INVESTIGATION OF THE LINE WIDTHS OF DISCRETE ENERGY LOSSES IN Ar^+—Ar *COLLISIONS*

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A method for investigation of inelastic energy loss spectra in atomic collisions is developed, based on measurement of the kinetic energies of the two atomic particles after the collision. The method can be used to determine the natural widths and to make more refined determinations of the discrete loss line energies for Ar^+—Ar collisions previously studied by a different method[1]. The line widths and energies are measured at an initial kinetic energy of 25 keV and initial scattering angle of the incident ions of 16°; various elementary processes of changing the charge state of the two particles could occur in these cases. Under the collision conditions studied, three loss lines R_I, R_{II} and R_{III} are excited; the line energies are of the order of hundreds of eV and the line widths amount to several dozen eV. It is shown that the R_{II} and R_{III} lines can be regarded as being the result of simultaneous excitation of lowest-energy line R_I and respectively of one ($R_{I-II} \approx 258$ eV) or two (R_{I-II} and $R_{II-III} \approx 272$ eV) levels with a natural width ~65 eV. Excitation of each of the levels (R_{I-II}, R_{II-III}) is accompanied by additional removal on the average of 1.5 electrons from the colliding particles. Estimates show that at small internuclear distances the system consisting of two atomic particles produced in the collision, the Ar_2^+ quasimolecule, should be in a highly excited state. The discrete inelastic energy losses can be attributed to excitation and decay of auto-ionization states produced in the collisions. The R_I line is ascribed to autoionization transitions in the outer (M) shells of the particles accompanied by excitation of a large number of M electrons. The R_{I-II} and R_{II-III} levels are ascribed to auto-ionization arising on removal of electrons from the inner (L) shells. For explanation of the level widths and energies it is suggested that the model of L-vacancy formation for Ar^+—Ar proposed by Fano and Lichten[7,10] be extended by taking into account auto-ionization transitions in the quasimolecule, i.e., before the separation of the particles.

INTRODUCTION

INVESTIGATIONS of the elementary act of atomic collisions at energies of the order of several keV by the coincidence method have shown that in collisions connected with the interaction of the electron shells the spectrum of the inelastic energy loss has a discrete character and consists of individual lines[1-4]. The energy of the heretofore investigated lines lies in the interval from several dozen to several hundred eV. Measurements of the energies of the discrete lines were carried out for the cases of collisions of ions and atoms of different inert gases, and also at different initial kinetic energies, distances of shortest approach between nuclei, and accompanied by various processes of the charge state of the particles. At the same time, another no less important problem, that of the width of the lines of inelastic energy losses, has so far remained uninvestigated.

The existing estimates of the line widths are crude and contradictory to a considerable degree. Thus, according to the estimate given in one of our first papers[1], the natural line width in Ar^+—Ar collisions does not exceed several dozen eV. According to later data[3], the line width in such collisions amounts to ~200 eV, although the authors of[3] do note the low accuracy of their width measurements. The lack of reliable data on the line width is connected with the features of the experimental procedure used to study the spectra of inelastic losses. In[1-4], the inelastic-loss width was determined by simultaneously measuring the scattering angle ϑ of the incoming particle and the emission angle φ of the recoil particle—the partners of the same elementary collision act (the angles ϑ and φ are reckoned from the direction of the momentum of the incoming particle prior to the collision). Such a procedure, however, has an essential shortcoming in that the energy resolution during the measurement of the elastic loss depends not only on the angular resolution of the collimator used to separate the scattered particles from the recoil particles, but also on the thermal motion of the target-gas atoms. The strong influence of the thermal motion on the resolution is due to the fact that the momenta of the thermal motion add up vectorially with the momenta of the particles after the collision. By way of an example we note that for Ar^+—Ar collisions at an initial energy $T_0 = 25$ keV and an incoming-particle scattering angle $\vartheta = 15°$, the energy resolution amounted to ~140 eV in our investigations.

The main purpose of the present investigation was to set up experiments that ensure an appreciable increase in the accuracy of the widths and line shapes of inelastic energy loss. The problem was solved by changing over to a direct measurement of the elastic loss R as a difference between the initial kinetic energy of the incoming particle T_0 and the sum of the kinetic energies of the fast scattered particle T_1 and the recoil particle T_2 after the collision:

$$R = T_0 - (T_1 + T_2). \quad (1)$$

It is obvious that in such a method of measuring R the thermal motion will cause only a negligibly small error, equal to the kinetic energy of the gas atoms at room temperature (~ 0.03 eV).

