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Inelastic Collisions of Slow Alkali or Alkaline-earth Ions with Cadmium Atoms

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Detailed studies have been made of the excitation processes in collisions of ions of Li^+, Na^+, K^+, Rb^+, Cs^+, Mg^+, Ca^+, Sr^+, and Ba^+ with cadmium atoms in the energy range from threshold to 1000 eV. Cross sections have been determined for excitation of a number of lines of the target atoms, the incident ions, and magnesium atoms. It is established that the excitation functions of spectral lines of cadmium change appreciably with the mass and type of the incident ion: the excitation functions of the lines of resonance-doublet components of alkaline-earth ions coincide except for barium. In interaction of alkali-metal ions with cadmium atoms, only the target atoms are excited, and in the case of collisions of Cd atoms with the ions Ca^+, Sr^+, and Ba^+, both the target atoms and the incident ions are excited. The reaction $Mg^+ + Cd$ occurs with a high probability simultaneously in three inelastic channels leading to excitation of Cd and Mg atoms and Mg^+ ions. The excitation efficiency of the spectral lines is analyzed and it is suggested that the observed fine-structure peaks in the optical excitation functions of the lines are the result of the quantum-mechanical interaction of two or more closely spaced inelastic reaction channels.

A number of recent articles have reported observation of fine structure (oscillations) in the excitation functions of spectral lines arising in collision of low-energy ions with atoms. Structure in the excitation curves of lines appears most distinctly in the processes $He^+ + He$,[1] $He^+ + H_2$,[2] $Na^+ + Ne$,[3] $Zn^+ + Cd$,[4] and others. Discovery of the causes of oscillations in the cross sections for these inelastic processes is the subject of further intense experimental and theoretical investigation. Thus, Tolk et al.[5] in studying the same spectral lines of He I, Ne I, and Ne II in the processes $He^+ + Ne$ and $Ne^+ + He$ has observed a completely different nature of the energy dependence of the excitation cross sections for the spectral lines: in the first case a number of fine-structure peaks are observed which are regular on an inverse-velocity scale, and in the second case the excitation functions of the lines have a monotonic nature. A similar pattern is observed also in the reactions $Zn^+ + Zn$ and $Cd^+ + Zn$.[6]

A qualitative theoretical explanation of the structure in the excitation functions of the 3^1S and 3^3S levels of the helium atom has been given by Rosenthal[7] on the basis of assumption of interference of two vacant states of the quasimolecule coherently excited in a $He^+ + He$ collision. A further development of this theory has been given by Ankudinov et al.,[8] who obtained expressions for the cross sections for inelastic processes with inclusion of the nonadiabatic interaction at large internuclear distances of two or more inelastic channels, and expressions for the amplitude, period, and phase of the oscillations in the total excitation cross sections.

The present work was undertaken in order to obtain detailed information on the nature of the excitation of energy levels in collisions of the slow ions Li^+, Na^+, K^+, Cs^+, Mg^+, Ca^+, Sr^+, Ba^+, and Rb^+ with cadmium atoms. The choice of these pairs of interacting particles permitted tracing of the various inelastic channels leading to excitation of spectral lines, the excitation efficiency for spectral lines of the target atoms by ions of various configurations and masses, and also the conditions for appearance of oscillations in the excitation functions of spectral lines.

APPARATUS AND METHOD OF MEASUREMENT

In a preceding article[4] we described in detail apparatus designed to study excitation processes in collision of low-energy ions with atoms. It consists of an ion source, a vapor-filled cell, spectral apparatus, and equipment for photoelectric detection of the radiation.

In performing the present experiments, we made design changes in the individual parts of the equipment which substantially improved the experimental conditions. Improvements were made in the ion source, the system for acceleration and focusing of the ion beam, and the techniques for determination of the concentration of atoms in the vapor-filled cell.

