EXCITATION OF ATOMS IN SLOW ION-ATOMIC COLLISIONS

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We describe apparatus used to study the threshold behavior of the optical excitation functions of spectral lines of cadmium and zinc atoms, produced when slow zinc ions (0—1200 eV) interact with cadmium atoms, and we determined the absolute cross sections for the excitation of 13 spectral lines of these atoms. Fine-structure maxima were observed on most of the measured curves, and certain features of the excitation of atoms in slow ion-atom collisions were established. The results of the measurements are compared with the theoretical calculations by the Grizinski formula.

A large number of papers have been published recently on elastic and inelastic scattering of low-energy electrons by atoms and molecules. Much less attention has been paid to similar investigations of collisions of heavy particles[1]. This is due to difficulties of experimental character and to the theoretical evidence[2] that such processes have low efficiency at low energies. There exist at present, however, experimental data pointing to considerable cross sections of reactions between ions and atoms in the near-threshold energy region. In[3] it was found, in a study of collisions of He$^+$ ions with helium atoms, that the energy dependence of the excitation cross sections of the spectral lines of the helium atoms from threshold to 5 keV has a strongly pronounced oscillatory character. A similar picture was observed also in the case of interaction of alkali-metal ions with inert-gas atoms[4]. The large role played in the formation of excited particles in a plasma by collisions between low-energy ions and atoms is mentioned also in[5,6]. Considerable interest attaches therefore to a determination of the effective cross sections for the excitation of spectral lines of atoms in slow ion-atom collisions, in order to clarify the mechanism whereby excited particles are produced in different types of low-temperature plasma, and for further development of the theory of collisions of heavy particles, especially in the near-threshold energy region. Such research has become more urgent in connection with searches for new active gaseous media for lasers[7].

We have developed and constructed apparatus for the study of the excitation of atoms by slow ions, investigated the excitation functions, and determined the effective cross sections for the excitation of the spectral lines of the Cd and Zn atoms in collisions between Zn$^+$ ions and Cd atoms.

EXPERIMENTAL SETUP

Experiments on the excitation of atoms by slow ions were performed by an optical method using a gas-filled cell. The main units of the setup are an ion source with a deflecting system, a collision chamber, a spectral instrument, and a block for the registration of the radiation. A schematic section through the vacuum chamber is shown in Fig. 1. Zinc ions produced in the source 1 are extracted by electrode 2 are deflected by the field of a 127° cylindrical electrostatic capacitor 4,

fall in collision chamber 9 and are detected by ion collector 10. The radiation resulting from the interaction is focused through a quartz window 8 unto the entrance slit of a monochromator and is registered with a photomultiplier.

To obtain the ion beam, a compact and economical ion source with electron bombardment was developed. It consists of a tungsten cathode, an anode (the case of the source), and a reservoir with the material. The metal vapor flows from the reservoir into the discharge chamber, is ionized by electron impact, is drawn from the anode side through a slit measuring 0.2 × 1.5 mm, and is focused at the entrance to the 127° selector with average radius 15 mm and height 24 mm. Further acceleration or deceleration of the ions is produced at the exit from the selector by electrode 5 with a slit 0.8 × 3 mm. The collision chamber is a parallelepiped (see Fig. 1) made of stainless steel, with inside bore of 20 mm diameter and 50 mm length. On one side, the cell is hermetically fastened to the accelerating electrode 5, and on the other end, the ion collector 10, 40 mm long, is introduced into the cell through a quartz
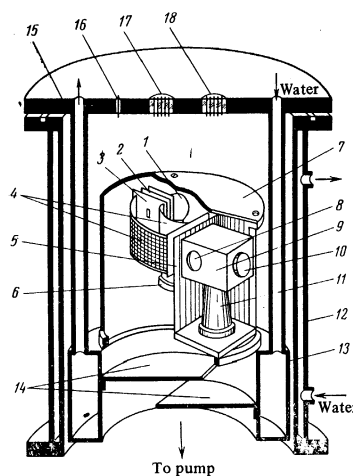