EXPERIMENTAL PROCEDURE

The experimental setup was described earlier[5], and had much in common with the setup used in our earlier investigations. A narrow beam of fast particles was passed through the gas. The two colliding particles were separated by two collimators. The positions of these collimators relative to the direction of the primary beam has made it possible to determine the scattering angles ϑ and φ of the two particles. The particles passing through the collimators were analyzed with respect to energy and with respect to charge in electrostatic analyzers, and were registered with detectors; the pulses from the detector outputs were fed to a delayed-coincidence system. We recall that in our earlier investigations we determined only the final charge states of the particles, for which we used magnetic analyzers with low resolution. Thus, all the previous capabilities of the setup (simultaneous measurement of the angles ϑ and φ and of the final charge states m and n of both particles) were retained in the modified procedure. In addition, it was possible to determine simultaneously the kinetic energies T_1 and T_2 of both particles after the collision.

The employed electrostatic energy analyzers were parallel-plate capacitors with a particle entrance angle 45°. Such analyzers, as is well known, have a first-order focusing with respect to the entrance angle. The distance between the entrance and exit slits of the analyzers was 400 mm. The slit widths could be varied without breaking the vacuum in a range 0—400 μ with accuracy ± 4 μ, making it possible to reduce the proper energy resolution of the analyzers $\Delta T/T$ to practically 5×10^{-5} (relative to the peak widths at half the height). The total width of the apparatus function of the setup depended, naturally, on the energy resolution of both analyzers, and also on the energy scatter in the primary ion beam. We used in the investigation a high-frequency ion source, and the energy scatter of the ions produced by it was measured to be ~7 eV (by scatter or width we shall henceforth have in mind the width of the distribution at half the height). The total width of the apparatus function in most experiments of the present investigation was 19 eV.

It is clear from the foregoing that the use of an analysis of the kinetic energies of both particles after the collision has made it possible to increase greatly the resolution in the investigation of the inelastic-loss energy spectra. It should be noted, however, that the use of such a procedure entails a considerable complication of the measurement technique.

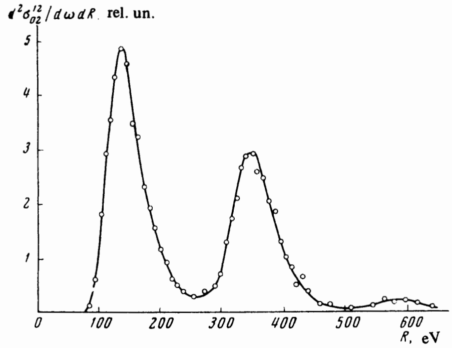

FIG. 1. Dependence of the differential cross section of the process $Ar^+ + Ar \rightarrow Ar^{2+} + Ar^{2+} + 3e$ on the inelastic energy loss. Incident-particle energy $T_0 = 25$ keV. Incident-particle scattering angle $\vartheta = 16°$.

MEASUREMENT RESULTS AND DISCUSSION

1. Energies and Widths of Discrete Loss Lines

By analyzing the kinetic energies of the atomic particles, we investigated the inelastic-loss spectra in Ar^+—Ar collisions. The initial energy of the incident particles was 25 keV, and their scattering angle was $\vartheta = 16°$. The choice of the pair, of the initial energy, and of the scattering angle is connected with the fact that under these conditions in our earlier investigations we observed simultaneous excitation of free inelastic energy-loss lines with nearly equal probabilities.

The inelastic-loss spectra were investigated in various processes wherein the charge states of the particles were changed. By way of an example, Fig. 1 shows the inelastic-loss spectrum obtained in the study of the process $Ar^+ + Ar \rightarrow Ar^{2+} + Ar^{2+} + 3e$ (abbreviated designation 1022). The spectrum consists of three well-resolved lines with energies 141, 350, and 588 eV. The line widths on Fig. 1 amount to 62, 82, and 85 eV respectively. These widths $(\Delta R_l)_e$, obtained as a result of the experiment, are made up of the natural line width ΔR_l and the width of the apparatus function of the setup $(\Delta R)_a$. By approximating the obtained line shape and the apparatus-function shape by Gaussian distributions, it is easy to obtain a relation for the natural line width

$$\Delta R_l = \sqrt{(\Delta R_l)_e^2 - (\Delta R)_a^2}. \quad (2)$$

For the present example, the natural line widths calculated in accordance with (2) are 62, 79, 82 eV. We see that at the obtained energy resolution, the apparatus broadening of the lines is quite insignificant and the widths of the experimental contours are determined almost completely by the natural line widths.

The measured energies R_l and the natural widths ΔR_l of the inelastic loss lines determined from (2) for different processes (10 mn) in which the charge states are changed are listed in Table I. The table lists also the errors in the determination of the energies σ_{R_l} and of the widths $\sigma_{\Delta R_l}$, corresponding to a confidence probability 68%.