In place of the ion source used previously,[4] we developed, tested, and put into use a highly versatile ion source which could operate in three modes: with electron bombardment, with surface ionization, and as a low-voltage discharge. The electrostatic cylindrical deflecting system was replaced by an ion-optical system with axial symmetry. Finally, the temperature of the cadmium-filled tube was determined not by a thermocouple but by a highly accurate mercury thermometer.

As a result of these improvements, the stability of the ion current and the length of service of the source in operation with various metals were increased; an increase was obtained in the ion current in the collision chamber, which varied from 0.8 to 5 μA, depending on the ion mass and the mode of operation of the source; the volt-ampere characteristics of the ion current at the collector were substantially improved (current saturation set in at an accelerating voltage of 10–20 V); and the spread in ion velocities was decreased to a half-width of 1–5 eV.

It is well known that in study of inelastic processes in collision of ions with atoms, it is necessary to choose the optimal experimental conditions and to exclude extraneous factors which distort the shape of the measured curve. For this reason control experiments were carried out during the work for each pair of interacting particles.

In the first place, we determined the range of cadmium vapor pressure in the cell and ion current in the beam for which single collisions were observed. It turned out that in the region studied by us (pressures of 1×10^{-3}–9×10^{-3} torr, currents of 0.5–5 μA) a linear dependence of spectral line intensity on vapor pressure and ion current is preserved.

In the second place, we observed that for certain potentials on the electrodes of the ion-optical system, secondary elements from the electrode surfaces enter the collision chamber. For this reason the potential on the focusing electrode was set below the potential of the accelerating electrode, and the ion collector was lengthened substantially. This precaution completely removed the possibility of appearance of electrons at the place of observation of the radiation.

In the third place, for the combination Mg^+-Cd we made measurements of the excitation functions of the lines both in the apparatus described previously[4] and in the improved apparatus. Comparison of the curves obtained showed that the excitation functions of the lines agreed within experimental error.

In the fourth place, we have shown[9] that for slow ion-atom collisions the effect of transfer of excitation energy of ions to the target atoms is important. Since the ions Ca^+, Sr^+, and Ba^+ have low-lying metastable levels, measurements of the excitation functions in these cases were made with the ion source operated in the surface-ionization mode or with electron bombardment, with the ionizing-electron energy being chosen below the threshold for excitation of the metastable level.

Absolute excitation cross sections for the lines were determined by three methods: photoelectric comparison of the intensities of the spectral lines with radiation from a standard lamp, from the known cross sections for electron excitation of cadmium lines, and by use of a light-detecting system calibrated in absolute units. The spectral-line excitation cross sections measured by these three independent methods agreed within 50–100%.

EXPERIMENTAL RESULTS

Analysis of the spectrum of radiation arising from interaction of alkali or alkaline-earth ions with cadmium atoms showed that, depending on the type of incident particle, the reactions studied proceed by one, two, or

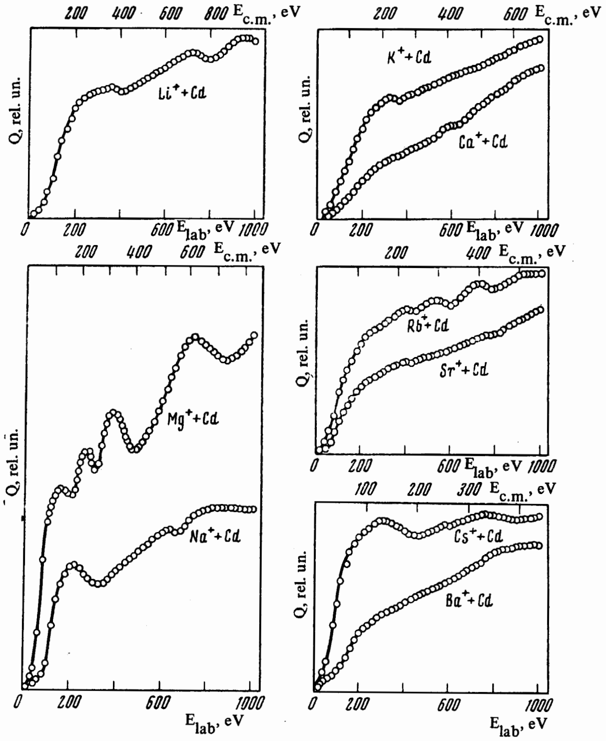

FIG. 1. Excitation functions for cadmium resonance lines 3261 Å (5^1S_0-5^3P_1) in various processes.