FIG. 1. Schematic section through vacuum chamber: 1—discharge chamber of ion source, 2, 3—drawing and focusing electrodes, 4—deflecting electrodes, 5—accelerating electrode, 6—source of zinc vapor, 7—case of differential-pumping chamber, 8—quartz window, 9—gas cell, 10—ion collectors, 11—source of cadmium vapor, 12—case of vacuum chamber, 13—water trap, 14—shutters of water trap, 15—upper flange of chamber, 16—18—electric leads.

insulator. The collision chamber is filled with the investigated vapor from the reservoir 11 through a vapor duct in the form of a tube of 6 mm diameter. The vapor pressure in the collision chamber was determined from the temperature of the coldest part of the reservoir, which was measured with a differential thermocouple. The ion source and the deflecting system were placed in a differential evacuation chamber 7, which together with the gas cell was mounted on the flange 15 of vacuum chamber 12. The evacuation of the chamber was by means of vacuum unit VA-05-4 with an evacuation rate of 250 liters/sec. The pressure of the residual gases in the chamber was 2×10^{-7} Torr, and under working conditions 5×10^{-7} Torr.

To separate the investigated line, we used the spectral instruments ISP-51 in the visible region and ZMR-3 in the ultraviolet region of the spectrum. The registration was effected photoelectrically, using photomultipliers FÉU-18A, FÉU-64, and FÉU-79 in conjunction with dc amplifier U1-2.

The absolute measurements of the excitation cross sections were performed by photoelectric comparison of the intensities of the spectral lines of the atoms excited by the ion beam with the intensity of a standard tungsten lamp CI8-200U, graduated at the Research Institute of the State Committee for Standards, Measures, and Measuring Instruments of the USSR.

MEASUREMENT RESULTS AND DISCUSSION

During the course of the investigations we measured the excitation functions of resonant intercombination and single lines of cadmium and zinc atoms, and also lines coming from the first-excited triplet S and D and singlet D levels of the cadmium and first-excited triplet S and singlet D levels of zinc. In addition, we determined the absolute cross sections for the excitation of 13 spectral lines of these atoms. The investigated metals were spectrally pure and prior to loading into the ion source and the gas cell they were additionally distilled in high vacuum. The differential evacuation chamber and the gas cell were heated in vacuum at 250—300° C for 25—30 hrs.

The measurements were performed at a cadmium-vapor pressure in the cell $8 \times 10^{-4} - 5 \times 10^{-3}$ Torr, an ion-beam current 0.3—1.5 μA, and a half-width of the ion-velocity scatter 3—5 eV. Under these conditions there was observed a linear dependence of the intensity of the investigated spectral line on the ion-beam current and on the cadmium-vapor pressure. Additional experiments have shown that a change of the potentials of the ion-optical system does not influence the form of the excitation functions[1]. The absolute excitation cross sections of the cadmium atoms were determined also by comparing the intensities of the spectral lines in collisions between the ions and the atoms with the intensities of the same lines excited by an electron beam, the cross sections being taken from the work of Shevera[9]. The cross sections obtained in this manner coincided within 30—60% with direct measurements with a standard lamp.

The results of our measurements are shown in Figs. 2—5 and in the table. The figures show only one line each from the group of lines starting from one upper level, since their excitation functions are identical. The lower abscissas of the diagrams represent the energies of the incoming ions in the laboratory frame, and the upper abscissas represent the particle-interaction energies in the c.m.s., and the ordinates represent the cross sections for the excitation of the corresponding spectral lines. The curves in the figures are averages of 5—20 individual measurements.

It is seen from the curves and from the table that collisions between zinc ions and cadmium atoms excite effectively the atoms of the target (cadmium), and that the charge exchange results in emission of the lines of excited zinc atoms. This feature of the interaction between zinc ions and cadmium atoms is possibly connected with the fact that the energy defects of the reaction of direct excitation and charge exchange with excitation are very close (see the table).