The inelastic-loss spectra for all the investigated 10 mn processes are similar to the spectrum shown in Fig. 1, differing from the latter only in the probability of excitation of the different lines. In none of the cases was a finer structure observed at the resolution employed in the investigation (19 eV). The investigated lines have a somewhat asymmetrical shape, characterized by a steeper leading (low-energy) front. This asymmetry is most noticeable in the excitation of the line R_I, corresponding to the smallest energy loss, although in all cases the asymmetry is quite low.

2. Inelastic Energy Loss in Various Elementary Processes

In our earlier investigations[1,2] it was established that the energy R_l of the inelastic-loss lines changes on going from one process to another, but the changes in the loss are connected principally with the change in the energy consumed in ionization Σu_i. This means that the "excess" lost energy, i.e., the difference between the total inelastic losses and the energy consumed in ionization, $R_l^* = R_l - \Sigma u_i$, remains practically constant for the different processes, when compared with the total inelastic energy R_l, which changes in a wide range, depending on the process. Such an approach has made it possible to consider instead of the total inelastic loss R_l the excess inelastic loss R_l^*, and to separate three lines whose energies in first approximation do not depend on the scheme of the process. The scatter in the values of R_l^* for different processes do not exceed the experimental errors[1,2]. In the present investigation it was established that the changes of the values of R_l^* on going from one elementary process to another have a systematic character. Figure 2 shows the variation of the line energy R_l^* as a function of the ionization energy, as determined in the present measurements by the scheme of the elementary process. We see that although for all three lines the function $R_l^*(\Sigma u_i)$ is relatively weak, the observed changes in R_l^* exceed the experimental errors. Attention is called to the different dependence of the excess loss on the scheme of the process for different lines. Whereas the energy of the first line R_I^* increases with increasing ionization loss, in the two other lines the situation is reversed. In one of our earlier investigations[6] it was shown that the line R_I^* differs from R_{II}^* and R_{III}^* in one other respect. Whereas in the excitation of the lines R_{II}^* and R_{III} there exist approximately the same correlation between the final charges of the particles m and n, there is practically no such correlation in the excitation of the line R_I^*.

3. Connection Between the Different Inelastic-loss Lines

The energy of the R_I^* line shows that this line can be connected only with excitation and ionization processes occurring in the outer shells of the colliding particles. As follows from the present results and the earlier one[1-4], the probability of realizing these processes in collisions with intersection of the electron shells is quite high, since under the conditions of such collisions no cases were observed without excitation of any of the inelastic-loss lines (R_I, R_{II}, R_{III}) and without ionization. Therefore, it is quite natural to assume, as was done in[7], where the discrete energy loss for Ar^+—Ar was interpreted, that the excitation of the outer shell occurs whenever the collision is connected with a closer approach of the nuclei and the inelastic-loss lines R_{II} and R_{III} are observed. It

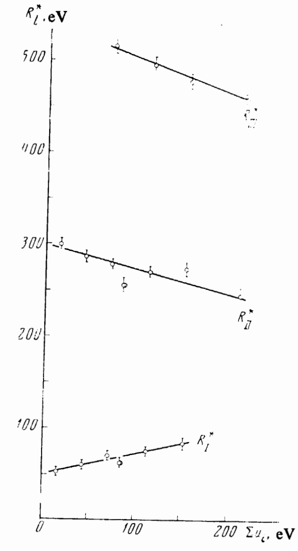

FIG. 2

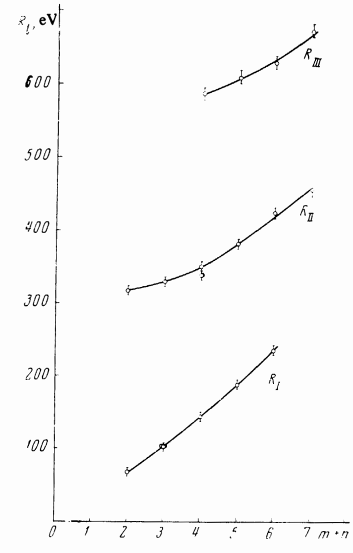

FIG. 3

FIG. 2. Dependence of the excess inelastic energy loss (R^*_I, R^*_{II}, and R^*_{III}) on the ionization loss in the excitation of three discrete loss lines.

FIG. 3. Dependence of the energy of the discrete inelastic-loss lines on the total final charge of the atomic particles.

Table I.