Cross sections and excitation thresholds for spectral lines of Cd, Mg, Mg^+, Ca^+, Sr^+, and Ba^+

Element	λ, Å	ΔE_{lab}^{theor}, eV	ΔE_{lab}^{exp}, eV	$Q \cdot 10^{19}$ cm² for E_{lab} = 600 eV
		Mg^++ Cd		
Cd	3261	4.6	10	—
	3466—68	8.9	20	3.2
	3610—14	8.9	20	9.5
	4678	7.7	10	1.6
	4800	7.7	10	5.0
	5086	7.7	10	7.5
Mg	2852	6.9	10	110
	3829—38	8.8	80	8.0
	4571	4.9	80	1.8
	5167—84	7,8	80	6.6
Mg^+	2795	5.4	8	84
	2803	5.35	8	40
	2928—36	10,5	12	2.5
		Sr^++ Cd		
Cd	3261	6.72	20	-
Sr^+	4078	5,4	10	50
	4216	5.2	10	30
		Li^+ + Cd		
Cd	3261	3.9	10	—
		Na^++ Cd		
Cd	3261	4.56	20	—

Element	λ, Å	ΔE_{lab}^{theor}, eV	ΔE_{lab}^{exp}, eV	$Q \cdot 10^{19}$ cm² for E_{lab} = 600 eV
		Ca^+ + Cd		
Cd	3261	5.13	20	—
	3466—68	10	80	1.0
	3610—14	10	80	3.0
	4678	8.6	10	0.9
	4800	8.6	10	2,9
	5086	8.6	10	4.4
Ca^+	3159—81	9.5	10	1.8
	3706—37	8.7	20	2.1
	3934	4.25	8	94
	3968	4.2	8	45
		Ba^++ Cd		
Cd	3261	8.37	12	—
Ba^+	4554	6	8	25
	4934	5.5	8	16
		K^++ Cd		
Cd	3261	5.13	30	—
		Rb^++ Cd		
Cd	3261	6.65	15	—
		Cs^++ Cd		
Cd	3261	8.3	20	—

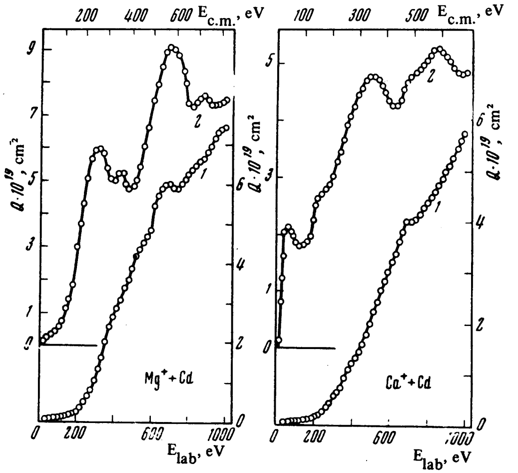

FIG. 2. Excitation cross sections for spectral lines of the cadmium atom as a function of energy: 1–3610Å (5^3P_2-5^3D_1), 2–5086Å (5^3P_2-6^3S_1).

three inelastic channels. The first channel leads to excitation of energy levels of the target atoms, the second to excitation of the incident ion, and the third to excitation of atoms produced as the result of charge exchange. It turned out that the first channel is open in all the reactions studied by us, that the first two channels appear simultaneously in collision of cadmium atoms with alkaline-earth ions, and finally that all three possible channels are observed only in the process Mg$^+$ + Cd.