Another important result is the fact that the experimentally observed thresholds for the excitation of the lines coincide, within the limits of experimental error, with the energy defects of the reactions ΔE, whereas for the case of $He^+ + He$[3] the experimental threshold is larger than ΔE.

It is also remarkable that, in contrast to the Massey adiabatic hypothesis[2], the curves measured by us have a series of fine-structure maxima in the investigated energy interval. The structure of the excitation functions could be observed because of the good homogeneity of the ion beam and the high sensitivity of the photoelectric method of registration of spectral-line intensity.

Let us stop to discuss some features of the curves obtained by us. There is a distinct maximum on the excitation function of the singlet λ 2288 Å resonance line

λ, Å	Series	Number of maxima	E_{lab}^{exc}, eV (ΔE)	Positions of maxima, eV	Q, 10^{18} cm² at E_{lab} = 600 eV	
					Experiment	Calculation
			Cd			
2288	$5^1S_0 — 5^1P_1$	1	8.5	18	—	—
2881	$5^3P_1 — 6^3D_{12}$	—	12.8	—	—	—
2981 2982 }	$5^3P_2 — 6^3D_{123}$	2	12.8	45. 1000	0,1	0.3
3261	$5^1S_0 — 5^3P_i$	3	6	33. 105. 500	—	—
3466 3468 }	$5^3P_1 — 5^3D_{12}$	3	11.5	100. 400. 750	1.3	—
3611 3613 3614 }	$5^3P_2 — 5^3D_{123}$	3	11.7	100. 400. 750	3,9	17.0
4416	$5^2P_{1/2} — 5^2D_{3/2}$	2	12.8; 27.6	60. 120	1,1	1.3
4678	$5^3P_0 — 6^3S_1$	3	10,1	42. 500. 800	0,5	
4800	$5^3P_1 — 6^3S_1$	3	10,1	42. 500. 800	1,5	12
5086	$5^3P_2 — 6^3S_1$	3	10,1	42. 500. 800	2,3	
6438	$5^1P_1 — 5^1D_2$	5	11.5	36. 120. 285. 440. 760	2,1	9
			Zn			
2139	$4^1S_0 — 4^1P_1$	3	8.5	24. 220. 900—1000	—	—
3076	$4^1S_0 — 4^3P_1$	2	5.7	10. 150	>4 *	5,3
4680	$4^3P_0 — 5^3S_1$	5	9.9	60. 260. 340 620. 1050	1.0	
4722	$4^3P_1 — 5^3S_1$	5	9.9	60. 260. 340 620. 1050	2,7	8,8
4811	$4^3P_2 — 5^3S_1$	5	9.9	60. 260. 340 620. 105C	4.2	
6362	$4^1P_1 — 4^1D_2$	5	11,5	70. 125. 490 640. 850	1,5	6,4

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[1] To exclude excited ions from the beam, the energy of the ionizing electrons in the source was chosen to be lower than the threshold of excitation of the resonant line of the zinc ion [8].