10mn	Σu_i	R_I	R_{II}	R_{III}	ΔR_I	ΔR_{II}	ΔR_{III}	σ_{R_I}	$\sigma_{R_{II}}$	$\sigma_{R_{III}}$	$\sigma_{\Delta R_I}$	$\sigma_{R\Delta_{II}}$	$\sigma_{\Delta R_{III}}$
1011	16	68	317		39			4	6		5		
1021	43	102	330		49	82		4	6		5	6	
1022	71	141	350	588	62	79	82	4	5	8	4	5	12
1031	84	146	340		62	73		5	6		7	5	
1013	84	146	340		69	73		4	5		7	5	
1032	112	187	383	609	80	85	108	5	6	8	6	4	7
1033	152	236	426	631		94	102	6	7	8		5	5
1043	212		458	676		110	109		9	9		11	7

follows from this assumption that the lines R_{II} and R_{III} can be regarded not as independent, but as a result of a simultaneous excitation of the line R_I, which is connected with processes on the outer shell, and respectively one (R_{I-II}) or two (R_{I-II} and R_{II-III}) additional levels. Measurements performed in[2] and in the present work have shown that in Ar^+—Ar collisions (T_0 = 25—50 keV), on going over from excitation of the R_I line to the excitation of the R_{II} line, and from R_{II} to R_{III}, the average number of the electrons removed by the collision increases approximately by 1.5. It can be assumed that an increase of the number of removed electrons is connected with autoionization decay of the additional levels R_{I-II} and R_{II-III}. This means that the energy of the additional level $R_{l-l'}$ cannot be determined from the energies R_l and $R_{l'}$ measured for the same elementary process.

The energy of the level $R_{l-l'}$ can be determined with account taken of the change of the average number of removed electrons by representing the data of Table I in the form of a dependence of the line energy R_l on the total final charge of the particles m + n, as is done in Fig. 3. Using the data of Fig. 3, we can determine the energy $R_{l-l'}$ with the aid of the relation

$$R_{l-l'} = R_{l'}(m+n+1.5) - R_l(m+n). \qquad (3)$$

Similar reasoning leads to the possibility of determining also the widths of the additional levels $R_{l-l'}$.

To this end, it is necessary to plot the dependence of the line width ΔR_l on the total final charge of the particles m + n, on the basis of which the width of the additional level $\Delta R_{l-l'}$ can be determined from the relation

$$\Delta R_{l-l'} = \sqrt{[\Delta R_{l'}(m+n+1.5)]^2 - [\Delta R_l(m+n)]^2}. \quad (4)$$

The energies and widths of the additional levels $R_{l-l'}$ were calculated with the aid of relations (3) and (4) for different values of the total final charge m + n; the results are shown in Figs. 4a and b. We see that both the energies and the widths of both additional levels R_{I-II} and R_{II-III} are quite close to each other. It follows from Figs. 4a and b that the energies and widths of the additional levels are not very sensitive to changes of the final charge state of the particle. Thus, when m + n changes from 2 to 5—6 (corresponding to a change in the ionization loss by more than 100 eV), the energy of the level R_{I-II} change in the ionization loss by more than 100 eV), the energy of the level R_{I-II} changes by only 20 eV. As to the level R_{II-III}, the scatter in its energies, as determined for different values of m + n, is smaller than the energy error, which amounts to ~6 eV.

4. Mechanism Producing the Discrete Energy Loss

The collisions investigated in the present study were relatively slow—relative velocity of the particle was approximately one sixth the velocity of the electron in the hydrogen atom as given by Bohr. At such a velocity, according to the usual notion of quasiadiabatic interaction the probability of the inelastic processes that call for an energy consumption on the order of tens and hundreds of eV should be very low. At the same time, the experimental data[1-4] show that at the investigated approach distances r, on the order of the dimensions of the shells, the probability of excitation of the lines of the inelastic losses is close to unity, and the total excitation cross section exceeds 10^{-17} cm^2 even for the lines R_{II} and R_{III}. One can therefore assume that as a result of the shifting of the terms of the Ar^+—Ar system when the nuclei come close together the real transition energy is appreciably smaller than the inelastic loss, which amounts to the difference between the energies of the initial and final states of the system at $r \to \infty$. The appreciable probability of the large inelastic losses may also be connected with the presence of a large number of electrons from which transitions are possible.