Figures 1 to 5 and the table show the results of our investigation. The left and right coordinates in the plots give the excitation cross sections of the lines, and the abscissa scale at the bottom gives the kinetic energy of the incident ion, while the upper scale is the relative energy of the interacting particles. The curves obtained are the result of averaging 10–20 individual measurements.

In the first channel the cadmium resonance intercombination line λ = 3261 Å is excited most intensely. The excitation function plotted for this line in Fig. 1 shows that it depends in a complicated way on the atomic number and electron-shell configuration of the incident ion. As can be seen from the figure, in interaction of alkali-metal ions with atoms of cadmium, one or more fine-structure peaks is observed in the excitation function of the Cd resonance line, while in collisions of cadmium atoms with alkaline-earth ions the structure is absent except for the reaction Mg$^+$-Cd, in which the peaks appear very distinctly. On the other hand, collisions of ions which are close in mass (Mg$^+$ and Na$^+$, K$^+$ and Ca$^+$, Rb$^+$ and Sr$^+$, Cs$^+$ and Ba$^+$) but with a different outer electron shell lead to a completely different energy dependence of the excitation cross section of the cadmium resonance line.

In bombardment of Cd atoms by Mg$^+$ and Ca$^+$ ions, the lower triplet S and D levels are also strongly excited. The cross sections for excitation of lines from these levels[1] are shown in Fig. 2 as a function of energy. It should be noted that in the remaining cases the intensities of these same spectral lines of the cadmium atom were too low for their excitation functions to be measured.

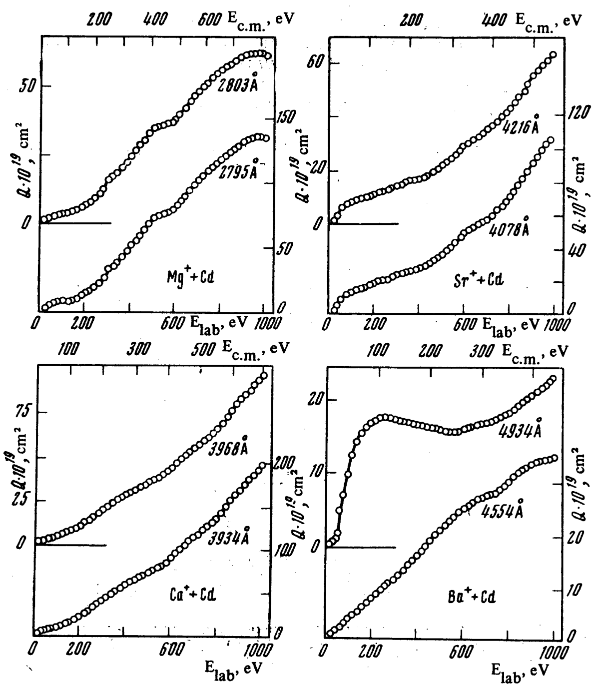

FIG. 3. Cross sections for excitation of resonance lines of alkaline-earth ions as a function of energy.

In contrast to the 3261 Å resonance line, structure appears distinctly in the excitation functions of the 5086 Å line (see Fig. 2) for excitation both by Mg$^+$ ions and Ca$^+$ ions. However, the rate of rise of the curve at threshold is greater in the case of Ca$^+$ + Cd, and the cross section at the peak for the reaction Mg$^+$ + Cd is almost twice the cross section for excitation of this same line by Ca$^+$ ions. The excitation functions of the 3610 Å line (Fig. 2) are similar in excitation by Mg$^+$ and Ca$^+$ ions: a weak rate of rise at threshold with a break in the 200–250 eV region, and in addition the absolute values of the excitation cross sections are very nearly the same.

