# AUTO-IONIZATION STATES IN ARGON

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Submitted January 3, 1969

Zh. Eksp. Teor. Fiz. 57, 27–37 (July, 1969)

Results of an investigation of the energy spectrum of electrons released in collisions between protons and argon atoms are presented. The electron spectrum was investigated for electron energies $E_e$, between 5 and 300 eV and a proton energy T = 20 keV. The structural part of the spectrum is identified and its nature is explained. It is shown that the observed spectral lines are due to auto-ionization transitions in argon. More precisely, the structure of the low-energy part of the spectrum ($E_e$ = 5–16 eV) is due to excitation of the argon M shell; the spectral range corresponding to electron energies $E_e$ = 28–70 eV is connected with Auger transitions of the Koster-Kronig type, and the spectral ranges at electron energies 100–240 and 240–300 eV are due to excitation of the $L_{23}$ and $L_1$ subshells in Ar respectively. It is pointed out that $3s3p^5 4s4p$ auto-ionization states may be produced and decay into excited $Ar^+$ ions. Spectral lines connected with Auger transitions from the initial states, in which the inner electron goes over into the excited optical level, have been observed, as well as lines due to three-electron Auger transitions.

## INTRODUCTION

In our earlier paper[1] we presented preliminary results of an investigation of the energy distribution of the electrons released when $Ar^+$ ions collide with Ar atoms. We observed a clearly pronounced structure, whose presence we attributed to auto-ionization transitions in the isolated Ar atom excited in the collision. In the case of the $Ar^+$-Ar pair, however, the identification of the individual spectral lines is made difficult by the presence of "Doppler" peaks connected with emission of the electron from the fast particle. It was therefore advisable, before investigating the $Ar^+$-Ar system in detail, to study the energy distribution of the electrons released from the Ar atom by collision with an elementary particle (a proton), for in this case the observed spectrum is connected only with the release of electrons from the target (argon) atom.

The energy spectra of the electrons released in collisions with Ar atoms were investigated earlier by Blaut[2] and by Rudd et al.[3,4] However, the information obtained in these investigations offers only an approximate representation of the character of the investigated spectra. Thus, in Blaut's work[2] the energy interval between the investigated measurement points greatly exceeded the resolution of the instrument, so that the structure features of the energy spectrum were not revealed, and the data obtained by him give an idea of only the averaged electron distribution function. In[3,4] the structure of the energy distribution was investigated in greater detail, but only a small number of the observed spectral line were identified. A detailed plot of the energy distribution, from which it is possible to determine the positions of the individual peaks, is given by Rudd et al.[3] only for small electron-energy intervals (3–20 and 200–212 eV).

The procedure used in our experiment had higher sensitivity and a low relative measurement error, and made it possible not only to establish with sufficient accuracy the positions of the individual peaks noted in earlier investigations, but also to observe new singularities on the electron energy distribution curve. In addition, we obtained an almost complete identification of the spectral lines and attempted to explain the nature of the observed peaks.

A detailed description of the experimental procedure is contained in our paper[5]. The electron-energy analyzer was a cylindrical electrostatic analyzer of the Blaut type[2], which we perfected. The electron emission angle was 54.5° relative to the axis of the primary beam. The energy resolution of the instrument was ~0.7%, and the relative error of a single measurement, including the statistical error of the counting channel and the error in the measurement of the primary current, did not exceed ±6%.

We present in this paper the results of an investigation of the energy spectrum of the electrons released in collisions between $H^+$ ions and Ar atoms at a colliding-particle energy 20 keV and in the electron energy interval 5–300 eV. In the entire electron energy interval, the distance between the successive measurement points was smaller than the energy resolution of the instrument. At each value of the analyzer voltage, we made usually 3–4 four single measurements, the statistics of each measurement being $10^3$–$10^4$ counts. The argon pressure in the collision chamber was $1.5 \times 10^{-4}$ mm Hg, and the pressure of the residual gas was $\sim 6 \times 10^{-6}$ mm Hg. The measurement results are expressed in terms of the absolute magnitude of the differential electron-production cross section per unit solid angle $d\Omega$ and per unit electron energy interval $dE_e$.