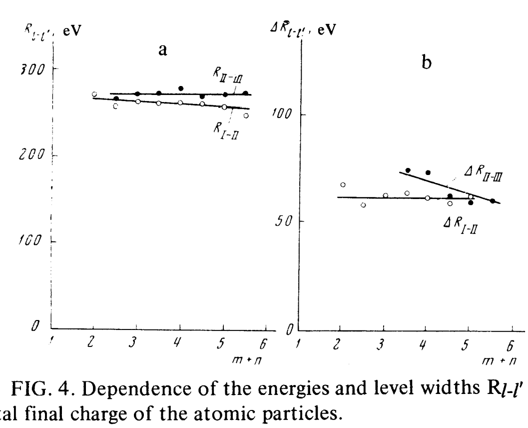

FIG. 4. Dependence of the energies and level widths $R_{l-l'}$ on the total final charge of the atomic particles.

It is clear that processes wherein electrons are knocked out and acquire appreciable kinetic energy are impossible at the investigated low velocities of the atomic particles. Apparently the explanation of the discrete inelastic energy losses can be based only on the consideration of intermediate autoionization states which are produced upon collision. As follows from an analysis of the excess loss, the autoionization levels should lie high above the limit of the continuous spectrum, and their energy should exceed greatly the binding energies of the outer-shell electrons of the colliding nuclei.

Unfortunately, there is still no universal inelastic-collision model suitable for the description of the currents of the levels. Nonetheless, following the publication of the first results of the studies of the discrete loss, several approaches to their interpretation were proposed, which to be sure had as a rule a qualitative and descriptive character. We consider below the possibility of using these approaches for the analysis of the results obtained in the present investigation.

a) The line R_I. According to the phenomenological description given in Sec. 3, each of the inelastic-loss lines R_l includes the processes of excitation of the outer shells of both particles. In pure form, these processes become manifest when the line R_I is excited. Multiple ionization observed upon excitation of the line R_I indicates that a large number of outer-shell electrons take part in the excitation processes. In addition, as shown in one of our earlier papers[8], the number of removed electrons is well described by the statistical relations. It can be assumed in this connection that the statistical approach is the most rational method of describing the considered processes in the outer shells.

The statistical character of the formation of autoionization states and of the process of removal of electrons during the decay of these states was used in a recent paper[9] devoted to the explanation of the first inelastic-loss line R_I^* in Ar^+—Ar collisions. The authors of[9] regard the line R_I^* as a result of the excitation of a certain number of outer electrons. According to the calculation[9], the excitation energies of any pair of outer electrons suffices to remove one of these electrons, if the other electron goes over at the same time to a lower free level. Therefore the most probable autoionization scheme is treated in[9] as a series of paired Auger transitions, as a result of which half of the excited electrons is removed. The weak dependence of the excess loss R_I^* on the scheme of the process is attributed[9] to the smaller growth of the excitation potentials are compared with the ionization potentials with increasing number of excited and hence removed electrons. The values of the excess losses R_I^* calculated in[9] for different 10 mn processes are in good agreement with the experimental data. Table II lists the results of the present measurements and the calculation of[9]. Good agreement is observed for all 10 mn processes with the exception of 1011. In the investigated conditions, the relative probability of the 1011 process is lower, and the discrepancy between the experimental and calculated values of R_I^* can be contributed to the fact that this process is connected to a

Table II.

10 mn	Σu_i	R^*_{I} th	R^* exp
1011	16	22	52
1022	71	72	70
1013	84	58	62
1031	84	58	62
1033	152	90	84

considerable degree with excitation and autoionization schemes that differ from the paired Auger-transition scheme.

It should be noted that the construction of the detailed model of the autoionization state, connected with simultaneous excitation of a large number of electrons from interacting particles, is hardly possible at the present status of the theory of atomic collisions. The model of collective excitation of electrons, considered in[10], is not yet sufficiently well developed to be able to carry out a quantitative calculation of the spectrum of the excited states of a system of colliding particles. Therefore, even though the model of collective excitations, as noted in our earlier papers, agrees qualitatively with the experimental data on inelastic atomic collisions, it is impossible to carry out at present a more detailed comparison of this model with experiment. The collective-excitation model was successfully used for the analysis of the simplest cases—the interaction of electrons and photons with atoms. Theoretical investigations[10-12] have shown that calculations using the model of collective excitations ensure in many cases much more accurate description of the experimental data[13,14] than the usual calculation which takes into account only the single-electron transition[15].