In the second channel, in collisions of Mg$^+$, Ca$^+$, Sr$^+$, and Ba$^+$ ions with cadmium atoms, the resonance levels of these ions are most efficiently excited. Excitation cross sections of the corresponding resonance lines of alkaline-earth ions are shown in Fig. 3. As can be seen, for most levels an almost linear energy dependence is observed, with insignificant steps, and the excitation functions of the doublet components are identical in the case of Mg$^+$, Ca$^+$, and Sr$^+$. In contradiction to this, they differ strongly for Ba$^+$: while the strong component 4554 Å of Ba II shows an almost linear dependence of the cross section on energy, the rate of rise of the excitation function of the weak component 4934 Å of Ba II near threshold is appreciably higher and the curve reaches a peak at E_{lab} = 200 eV. The absolute cross section for excitation of the resonance lines, as can be seen from Fig. 3, is greatest for the Ca II line 3934 Å and amounts to $\sim 2 \times 10^{-17}$ cm^2, and is least for the Ba II line 4934 Å, amounting to $\sim 2 \times 10^{-18}$ cm^2 for E_{lab} = 1000 eV.

Other levels of alkaline-earth ions are efficiently excited only in light particles (Mg$^+$ and Ca$^+$). For ex-

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[1] The excitation functions of lines originating from the same upper level agree within experimental error.

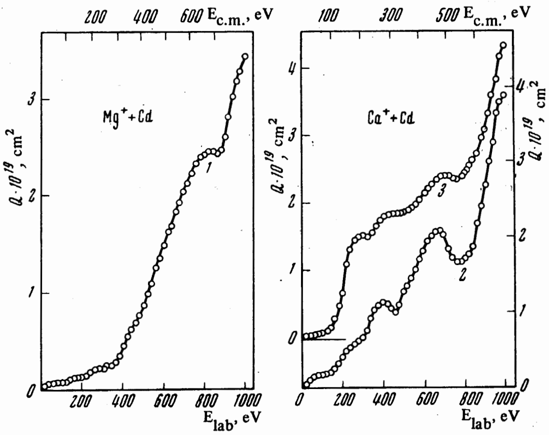

FIG. 4. Cross sections for excitation of spectral lines of magnesium and calcium ions as a function of energy: 1–2936Å ($3^2P_{3/2}$-$4^2S_{1/2}$); 2–3179Å ($4^2P_{3/2}$-$4^2D_{5/2}$); 3–3737Å ($4^2P_{3/2}$-$5^2S_{1/2}$).

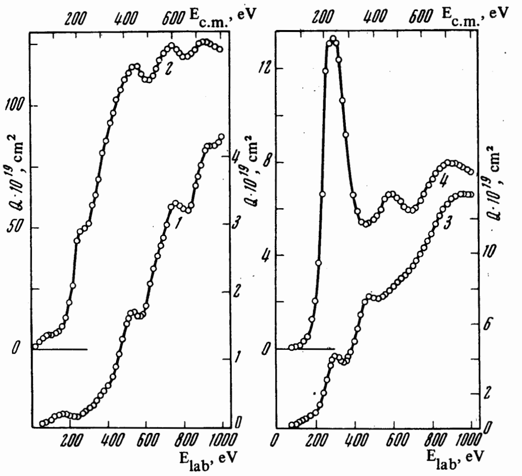

FIG. 5. Cross sections for excitation of spectral lines of the magnesium atom arising in the process Mg^+ + Cd, as a function of energy: 1–4571Å (3^1S_0-3^3P_1); 2–2852Å (3^1S_0-3^1P_1); 3–3838Å (3^3P_2-3^3D_3); 4–5184Å (3^3P_1-4^3S_1).

ample, in the excitation functions measured by us for the spectral lines of these ions: 2928/36 Å of Mg II, 3706 Å and 3737 Å of Ca II (see Fig. 4), an insignificant rise in the curve is observed at threshold, and then a sharp increase in the rate of rise of the cross section and appearance of a number of peaks which appear particularly clearly in the excitation function of the Ca II line 3179 Å.