## MEASUREMENT RESULTS AND DISCUSSION

Figure 1 shows the complete spectrum of the electrons released in collisions between $H^+$ ions and Ar atoms, for the entire investigated electron energy range 5–300 eV. The values of the cross sections $d^2\sigma/dE_e d\Omega$ and electron energies $E_e$ are drawn in a logarithmic

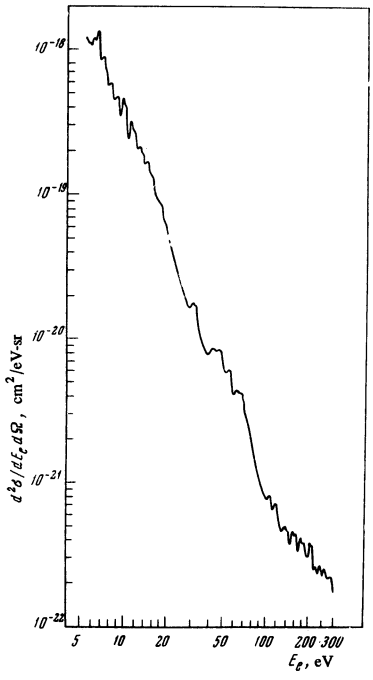


FIG. 1. Energy spectrum of electron released in collisions of $H^+$ ions with Ar atoms. Collision energy T = 20 keV. Electron emission angle $\theta$ = 54.5°. Ordinates – differential electron-production cross sections, abscissas – electron energy.

scale. Just as in the case of collisions between $Ar^+$ ions and Ar atoms[1], it is possible to separate in the electron energy distribution a continuous spectrum and a superimposed structure. As seen from the plot, the cross sections corresponding to the continuous part of the spectrum (the "base") decrease sharply with increasing electron energy, from $\sim 10^{-18}$ to $\sim 10^{-22}$ cm$^2$/eV-sr. The "base" may be due either to direct ionization or to a superposition of the bases of auto-ionization peaks. The peak-superposition effect is apparently more clearly pronounced at high electron energies, as is evidenced by the change in the slope of the energy-distribution curve in the high-energy part of the spectrum.

For a detailed study of the structure part of the curve, we divided the entire electron spectrum into individual sections and separated three main groups of lines corresponding to the three main types of auto-ionization transitions connected with the excitation of the M, $L_{23}$ and $L_1$ shells of argon. During the course of the discussion of the results we shall compare, where possible, our present data with the known analogous data obtained by others.

## 1. Transitions Connected with M-shell Excitation

To identify the numerous spectral lines connected in the low-energy region (5—16 eV) with excitation of the M shell of Ar, we determined the energies of the auto-ionization states of argon. We used a calculation method[6,7] based on the relation

$$E = E_\infty - 13{,}6Z^2/(n-\mu)^2, \qquad (1)$$

where E is the energy of the auto-ionization state, $E_\infty$ is the limit of the series, Z is the charge of the atomic remainder, n is the principal quantum number, and $\mu$ is the quantum defect.

In the determination of the quantum defect we used the following empirical regularities established by Samson,[6] which are well satisfied in the case of Ne[7]:

1. The quantum defect for the state connected with excitation of the s electron of the inert-gas atom is approximately equal to the quantum defect of the analogous state of the alkali-metal atom that follows it in the periodic system. For example the configuration $3s3p^64p$ of Ar I corresponds to the configuration $3s^23p^64p$ of K I. In the case of more complicated configurations connected with simultaneous excitation of 3s and 3p electrons, we have used the values of the quantum defect for the corresponding configuration of the Ca atom.

2. On going over to a heavier inert-gas atom (say from Ne to Ar), the quantum defect increases by unity. This regularity was used to determine the energy of the states of the configuration $3p^44s4p$. The values of the quantum defect for analogous Ne states were taken from the paper of Edwards and Rudd[7].

Calculations of this kind enabled us to determine the energies of various auto-ionization transitions, which could then be compared with the positions of the experimentally observed peaks and thus identify the electron spectral lines[1)].

The table lists the energies of certain initial auto-ionization states calculated from formula (1) and corrected on the basis of the experimental data on the spectral line positions connected with the decays of these states[2)]. For convenience, we use in the table the state designations adopted for the LS coupling. Whenever some configuration was characterized by an intermediate coupling, we used the rules of the transition to the limit[9].

