance levels and what fraction with the excitation of metastable levels.

3. Processes of Excitation of the K⁺ Ion

It is seen from the inelastic-loss spectra that when K⁺ ions are excited the levels excited with the largest probability are those with the configurations $3p^54s$ and $3p^53d$. The cross section for the excitation of these levels, obtained by integrating the areas of the third peaks in the loss spectra with respect to the scattering angle, is 4×10^{-17} cm².

An analysis of the inelastic-loss spectra also makes it possible to estimate the cross section for the excitation of levels with configuration $3p^54p$. Accurate to a coefficient 1.5, the cross section amounts to 1.6×10^{-17} cm². The value of the coefficient is determined

mainly by the error with which it is possible to separate the region of the inelastic-loss spectrum corresponding to excitation of only $3p^54p$ levels of the K⁺ ion. Thus, when the K⁺ ion is excited, unlike Ar, the excitation of the lowest level is more favored. It follows from these data that the resonance levels of the configurations $3p^54s$ and $3p^53d$ are excited mainly directly.

The excitation of the resonance lines of the K⁺ ions in collisions with Ar atoms was previously investigated in^[11,12]. It was observed that a noticeable intensity is possessed only by the resonance lines with wavelengths 600.7, 607.9, and 612.6 Å, corresponding to transitions from the $4s[1/2]^0$, $4s'[1/2]^0$, and $3d[1/2]^0$ levels to the ground level (see Fig. 2, lines 6–8).

Absolute measurements^[12] showed that the total cross section for the excitation of the resonance lines of the K⁺ ion at an ion energy 2.0 keV is 4×10^{-17} cm², i.e., it coincides with the cross section for the excitation of the states $3p^54s$ and $3p^53d$ determined from the inelastic-loss spectra.

It should be borne in mind that in the present study we measured the summary cross section for the excitation of the resonance and metastable levels of the 4s and 3d configurations of the K⁺ ion, whereas the measurements in^[12] were made on the resonance levels of these configurations. One should therefore expect the cross section obtained in the present paper to be larger than the cross section obtained in^[12]. The equality of these cross sections may be due to the fact that in atomic collisions the cross sections for the excitation of metastable levels of the configurations 4s and 3d from the ground level are noticeably smaller than the cross sections for the excitation of the resonance levels. The last premise, however, calls for a serious verification, since the measurement procedure used in^[12] can exaggerate the cross section somewhat, owing to the uncontrollable contribution of a number of Ar II lines excited simultaneously with the resonance lines.

4. Relative Role of Autoionization Processes and Processes of Ionization with Excitation of Ar

The fourth peaks in the spectra of the inelastic energy losses (Fig. 1) correspond to processes of autoionization and ionization with formation of Ar⁺ ions in excited states. It is seen from the figure that among these processes the highest probability is possessed by those whose excitation requires an energy expenditure of 25–35 eV. In the loss region from 25.3 to 29.2 eV, the energy is expended mainly on excitation of the autoionization levels of Ar, corresponding to the transitions of the 3s electron from the ground state to the s, p, and d states.

The limit of the s, p, d series of the autoionization levels corresponds to an energy loss $Q = 29.24$ eV (excited-ion state $3s3p^6\ (^2S_{1/2})$). That fraction of the fourth peak of the inelastic-loss spectrum which is located in the region $Q > 29.24$ eV is connected with energy loss to autoionization processes corresponding to excitation of two 3p electrons, and to ionization into the excited states of the Ar⁺ ion.

It was mentioned earlier (Sec. II.1) that the processes corresponding to the fourth peaks play the decisive role in the ionization of Ar. Let us determine the rela-

tive contribution of these processes to the excitation of the fourth peak of the loss spectrum. It is seen from the figure that when the ions are scattered through angles $\vartheta \leq 3^\circ$ the maximum of the peak lies in the region $Q < 29.2$ eV and, consequently, the largest contribution to the ionization cross section in the scattering of ions through angles $\vartheta \leq 3^\circ$ is made by the autoionization processes connected with the excitation of the 3s electron. With increasing scattering angle, the contribution of these processes decreases and amounts to less than 50% at 4° . An analysis of the inelastic-loss spectra in the region of the fourth peak shows about half of the ionization cross section is due to autoionization processes corresponding to excitation of 3s electrons, and about half to processes corresponding to excitation of two 3p electrons, and possibly to processes of ionization to the excited states of the Ar^+ ion.

To determine the relative contribution of the latter processes, we turn to the emission spectra of Ar^+ (Fig. 2). It follows from the figure that among the processes of ionization into excited states of the ion, those capable of contributing to the fourth peaks of the inelastic-loss spectra can be processes leading to the emission of lines 2 and 3 (resonance lines of Ar^+ ion), 9–11, and 12 and 13 (see the caption of Fig. 2). In the estimate obtained above (Sec. II.2) for the relative intensities of the emission-spectrum lines it was found that the total intensity of the resonance lines of the ion (lines 2 and 3) is approximately equal to the total intensity of the resonance lines of the atom (lines 4 and 5). According to our estimates, the cross section for the excitation of the resonance lines of the atoms is $\sim 2.5 \times 10^{-17}$ cm². The cross section for the excitation of the resonance lines of the ion (λ 919.8 Å and λ 932.05 Å) should be approximately the same. The excitation cross section of these lines consists of the cross sections for ionization with excitation, charge exchange with excitation, and cascade transitions to the $3p^6\ ^2S$ level of the Ar^+ ion, which is the upper level for the resonance lines. Therefore the cross section of the process of ionization with excitation of the $3p^6\ ^2S$ state should be somewhat smaller than 2.5×10^{-17} cm², and consequently the contribution of this process to the ionization cross section (excitation cross section of the fourth peak of the loss spectrum) does not exceed 15%.

Processes of ionization with excitation of the lines

9–11 and 12 and 13 also make no noticeable contribution to the ionization cross section. This conclusion follows from an estimate of the contribution made to the fourth peaks of the loss spectra by those parts of these peaks lying in the loss region 34.0–34.4 eV and those in the region 32.4–33.0 eV, corresponding to ionization with excitation of the line groups 9–11 and 12 and 13, respectively. It is seen from Fig. 1 that the contribution of these parts to the fourth peaks of the loss spectra is insignificant (<10%). The excitation of lines 9–13 is apparently connected with the process of charge exchange with excitation of the Ar^+ ion.

Comparing the estimates of the cross sections for the excitation of the lines of the ion 2, 3, 9–13 with the total ionization cross section and recognizing that the excitation of the indicated lines may be connected with the processes of charge exchange and excitation of the ion, we conclude that in slow collisions between K^+ ions and Ar atoms ($T_0 = 2.0$ keV) the decisive role in the Ar ionization is played by autoionization processes.

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Translated by J. G. Adashko

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