In the case of interaction of Mg^+ ions with cadmium atoms, in addition to the two reaction channels discussed above, a third channel is also observed—charge exchange of the ions with transition to an excited state. In Fig. 5 we have shown excitation functions of the most intense spectral lines of the neutral magnesium atom—single and intercombination resonance lines and lines originating from the first excited triplet S and D levels. As can be seen, the energy dependence of the excitation cross sections for these lines is not monotonic, and there is a rather distinct structure in the energy region studied. The absolute value of the cross section for excitation of the Mg I singlet resonance line 2852 Å is close to the excitation cross section for the resonance lines of the magnesium ion, and those of the other lines to the cross sections for excitation of the Cd atom lines originating from similar upper S and D levels.

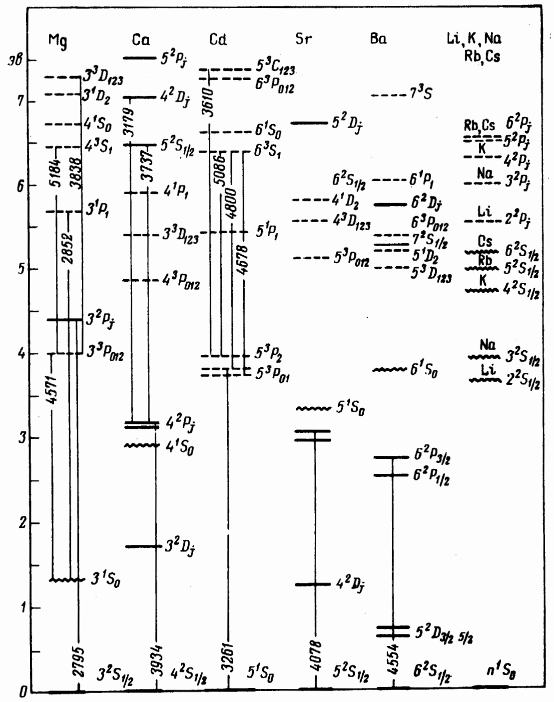

FIG. 6. Energy diagram of inelastic channels in collisions of alkali or alkaline-earth ions with cadmium atoms: solid lines—ion energy levels; dashed lines—neutral-atom energy levels; wavy lines—charge-exchange energy defects.

DISCUSSION OF RESULTS

The extensive experimental data obtained by us on excitation functions (Figs. 1–5) and excitation cross sections (the table) show that in most cases the determining factor in the probability for occurrence of a reaction by a given channel is the energy defect ΔE. Levels with a minimal energy defect are excited most efficiently. Furthermore, it turns out that when there are two or more levels closely spaced in energy, the characteristic fine-structure peaks appear in the excitation functions.

If we consider the absolute cross sections for excitation of spectral lines giving a cascade contribution (see the table), we can conclude that they do not play an important role in population of the initial levels (at least of the resonance levels). Therefore the excitation functions shown in the figure for lines almost completely reflect the nature of the excitation of the corresponding energy levels of the atoms and ions.

Proceeding from what has been said above, let us dwell in more detail on the results obtained by us. For this purpose we will use the energy scheme of the elastic channels, which are shown in Fig. 6. These channels arise in the following ways:

$$A^+ + Cd \rightarrow A^+ + Cd^* + \Delta E,$$
$$A^+ + Cd \rightarrow A^{+*} + Cd + \Delta E,$$
$$A^+ + Cd \rightarrow A^* + Cd^+ + \Delta E,$$
$$A^+ + Cd \rightarrow A + Cd^+ + \Delta E,$$

where A^+ is the incident ion, and ΔE is the energy defect of the corresponding process. The first three channels studied by us lead to excitation of the interacting particles; study of the fourth channel—simple asymmetric charge exchange—did not enter into the problem of the present experiment.