Figure 2 shows a section of the spectrum in the electron energy band $E_e$ = 5—9 eV. Two intense line groups can be separated on the curve, at $E_e$ = 5.85—5.95 eV and $E_e$ = 6.25—6.35 eV. A sharply pronounced peak at 6.21 eV was observed also in[3,4]. The spectral lines observed in this energy interval are due to the transitions $3s3p^54p-3p^4$ and $3s3p^54s4p-3p^44p$. An interesting feature of the latter is that the final state of the produced ion is excited, and consequently auto-ionization transitions of this kind should be accompanied by optical transitions. In this case one should expect emission of the lines corresponding to the transitions 3s—4p and 3d—4p in the visible region of the spectrum ($\lambda$ = 3000—6000 Å). The excitation cross sections of the auto-ionization states $3s3p^54s4p$ in the case investigated by us can reach values on the order of $10^{-19}$ cm$^2$.

Another feature of the transitions considered in the table is connected with the fact that the excitation of certain auto-ionization states, for example the state $3s3p^5(^3P)4p(^4P)$, is accompanied by spin flip and by the

[1)] Because of space limitations, we are unable to present a table with all the spectral lines identified.

[2)] The final-state energies were determined from the spectroscopic tables [8].

| Auto-ionization state | E, eV | Auto-ionization state | E, eV |
|---|---|---|---|
| $3s3p^5(^3P)4p(^4D)$ | 48.96±0.05 | $3s3p^5(^1P)4s(^2P)4p(^2D)$ | 45.5±0.1 |
| $3s3p^5(^3P)4p(^2D)$ | 49.13±0.05 | $3s3p^5(^1P)4s(^2P)4p(^2P)$ | 45.9±0.1 |
| $3s3p^5(^3P)4p(^4P)$ | 49.23±0.05 | $3s3p^5(^1P)4s(^2P)4p(^2D)$ | 46.8±0.1 |
| $3s3p^5(^3P)4p(^2P)$ | 49.33±0.05 | $3s3p^5(^1P)4s(^2P)4p(^1P)$ | 46.9±0.1 |
| $3s3p^5(^1P)4p(^2D)$ | 52.75±0.05 | $3s3p^5(^1P)4s(^2P)4p(^1S)$ | 47.6±0.1 |
| $3s3p^5(^1P)4p(^2P)$ | 52.92±0.05 | $3p^4(^3P)4s(^4P)4p(^3P)$ | 28.2±0.3 |
| $3s3p^5(^1P)4p(^2S)$ | 53.55±0.05 | $3p^4(^3P)4s(^2P)4p(^3P)$ | 28.7±0.3 |
| $3s3p^5(^3P)4s(^4P)4p(^3P)$ | 41.84±0.05 | $3p^4(^3P)4s(^2P)4p(^1P)$ | 29.7±0.3 |
| $3s3p^5(^3P)4s(^2P)4p(^3D)$ | 41.84±0.05 | $3p^4(^1D)4s(^2D)4p(^3P)$ | 29.9±0.3 |
| $3s3p^5(^3P)4s(^2P)4p(^3P)$ | 42.21±0.05 | $3p^4(^1D)4s(^2D)4p(^1P)$ | 31.0±0.3 |

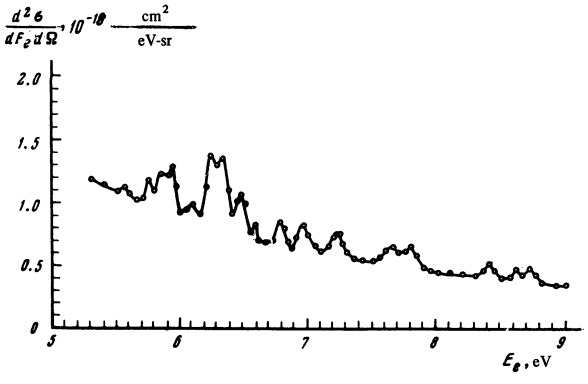


FIG. 2. Energy spectrum of electrons in the electron-energy interval $E_e$ = 5 - 9 eV. Resolution 0.07 eV.