*The cross section for the excitation of this line is obviously too low because of self-absorption of the resonant radiation.
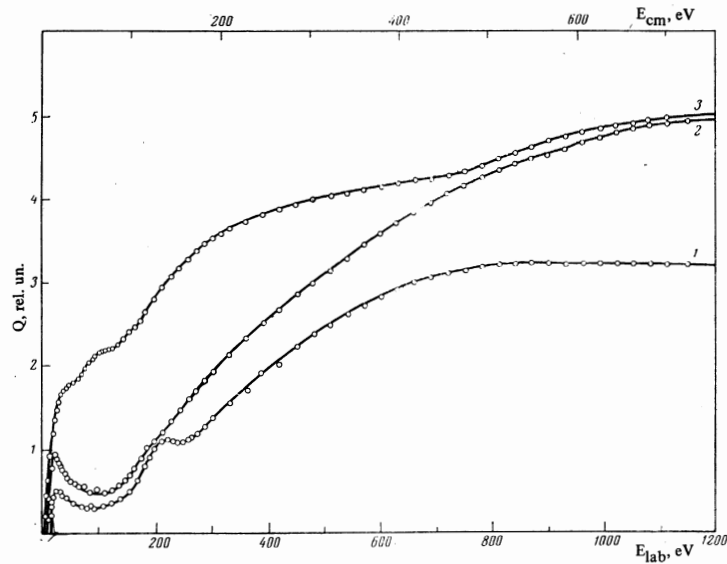
FIG. 2. Energy dependence of the cross sections for the excitation
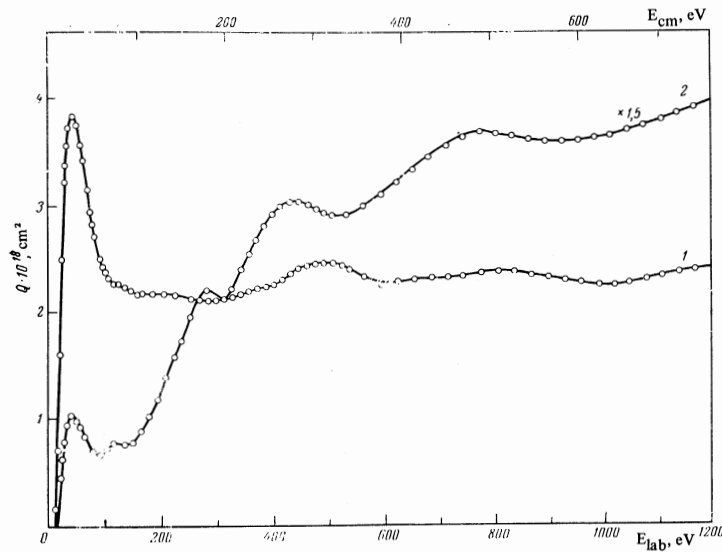of resonant lines of cadmium and zinc.

FIG. 3. Energy dependence of the cross sections for the excitation
of the spectral lines of cadmium: 1—λ5086Å, 2—λ6438Å.
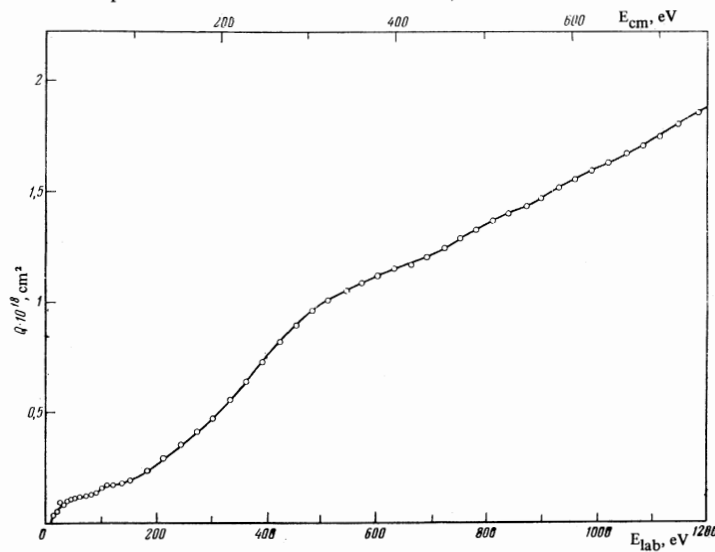