b) The lines R_{II} and R_{III}. The experimental data show sufficiently clearly that the appearance of inelastic-loss lines R_{II} and R_{III} is connected with the excitation of the internal shells. This is indicated both by the magnitude of the excess energies R^*_{II} and R_{III}, and by the fact that the probability of excitation of these lines increases sharply when a closest-approach distance of 0.25 Å, corresponding to the crossing of the L shells, is reached. The simplest mechanism of participation of the internal-shell electrons and atomic excitation is apparently removal of electrons and formation of vacancies. Such an excitation mechanism of autoionization states has been known for a long time, and was discussed already in our first paper devoted to discrete inelastic losses[11]. The possibility of formation of $L_{\mathrm{II,III}}$ vacancies in Ar^+—Ar collisions not only by removal of electrons but also by transition of a 2p electron to higher levels, was considered in[7,16]. According to the model developed in[7,16], the transition is the result of a shift of the lower field levels (molecular orbitals) of the quasimolecule Ar_2^+ when the internuclear distance changes, and the intersection with the outer unfilled levels. The energy of formation of the $L_{\mathrm{II,III}}$ vacancy in the Ar atom amounts to 249 eV when a 2p electron goes over to the continuous spectrum, and is somewhat lower in the case of a transition to an unfilled level of the atom. According to Lichten[16], this energy should not depend in first approximation on the state of the outer shell. In accordance with such a model, the energies $R_{\mathrm{I-II}}$ and $R_{\mathrm{II-III}}$ of the levels separated in this investigation should equal the energy of formation of the $L_{\mathrm{II-III}}$ vacancy. As seen from Fig. 4a, the energies of the levels $R_{\mathrm{I-II}}$ and $R_{\mathrm{II-III}}$ are actually not very sensitive to changes of the final charge states of the nuclei. However, the experimental values of the energies of these levels, namely 258 ± 8 and 272 ± 6 eV, themselves greatly exceed the energy of formation of the $L_{\mathrm{II,III}}$ vacancy. In addition, in accordance with the present measurements, the levels $R_{\mathrm{I-II}}$ and $R_{\mathrm{II-III}}$ have an appreciable width, which amounts to ~ 65 eV for each level. One can hardly attribute such a width of the levels $R_{\mathrm{I-III}}$ and $R_{\mathrm{II-III}}$ to the transitions of a 2p electron to different unfilled levels in the atomic particle, particularly if it is recognized that the energies of the levels $R_{\mathrm{I-II}}$ and $R_{\mathrm{II-III}}$ themselves change by not more than 23 and 7 eV when the charge states of the particles vary in a wide range.

Within the framework of the models considered here, one might attempt to improve the agreement with experiment by assuming that in addition to the $L_{\mathrm{II,III}}$ vacancies there can occur also L_{I} vacancies, which call for an energy loss of 326 eV, although the formation of L vacancies as a result of a shift in an intersection of terms at the investigated internuclear approach distances is less probable. However, from the numerical values of the energies of formation of the L_{I} and $L_{\mathrm{II,III}}$ vacancies it follows that were it possible to obtain in this case an average vacancy-formation energy in agreement with the energies of the levels $R_{\mathrm{I-II}}$ and $R_{\mathrm{II-III}}$, then the total width of the transition from the levels 2s and 2p would greatly exceed the experimentally observed 65 eV.

Summarizing the foregoing, we can state that the assumption that vacancies are produced in the internal shell makes it possible to explain qualitatively the presence of three discrete inelastic-loss lines and the existence of characteristic internuclear distances in Ar^+—Ar collisions. The qualitative agreement of the discrete losses and the vacancy-formation energies for isolated particles was noted also earlier, in the analysis of the results of measurements for Ar^+—Ar, in our earlier paper[11], and also in the work of Fano and Lichten[7,16]. At the same time, discrepancies are observed in the comparison of the more exact results obtained in the present paper with the vacancy-formation energies. Nonetheless, in our opinion, the presence of the indicated discrepancies is no reason for rejecting the explanation of the inelastic losses on the basis of the assumed vacancy formation in the internal shells. These discrepancies only indicate that the vacancy-formation model, in the form presented by Fano and Lichten, is too schematic and calls for further development.

c) Possible directions in which the molecular-orbit-shift model can be modified. According to Lichten[16] autoionization states with $L_{\mathrm{II,III}}$ vacancies have a lifetime exceeding the collision time, and their decay occurs when the colliding particles have already ceased to interact, and the autoionization transitions in the external shells have terminated. Therefore, in the existing form, this model can explain quantitatively the inelastic losses in atomic collisions only on the basis of transition energies in isolated atomic particles. The quasimolecular system produced during the time of the collision is essentially considered only in order to show how excitation can occur as a result of collision. However, the clarification of the problem of the ratio of the lifetime of the autoionization state to the collision time calls, in our opinion, for a more careful analysis. The calculation has shown[17], for example, that the probability of the Auger process in a quasimolecular system produced when a multiply charged ion collides with an atom can increase strongly, reaching 10^{15}—10^{16} sec^{-1}. On the other hand, the conclusion arrived at in[16], that the $L_{II,III}$ vacancies produced in collisions have a large lifetime, is based only on considerations that are valid for an isolated atom. Yet in a quasimolecular system there appear factors that are capable of increasing the probability of the autoionization transition. These factors include, for example, the increased overlap of the wave functions of the electrons of the L and M shells compared with the isolated atom, and also the increase of the total number of outer electrons with which the $L_{II,III}$ vacancies can interact in the quasimolecule. It is therefore not excluded that the activation transition can occur with high probability *also* within a collision time that amounts to $r_L/v \sim 10^{-16}$ sec even for L-shell interaction, and to $r_M/v \sim 10^{-15}$ sec for M-shell interactions (r_L and r_M are the radii of the L and M shells, and v is the velocity of the relative motion of the atomic particles).