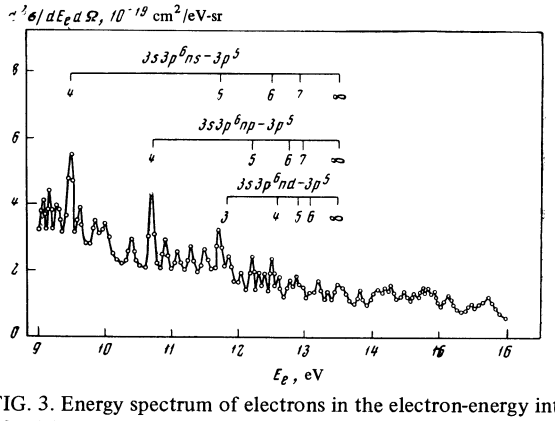


FIG. 3. Energy spectrum of electrons in the electron-energy interval $E_e$ = 9 - 16 eV. Resolution 0.1 eV. The positions of the lines corresponding to the transitions $3s3p^6\,nl - 3p^5$ are indicated on the top.

change of the multiplicity. Edwards and Rudd[7], who investigated the auto-ionization states of Ne, observed similar effects in collisions of Ne atoms with $He^+$ and $Ne^+$ ions, but not in the case of the $H^+$-Ne pair. They advanced the hypothesis that the main cause of formation of multiplet states is intense electron exchange that takes place in interactions of multielectron systems. A possible explanation of the excitation of multiplet states in the case of the $H^+$-Ar system is violation of the LS coupling and the appreciable mixing of Ar states with identical total angular momentum J, due to the spin-orbit interaction.

The electron energy spectrum shown in Fig. 3 pertains to the interval $E_e$ = 9—16 eV. In this energy region there are also intense peaks corresponding to electron energies 9.50, 10.75, 11.80, 12.20, and 12.55 eV. These peaks are connected with the excitation of auto-ionization states. The energies of some of the states—members of the indicated series—were determined in[6,10] from electron photoabsorption and inelastic scattering. The energies determined by us for the states pertaining to the first terms of the series $3s3p^6np$ are in good agreement with Samson's data[6], and the energy of the $3s3p^6 3d(^1D)$ state agrees well with the data of Simpson et al.[10] In the case of the state $3s3p^6 4s(^1S)$, the agreement between our data (25 and 26 eV) with the data of[10] (25.8 eV) is less satisfactory. The positions of the maxima corresponding to the excitation of the auto-ionization states belonging to the series $3s3p^6ns$, $3s3p^6np$, and $3s3p^6nd$ are noted in Fig. 3 in the form of vertical lines in the upper part of the figure.

The most significant among the other auto-ionization transitions in this electron energy region are the transitions $3s3p^5 4s4p - 3p^4 4s$ and $3p^4 4s4p - 3p^5$. The first of them leads to the formation of an excited state of the argon ion with subsequent emission of Ar II resonant lines.

The structure part of the spectrum obtained by us (Figs. 2 and 3) can be used to estimate the true widths of the spectral lines and consequently the lifetimes of the auto-ionization states produced in interactions between protons and argon atoms. To this end, we determined the apparatus function of the instrument and used the reduction rules considered in the paper by Bracewell[11]. As shown by control experiments with an auxiliary electron source[5], the apparatus function is described with a sufficient degree of accuracy by a Gaussian distribution. The reduction was carried out for the most intense and best resolved lines (e.g., the line at $E_e$ = 10.75 eV (Fig. 3)). Estimates have shown that the lifetimes of the auto-ionization states connected with the excitation of the M shell of Ar amount to $\sim 10^{-14}$ sec.

## 2. Transitions Connected with Excitation of the $L_{23}$ Subshell

Figure 4 shows a section of the electron spectrum in the energy interval $E_e$ = 100— 240 eV. The cross sections for the production of electrons in this energy region are small (on the order of $10^{-22}$ cm²/eV-sr), and the structure of the spectrum is due to various kinds of Auger transitions with filling of the vacancy in the $L_{23}$ subshells of argon. Transitions of the $L_{23}$ - MM type were investigated earlier[3,12,13] for the case when Ar atoms collide with high-energy protons