FIG. 4. Energy dependence of the cross sections of the excitation
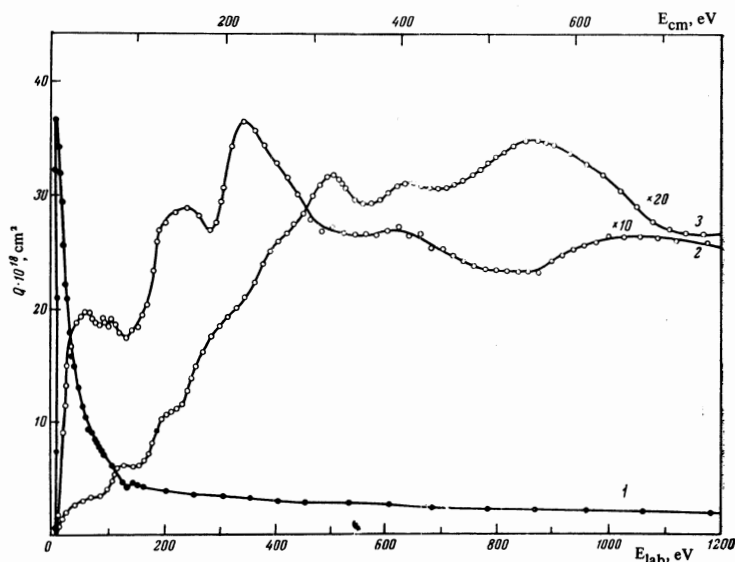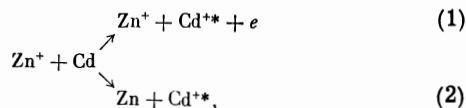of the spectral line λ4416Å of the cadmium ion.

FIG. 5. Energy dependence of the cross sections for the excita-
tion of the spectral lines of zinc: 1—λ3076Å, 2—λ4722Å, 3—λ6362Å.

of cadmium (see Fig. 2) near the threshold. A similar
maximum was observed also on the excitation function
of the λ 6438 Å cadmium line, although to be sure it is
somewhat farther from the threshold. Besides such a
distinct maximum near the threshold, the excitation
function of this line has also a number of maxima. The
absence of additional maxima on the excitation function
of the λ 2288 Å cadmium resonance line allows us to
conclude that the contribution made to it by the cascade
transitions from the 5^1D_2 level is negligible. A different
behavior is exhibited by the excitation function of the
triplet lines of cadmium. Whereas for the lines starting
from the 6^3S_1 level the cross section is maximal near
the threshold, for the line starting from the triplet
5^3P and 5^3D levels the cross section increases mono-
tonically with increasing energy of the incoming ions.
It is possible that the smooth behavior of the excitation
function of the intercombination λ 3261 Å resonance line
of cadmium is due to the large role of cascade transi-
tions from the $^3S_1$ and $^3D_1$ levels than in the case indica-
ted above.

The excitation of the λ 4416 Å cadmium-ion line
(Fig. 4) can proceed in two ways:

$$Zn^+ + Cd \nearrow Zn^+ + Cd^{+*} + e \qquad (1)$$
$$\searrow Zn + Cd^{+*}, \qquad (2)$$

and these reactions have strongly differing energy de-
fects (see the table). The threshold measured by us for
this reaction shows that the probability of the process
(2) in the near-threshold region is different from zero.
Starting with an energy E_{lab} = 27.6 eV, the process (1)
is turned on and with further growth of the energy of the
incoming Zn^+ ions the excitation function duplicates the
sum of both processes.

Just as in the case of cadmium, a distinct maximum
is observed near the threshold on the excitation func-
tions of the zinc lines (see Figs. 2 and 5). The strongest

excitation is that of the λ 3076 Å intercombination
resonance line. This is obviously connected with the
fact that the energy defect of this reaction is minimal.
The excitation function of the λ 2138 Å zinc line is
similar in form to the cadmium singlet resonance line.
A certain analogy is observed also in the behavior of
the excitation functions from the singlet D levels of zinc
and cadmium, although with certain peculiarities. How-
ever, a larger difference between the excitation func-
tions is observed for lines starting from analogous
triplet levels. Such a difference of the excitation func-
tions of the lines, in spite of the insignificant difference
between the energy defects of the reactions, is appar-
ently connected with different processes that lead to
population of the initial levels.