When account is taken of the possibility of autoionization transitions in the quasimolecule, large inelastic energy losses can be attributed to factors other than electron transitions upon intersection of molecular orbitals (MO). An important role can be played also by autoionization states of the quasimolecule itself, which exist only at low internuclear distances r. Within the framework of the MO model, the reason for the appearance of such autoionization states of the quasimolecule is the shift of the MO of the unfilled shells of Ar (3d, 4s, 4p), as a result of which the Ar^+—Ar system (which goes over into the Kr atom in the limiting case of $r \to 0$) turns out to be, at small values of r, in a state with a large number of vacancies in the internal shells. The excitation energy can be quite large in this case, and the probability of formation of such states should be close to unity, since the shift of the unfilled MO "downward" depends not on the probability of the transition at any particular intersection of the MO, but on the internuclear distance. We note that the occurrence of autoionization states with a large total excitation energy when the atomic particles come closer together follows not only from the MO analysis, but can also be explained on the basis of more general considerations. The binding energy of all the electrons E in a multi-electron atom, according to the Thomas-Fermi model, is connected with the charge Z of the nucleus by the relation $E = 20.8Z^{7/3}$ eV. In accordance with this relation, the difference between the electron binding energy in the two Ar atoms and in the unified system as $r \to 0$ (the Kr atom) is $A_{Kr} - 2E_{Ar} \approx 3E_{Ar} \approx 50$ keV. This means that the produced system can be strongly excited when the internuclear distances between the atomic particles become smaller.

The assumed possibility of autoionization transitions prior to the decay of the quasimolecule allows us to supplement the explanation of the present results. The appreciable widths of the observed R_{I-II} and R_{II-III} levels, and the difference between their energies and the energies of the transitions between the levels of the isolated Ar atom, can be connected with the change of the energy of the autoionization transitions with changing internuclear distance r. In such a case, the observed energy and line width will be determined by the dependence of the probability of the autoionization transition on r. This assumption could also explain the results of measurements of inelastic losses for other pairs[2], according to which the energies of the inelastic-loss lines depend on the relative velocity of the colliding particles. In addition, the energies and widths of the lines can be connected also with the decay of the autoionization states of another type, which were indicated above, namely states existing only in the quasimolecule.

Of course, the assumption that the autoionization states decay during the time of collision does not mean that only the properties of the quasimolecule appear in all the characteristics of the inelastic collision. As a result of the statistical character of the decay of the autoionization states in time, a certain fraction of the particles may turn out to be excited also after the particles move apart, and in such cases the properties of the isolated atoms or ions come into play (for example, in the form of characteristic groups of Auger electrons or emission lines). The ratio of the collision time and of the decay probability of the excited states in each concrete case will determine which properties (those of a system or those of isolated particles) will become predominantly manifest in the collisions.

The results of a study of the energy spectra of the electrons produced in atomic collisions are regarded in some papers as a confirmation of the Fano-Lichten model. Thus, an investigation of Ar^+—Ar collisions[18,19] revealed narrow electron lines, with energy ~200 eV, corresponding to Auger transitions occurring in the filling of $L_{II,III}$ vacancies. However, the total cross section for the production of such electrons was not measured, and it is impossible to state that each case of excitation of the lines of inelastic losses with high energy (R_{II} and R_{III}) corresponds to the emission of Auger electrons from the isolated particles. Moreover, it turns out that in Ar^+—Ar collisions there are produced, besides electrons with discrete energies, also electrons having continuous energy spectrum that extends to above 200 eV. The total cross section for the production of electrons having a continuous energy distribution greatly exceeds the cross section for the production of electrons with discrete energy values. Therefore the results of a study of the energy spectra of the electrons is in much better agreement with the assumed decay of the autoionization states predominantly in the quasimolecule. Within the framework of the latter assumption, it is easy to explain the occurrence of electrons with a continuous energy spectrum, since the transition energy depends on the internuclear distance at which the autoionization takes place. On the other hand, discrete lines of Auger electrons in the spectrum are a manifestation of the presence of a certain number of cases in which the excited states do not have time to decay within the collision time, and the transitions occur already in the isolated atoms or ions that move apart. In addition to this cause, there may also be another cause for the appearance of electrons with continuous energy spectra. A continuous electron energy distribution can be observed in the case when the decay of the excited state is accompanied by the emission of not one but several (even two) electrons, even if the total excitation energy has in this case a fixed value.