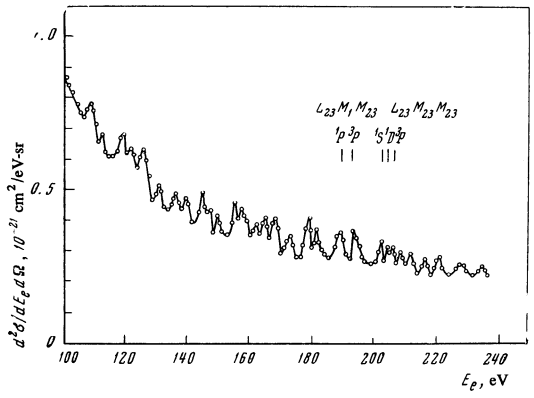


FIG. 4. Electron energy spectrum in the electron energy interval $E_e$ = = 100 - 240 eV. Resolution 1.5 eV. On the top are indicated the positions of the lines corresponding to Auger transitions in the neutral Ar atom, as given in [12].

and with photons. The peaks observed in the present investigation at energies 179, 189, 202, 204, 206, and 208 eV, which can be ascribed respectively to the transitions $L_{23} - M_1M_1(^1S)$, $L_{23} - M_1M_{23}(^1P)$, $L_{23} - M_1M_{23}(^3P)$, $L_{23} - M_{23}M_{23}(^1S)$, $L_{23} - M_{23}M_{23}(^1D)$, $L_2 - M_{23}M_{23}(^3P)$, and $L_2 - M_{23}M_{23}(^3P)$ agree well in their positions with the data of these references. The energies of some of the indicated transitions, as given by Carlson and Krause[12], are marked by vertical lines in the upper part of Fig. 4. In[14], where they investigated the excitation of the $L_1$ subshell of Ar by electron impact, they also observed electrons with energy $E_e$ =227 eV, due to the transition $2p^53s^23p^64s - 2p^63s^23p^5$. The peak seen on Fig. 4 at $E_e$ =2.28 eV can obviously be ascribed to the same transition. As can be seen from Fig. 4, however, the number of identified lines constitutes only a small fraction of all the lines observed in the energy interval 100—240 eV.

To obtain more detailed information on the nature of the lines of this section of the spectrum, we start from the following assumptions:

1. The lines corresponding to electron energies higher than the energies of the transitions $L_{23} - MM$ (>208 eV) are due to excitation of auto-ionization states in which the 2p electron of the argon does not go off to the continuum, but goes over to an excited optical level, for example, states of the type $2p^53s^23p^64s$. Auger transitions from similar initial states can take place both with participation of M and N electrons ($2p^53s^23p^64s - 2p^63s^23p^5$), both with participation of two M electrons ($2p^53s^23p^64s - 2p^63s^23p^44s$).

2. At energies $E_e$ smaller than the energies of the $L_{23} - MM$ transitions (< 179 eV), the observed lines are due to Auger transitions in which three electrons take part, wherein one of the electrons occupies a vacancy in the internal shell of the atom, the second goes off to the continuum, and the third goes over to an excited optical level, for example the transitions $2p^53s^23p^6 - 2p^63s^23p^34s$.

3. Auger transitions are possible not only in the neutral atom (i.e., from the states $2p^53s^23p^6$), but also in an ionized atom, say as a result of the "jolting" process (i.e., from $2p^53s^23p^5$ states).

In the calculation of the energy of the Auger transitions we used the data of[14] on the energies of the ground auto-ionization states of argon $2p^53s^23p^6$ and $2p^53s^23p^5$. The energies of the final states produced as the result of the Auger transition were determined from the spectroscopic tables[8]. In the case of configurations of the type $2p^53s^23p^64s$ ($2p^53s^23p^54s$) it was assumed that the binding energy of the 4s electron is approximately equal to its binding energy in the potassium atom (ion).

Starting from the results of the calculations, and taking into account the rather appreciable width of the peaks obtained in this energy region, we can assume that most of them represent superpositions of a large number of unresolved narrow peaks corresponding to separate Auger transitions. Thus, for example, the peaks at the energies 179, 189, and 193 eV may be connected not only with transitions in the neutral atom ($L_{23} - MM$), but also with transitions in the ionized argon atom ($L_{23}M - MM$).

The identification made in this manner indicates the presence of the following groups of Auger transitions connected with excitation of the $L_{23}$ subshell of Ar following interaction with protons:

1. Transitions from initial states $2p^53s^23p^6$.
2. Transitions from initial states $2p^53s^23p^5$.
3. Transitions from initial states of the type $2p^53s^22p^64s$, $2p^53s^23p^54s$, in which the 2p electron occupies an excited optical level.
4. Transitions of the type $2p^53s^23p^6 - 2p^63s^22p^34s$ with three electrons taking part, one of which goes over to an excited optical level.