As seen from the figures and from the table, for
lines starting from the same levels the absolute cross
sections for the excitation of the zinc and cadmium
lines practically coincide and are smaller by several
times than the cross sections for the excitation of the
same lines by electron impact[9].

The interpretation of the fine structure of the exci-
tation functions of the lines in collisions between heavy
particles is more complicated than in the case of elec-
tronic excitation. The point is that slow ion-atom inter-
actions can occur simultaneously via several competing
channels and formation of temporary complexes (quasi-
molecules), the energy states of which are still un-
known, is possible. Our results offer convincing evi-
dence that the Massey adiabatic theory[2] is incapable
of explaining even qualitatively the observed maxima
and the oscillatory character of the excitation functions
of the lines. According to this hypothesis, the smaller
ΔE the closer the maxima of the cross sections to the
threshold, but this is not confirmed experimentally.
For example, the cross section for the excitation of the
λ 3076 Å zinc line is maximal at E_{lab} = 10 eV, whereas
there is no clearly pronounced maximum on the excita-
tion functions of the λ 3261 Å cadmium line, in spite of
the considerable difference (0.3 eV) between the energy

defects of these reactions[2]. It seems to us that a qualitative interpretation of the results given above can be obtained in the future by using the theory of phase interference between two closely lying states of the quasimolecule, on the basis of which Rosental and Bobashev explained the oscillations in the case of interaction of He[+] ions with He atoms[10] and of Na[+] ions with Ne atoms[4].

We have also calculated the cross sections of certain levels in accordance with the Grizinski formula[11]. This formula, unlike the widely used Landau-Zener formula[2] does not employ any experimental parameters. The results of the calculations for the energy of the ions E_{lab} = 600 eV are given in the table. As expected, the values of the calculated cross sections exceed by several times the experimental ones. In addition, the calculated curve does not reflect in many cases the form of the experimental curves.

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[2] From the results of [3, 4, 10, 12] it also follows that the Massey adiabatic hypothesis is not applicable to processes of excitation of atoms by slow ions.

[1] V Intern. Conf. of the Physics of Electronic and Atomic Collisions, Abstracts of Papers, Leningrad, 1967.

[2] H. Massey and E. Burhop, Electronic and Ionic Impact Phenomena, Oxford, 1952.

[3] S. Dworetsky, R. Novik, W. W. Smith, and N. Tolk, Phys. Rev. Lett. 18, 939, 1967.

[4] S. V. Bobashev, ZhETF Pis. Red. 11, 389 (1970) [JETP Lett. 11, 260 (1970)].

[5] V. S. Aleĭnikov, Opt. Spektrosk. 28, 31 (1970).

[6] R. C. Jensen, G. J. Collins, and W. R. Bennet, Phys. Rev. Lett. 23, 363, 1969.

[7] V. S. Mikhalevskiĭ and M. F. Sem, Elektronnaya tekhnika (Electronic Engineering), Series 1, No. 9, 98 (1969).

[8] A. N. Zavilopulo, O. B. Shpenik, K. Z. Tsin, and V. L. Ovchinnikov, Fourth All-Union Conference on Physics of Electronic and Atomic Collisions, Abstracts of Papers, Riga, 1959, p. 56.

[9] V. S. Shevera, Author's Abstract, Kiev, 1965.

[10] H. Rosental, VI Intern. Conf. of the Physics of Electronic and Atomic Collisions, Abstracts of Papers, Cambridge, Massachusetts, 1969, p. 302.

[11] J. D. Garcia and E. Gerjuoy, Phys. Rev. 165, 72, 1968.

[12] S. V. Bobashev, Phys. Letters 31A, 4, 204, 1970.