Thus, our analysis shows that the inelastic energy loss in Ar^+—Ar collisions can be qualitatively explained on the basis of the notion of autoionization transitions in the quasimolecule Ar_2^+. To describe the losses connected with simultaneous excitation of many electrons of outer shells, the most fruitful approach is the statistical one. The inelastic losses corresponding to excitation of individual internal-shell electrons can be described on the basis of the vacancy-production assumption. However, concrete models using these assumptions require further development. An analysis of the experimental data points to the possible ways of such a development, namely the investigation of autoionization transitions in the quasimolecule, i.e., prior to the moving apart of the particles, and also allowance for the vacancies that result from the approach of the nuclei and exist at small internuclear distances.

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[1] V. V. Afrosimov, Yu. S. Gordeev, M. N. Panov, N. V. Fedorenko, Zh. Tekh. Fiz. **34**, 1624 (1964) [Sov. Phys.-Tech. Phys. **9**, 1256 (1965)].

[2] V. V. Avrosimov, Yu. S. Gordeev, M. N. Panov, and N. V. Fedorenko, ibid. **36**, 123 (1966) [**11**, 89 (1966)].

[3] Q. C. Kessel and E. Everhart, Phys. Rev. **146**, 16 (1966).

[4] Q. C. Kessel, M. P. McCaughey, and E. Everhart, Phys. Rev. **153**, 57 (1967).

[5] V. V. Afrosimov, Yu. S. Gordeev, M. N. Panov, and N. V. Fedorenko, Zh. Tekh. Fiz. **34**, 1613 (1964) [Sov. Phys.-Tech. Phys. **9**, 1248 (1965)].

[6] V. V. Afrosimov, Yu. S. Gordeev, A. M. Polyanskiĭ, and A. P. Shergin, ZhETF Pis. Red. **6**, 461 (1967) [JETP Lett. **6**, 3 (1967)].

[7] U. Fano and W. Lichten, Phys. Rev. Lett. **14**, 627 (1965).

[8] V. V. Afrosimov, Yu. S. Gordeev, M. N. Panov, and N. V. Fedorenko, Zh. Tekh. Fiz. **34**, 1637 (1964) [Sov. Phys.-Tech. Phys. **9**, 1265 (1965)].

[9] L. M. Kishinevskiĭ and É. S. Parilis, ibid. **38**, 760 (1968) [**13**, 570 (1968)].

[10] M. Ya. Amus'ya, ibid. **36**, 1409 (1966) [**11**, 1053 (1969)] M. Ya. Amusia, Abstr. V. Int. Conf. Phys. of Electronic and Atomic Collisions, Leningrad, (1967), p. 97.

[11] W. Brandt and S. Lundqvist, J. Quantitative Spectroscopy and Radiative Transfer **7**, 411 (1967).

[12] M. Ya. Amusia, N. A. Cherepliov, and S. J. Sheftel, Abstr. V. Int. Conf. Phys. of Electronic and Atomic Collisions, Leningrad, (1967), p. 130.

[13] A. P. Lukirskiĭ, I. A. Brytov, and T. M. Zimkina, Opt. Spektrosk. **17**, 438 (1964); D. L. Ederer, Phys. Rev. Lett. **13**, 760 (1964).

[14] V. V. Afrosimov, Yu. S. Gordeev, V. M. Lavros, and S. G. Shemelinin, Zh. Eksp. Teor. Fiz. **55**, 1569 (1968) [Sov. Phys.-JETP **28**, 821 (1969)].

[15] J. W. Cooper, Phys. Rev. Lett. **13**, 762 (1964).

[16] W. Lichten, Phys. Rev. **164**, 131 (1967).

[17] L. M. Kishinevskiĭ and É. S. Parilis, Zh. Eksp. Teor. Fiz. **55**, 1932 (1968) [Sov. Phys.-JETP **28**, 1020 (1969)].

[18] M. E. Rudd, T. Jorgensen, and D. J. Volz, Phys. Rev. **151**, 28 (1966).

[19] G. N. Ogurtsov, I. P. Flaks, S. V. Avakyan, and N. V. Fedorenko, ZhETF Pis. Red. **8**, 541 (1968) [JETP Lett. **8**, 330 (1968)].

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