### 3. Transitions connected with excitation of $L_1$ Subshell

Figure 5 shows the section of the spectrum obtained by us, pertaining to the high-energy part of the electron energy range, $E_e$ =240—300 eV. The plot shows clearly nine peaks corresponding to the energies 240, 247, 255, 260, 267, 273.5, 280, 288, and 292 eV. The appearance of electrons having these energies may be due to direct Auger transitions, which cause one of the outer 3s and 3p electrons to fill a vacancy in the $L_1$ subshell of Ar, while the other goes off to the continuum. The spectrum of the electrons released from the

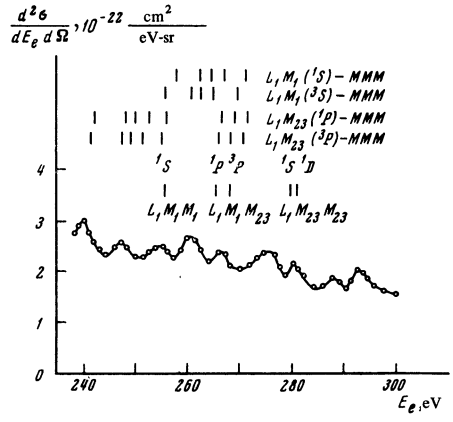


FIG. 5. Energy spectrum of electrons in the electron-energy interval $E_e$ = 240 - 300 eV. Resolution 2 eV. The lines in the upper half of the figure correspond to Auger transitions in Ar and $Ar^+$, according to the data of [14].

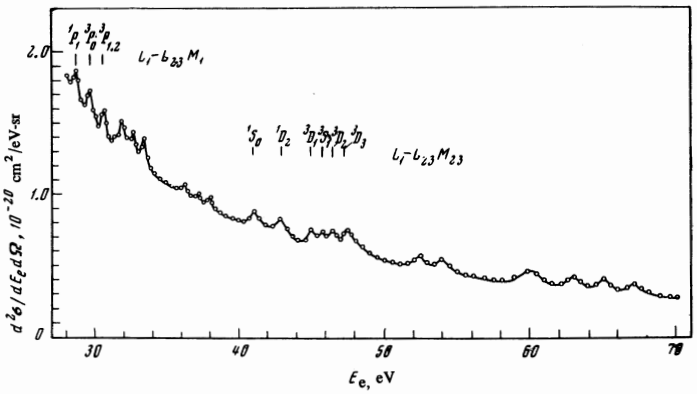


FIG. 6. Energy spectrum of electrons in the electron energy interval $E_e$ = 28 – 70 eV. Resolution 0.3 eV. On the top are indicated the positions of the lines corresponding to Koster-Kronig transitions in the neutral Ar atom according to [14].

Ar atoms by electron impact in this energy interval was investigated in[14]. The positions of the lines of the neutral (transitions $L_1$ – MM) and ionized argon atom (transitions $L_1M$ – MMM), as given in[14], are marked by the vertical lines in the upper part of the figure.

In the calculations of the energies of the Auger electrons, we used for the energy of the $2s2p^63s^23p^6$ configuration the value 326.5 eV determined in[14]. In the case of configurations of the $2s2p^63s^23p^64s$ ($2s2p^63s^23p^54s$) type it was assumed that the binding energy of the 4s electron is approximately equal to its binding energy in the potassium atom (ion), and in the case of the $2s2p^63s^23p^5$ configuration the energy determined from the formula

$$E(2s2p^63s^23p^5) \approx E(2s2p^63s^23p^6) + E(3p, \text{K II}),$$

where E(3p, K II) is the binding energy of the 3p electron in the $K^+$ ion. More accurate values of the energies of such states, obtained with allowance for the positions of the experimental peaks, turned out to be $E(2s2p^63s^23p^64s)$ = 321.9 ± 0.8 eV, $E(2s2p^63s^23p^64p)$ = 323.8 ± 0.8 eV, $E(2s2p^63s^23p^63d)$ = 324.2 ± 0.8 eV, and $E(2s2p^63s^23p^54s)$ = 345.5 ± 0.8 eV.