We will begin discussion of the processes studied by us with the first channel—excitation of cadmium atoms. As can be seen from Fig. 6, in collisions of alkali-metal ions with atoms of cadmium, excitation of the 5^3P_j level of Cd is energetically most favorable. However, competition is provided by the fourth channel—charge exchange to the normal state of alkaline-metal atoms to whose energy defects decrease in the transition from cesium to lithium, approaching the excitation energy of the cadmium resonance level. This competition obviously is also reflected in the energy dependence of the cross section for excitation of the cadmium resonance line 3261 Å (see Fig. 1) excited by alkali ions. This competition appears most strongly in the process $Na^+ + Cd$, for which the difference in the energy defects is least and amounts to only 0.05 eV. In just this case the sharpest peak is observed at the excitation threshold of the resonance line. Therefore we can suggest that the additional peaks in the excitation functions of the Cd line 3261 Å in excitation by alkali-metal ions appear as the result of competition (interference) of two inelastic channels which are close in energy—direct excitation of the level and charge exchange to the normal state of the alkali atom. A convincing confirmation of this suggestion would be the observation of oscillations in the charge-exchange functions, which should be in the opposite phase[3, 10] from the peaks in the excitation function of the cadmium resonance line.

The smooth behavior of the excitation function of the 3261 Å Cd resonance line in excitation by alkaline-earth ions is due, it appears to us, to the energy isolation of the initial level of the line. An exception is the process $Mg^+ + Cd$. If we turn to the inelastic channel scheme (Fig. 6), we see that in this case the 5^3P_j levels of Cd, the 3^3P_j levels of Mg, and the 3^2P_j levels of Mg^+ are located very close together. Hence we can assume that the clearly expressed oscillatory nature of the excitation function of the Cd line 3261 Å in the process $Mg^+ + Cd$ is the consequence of a quantum-mechanical interaction of the initial level of the line with the excited levels of the atom and ion of magnesium enumerated above. The idea of competition of two reaction channels for interpretation of the peaks and of the regularity in their appearance in the excitation functions of the lines in heavy-particle collisions was expressed for the first time by Bobashev.[3] However, in the case $Mg^+ + Cd$ we have succeeded in observing competition of three channels and, possibly, for this reason, the fine-structure peaks do not reveal a regular nature.

It should be noted that fine-structure peaks are absent in the excitation function of the cadmium resonance line in the process $Ba^+ + Cd$, in spite of the existence of a nearby charge-exchange channel (Fig. 6), which is 0.02 eV below the excitation threshold for the Cd resonance level. It is possible that in this case the interaction of the competing channels leads to a sharp peak in the charge-exchange function. An indirect confirmation of this idea is the fact that in the process $Zn^+ + Cd$[4] a sharp peak is observed at threshold in the excitation function of the 3076 Å Zn resonance line, which arises as the result of charge exchange, while the excitation function of the 3261 Å cadmium resonance line, whose energy defect is 0.2 eV higher, shows a smooth behavior.

The appearance of structure in the excitation functions of the 5086 and 3610 Å lines of cadmium also can be explained by interference of competing channels. Possible such channels are the initial 6^3S_1 level of Cd competing with the 4^3S_1 and 4^1S_0 levels of Mg, and the 6^3S_1 level of Cd competing with the $5^2S_{1/2}$ level of Ca II. This is indicated by the fact that some peaks in the excitation functions of the 5086 Å Cd line are in opposite phase to the peaks in the excitation functions of lines with energetically nearby levels (5184 Å of Mg I and 3737 Å of Ca II).

Let us turn now to discussion of the excitation functions of levels of the incident ions. Proceeding from the model proposed above of the interaction of two or more inelastic channels, we can easily understand the smooth behavior of the excitation curves of the resonance levels of the alkaline-earth ions (see Fig. 3). The fact is that the initial levels of these elements are strongly isolated energetically (Fig. 6), and therefore interference is absent and the excitation functions do not have singularities. The appearance of small steps in the excitation functions of the 2795 Å and 2803 Å lines of Mg II is apparently due to weak interaction of the initial levels of the 3^2P_j lines of Mg II with the 3^3P_j levels of Mg I and the 5^3P_j levels of Cd.

The difference in the behavior of the excitation functions for the components of the barium ion doublet 4554 and 4934 Å cannot be explained unambiguously at the present time. It is possible that this is due to the large difference (the greatest for the alkaline-earth elements) in the excitation energy defects of the initial sublevels of the barium ion (see the table).