The peaks at 255, 267, and 280 eV, which are apparently due principally to the transitions $L_1 - M_1M_1(^1S)$, $L_1 - M_1M_{23}(^1P, ^3P)$, and $L_1 - M_{23}M_{23}(^1S, ^1D)$, agree in their positions with the data of[14]. The remaining lines in the interval $E_e$ = 240–280 eV pertain to the transitions $L_1M$ – MMM in the ionized Ar atom and to transitions in which three electrons take part, analogous to those considered in the preceding section. The peaks at higher energies are due to Auger transitions from initial states in which the 2s electron occupies an excited optical level.

The section of the spectrum corresponding to the electron energies $E_e$ = 28–70 eV is shown in Fig. 6.

The nature of many electron spectral lines in the energy range 28–70 eV was explained in[14]. These lines are connected with Auger transitions of the Koster-Kronig type, in which the vacancy in the $L_1$ shell is filled by a 2p electron, and one of the M-shell electrons goes off to the continuum. The probability of the Koster-Kronig transitions is usually much larger than the probability of the direct Auger transitions to the $L_1$ vacancy. Thus, for example, according to the data of[14], the summary probability of the Koster-Kronig transitions in the case of Ar is approximately 30 times larger than that of direct Auger transitions. The positions of the most intense lines investigated in[14] and connected with the transitions $L_1 - L_{23}M_1$ and $L_1 - L_{23}M_{23}$ are shown in the upper part of Fig. 6. As seen from the figure, there is good agreement between the data of[14] and our present data.

In addition to the lines corresponding to the indicated transitions and considered in[14], we have observed two other groups of lines in the energy intervals $E_e$ = 52 – 54 and $E_e$ = 60 – 68 eV. The first of them is apparently connected with the transitions $2s2p^63s^23p^64s - 2p^53s^23p^54s$, and the other with the transitions $2s2p^63s^23p^54s - 2p^53s^23p^5$ in the ionized Ar atom. The peaks in the energy range 31 – 34 eV are apparently due to the transitions $2s2p^63s^23p^5 - 2p^53s^23p^4$. In view of the lack of data on the energies of the final states, it is difficult to identify the lines in this section of the spectrum.

On the whole, the electron spectrum connected with Auger transitions to the $L_1$ vacancy is characterized by the same features as the spectrum connected with the excitation of the $L_{23}$ subshell of Ar. These features consist in the fact that besides the ordinary Auger transitions of the L – MM type, transitions LM – MMM are possible in the ionized Ar atom, and also transitions from the initial states, in which the electron goes over from the internal shell to the excited optical level, as well as three-electron Auger transitions. A more complete understanding of the character of the observed spectra calls for a special theoretical study. In particular, it is of interest to determine the mechanism of formation of an atom ionized in the inner and outer shells by proton impact. A possible cause of a phenomenon of this kind may be the "jolt" effect resulting from a sudden removal of an electron from the inner shell of the atom. It is not excluded that a definite role is played also by the charge-exchange of a proton interacting with an internal electron of the atom. It is of definite interest also to clarify the question of the probability of Auger transitions in which three electrons take part, since this phenomenon has never been investigated theoretically so far.

## CONCLUSION

Our investigation of the energy distribution of the electrons released from argon atoms has yielded new information concerning the auto-ionization argon states

excited by proton impact. Particular interest attaches to the observed possibility of formation of $3s3p^5 4s4p$ states, the decay of which produces excited $Ar^+$ ions. It is not excluded that the excitation of such states may be reflected in the character of the processes occurring in an argon laser[15].

We observed for the first time spectral lines connected with the excitation of the 2s electron of Ar in collisions of atomic particles. The investigations have shown that Auger transitions accompanied by filling of the vacancy in the inner shell of the atom are highly variegated and can occur with the participation of either two electrons or three electrons.

It can be hoped that the results of this investigation will stimulate further theoretical investigations of the questions connected with ionization of inner shells of the atom and will contribute to a more complete understanding of the character of the inelastic processes occurring upon collision of atomic particles.

The authors are deeply grateful to N. V. Fedorenko for valuable advice and continuous interest in the work, and to V. M. Dukel'skiĭ for a discussion of the results.

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Translated by J. G. Adashko
5