Analysis of the excitation functions and the cross sections for excitation of resonance lines of ions of the alkaline-earth elements shows that in a certain range of energies the cross sections for excitation of the lines are proportional to the energy of the ion, and, furthermore, the following dependence of the cross sections on the ion energy E, mass m, and the energy defect of the process ΔE is observed approximately:

$$\sigma(E) \approx KE / m\Delta E^2,$$

where K is a constant equal to $\sim 6.6 \times 10^{18}$ amu-eV-cm^2.

Again, the resonance lines of the barium ion are an exception, their excitation cross sections being below the expected values. However, if we assume that this relation should be satisfied for the levels and not for the lines, it turns out that the excitation cross sections for the resonance levels of the ions Mg^+, Ca^+, and Sr^+ (taking into account the transition probabilities) are practically in agreement with the excitation cross sections of the lines, and the cross sections for excitation of the resonance sublevels of Ba^+ are substantially higher as a result of the high probability of transitions from the 6^2P_j levels to the 5^2D_j levels of the barium ion.[11]

Thus, the results obtained by us for the excitation functions of resonance lines of the alkaline-earth ions show that the well-known Massey criterion,[12] which is contradicted by the data of recent experimental studies,[1-6] is appropriate when there is an energetically well isolated channel for occurrence of a reaction involving interaction of heavy particles. An additional confirmation of this is the fact that the excitation functions of lines with energetically higher levels of the ions Mg^+ and Ca^+ (see Fig. 4) reveal a fine structure in the near-threshold energy region. It is just here that there are competing channels (see Fig. 6).

Competition of all three inelastic channels appears clearly in the interaction of Mg^+ ions with Cd atoms (see Fig. 6). Therefore efficient excitation occurs of target atoms, incident ions, and Mg atoms by means of charge exchange with transition to excited states. It is interesting also that the cross section for excitation of the Mg line 2852 Å is comparable in magnitude with the excitation cross section for the Mg II line 2795 Å, which has almost the same excitation energy. The cross sections for excitation of the Mg lines 5184 Å and 3838 Å is also close to those for the Cd lines 5086 Å and 3610 Å, whose excitation potentials are almost the same. Proceeding from this, we can consider as a reliably established fact that oscillations in excitation curves, as in the cases discussed above, appear as the result of the mutual influence of two or more inelastic channels of nearly the same energy.

Finally, it should be noted that a general feature of all channels which we have studied is the characteristic threshold behavior of the excitation cross sections: presence in the initial portions of the curves of a very weak rate of rise with a subsequent sharp rise in the excitation efficiency. This indicates that excitation at threshold is determined by two processes of different probability. In the region of the very weak rate of rise of the cross section, excitation of the levels occurs by means of the tunnel effect,[13] and not as the consequence of crossing (pseudocrossing) of terms of the quasimolecule. When the ions reach energies corresponding to the sharp break in the curve, on the other hand, transitions to the ground state occur according to the Landau–Zener scheme, whose probability greatly exceeds the probability of the tunnel effect. Therefore the small shift in the experimental excitation threshold of lines toward higher energies, observed in practically all cases (see the table), obviously must be assigned to insufficient sensitivity of the detecting equipment.

CONCLUSIONS

The results of our investigation can be reduced to the following:

1. In interaction of the low-energy ions studied with cadmium atoms the probability that the reaction occur by a given inelastic channel is determined by the energy defect of the reaction.
2. The observed structure (oscillations) in the excitation cross sections of lines is the result of the mutual influence of two or more inelastic channels with nearly the same energies.
3. For resonance levels of alkaline-earth ions a close to linear energy dependence of the excitation cross section near threshold has been established.
4. Transitions between final energy states of the interacting particles in the near-threshold region can occur both in the presence of crossings (pseudocrossings) of terms of the quasimolecule arising, and without crossing of terms, by means of the tunnel effect.

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