Cross sections for ionization of gallium and indium by electrons

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The method of crossed electron and modulated atomic beams was used to determine the cross sections σ^+ and σ^{2+} and the total ionization cross sections of gallium and indium in the electron energy range up to 200 eV. The results were compared with the available experimental data, as well as with the results of calculations carried out using the Born method, the Born method including exchange, and the semiclassical variant of the distorted wave method.

1. Reliable values of the cross sections of ionization of atoms by electrons are required in various branches of science and technology (for example, in the physics of a plasma and of a gas discharge, mass spectrometry, physics of the upper layers of the atmosphere, etc.). In the case of gallium and indium there has been only one experimental determination of the total ionization cross sections of aluminum, gallium, indium, and thallium.[1] The total ionization cross section σ_n is defined as follows:

$$\sigma_n=\sigma^{+}+2\sigma^{2+}+3\sigma^{3+}+\ldots, \tag{1}$$

where σ^{i+} is the cross section for the ith ionization. It was reported in Ref. 1 that the maximum of the total cross section of all four elements corresponds to an electron energy of about 100 eV. McGuire[2] reported calculations of the ionization cross sections of aluminum and gallium obtained in the first Born approximation and diverging considerably from the experimental data. The divergence was particularly marked in the case of aluminum, in which case the appearance of a maximum at 100 eV cannot be explained by multiple ionization effects. Bearing also in mind that elements belonging to the third group, particularly gallium and indium, are used widely in modern technology, we decided to repeat the measurements of the ionization cross sections of these elements and to obtain experimental data on the single and double ionization cross sections, because these can be compared in greater detail with the results of calculations.

2. A full description of the measurement method and of the apparatus was published earlier.[3] We shall therefore point out only the main features.

We used the technique of crossed electron and modulated atomic beams. An electron beam 1 mm in diameter was created by an electron gun of the tetrode type with an oxide cathode as an electron source and it was focused by a longitudinal magnetic field ($B\approx0.02$ T). The scatter of the electron energy in the range 5–200 eV did not exceed 0.5 eV at half the amplitude of the energy distribution. The atoms entered the equipotential region in the form of a modulated ($f\approx 10$ Hz) atomic beam created by an effusion source with resistance heating. The ions created by the ionization process were extracted by an electric field perpendicular to both beams and were directed to a collector of the total ion current or to a quadrupole mass spectrometer. The considerable difference between the method used by us and the techniques employed earlier in similar measurements[1,4-7] was the time separation of the processes of ion ionization and extraction. The electron beam and the extracting field were modulated by antiphase rectangular pulses with a modulation depth of 100% and a total period of 2 μsec. This method made it possible to use an electric field of fairly high amplitude ($E\approx5\times10^3$ V/m), which ensured complete extraction of ions both to the collector of the total ion current and to the input of the mass spectrometer. The extracting electric field had no influence on the energy and the energy scatter of electrons.

The signals from the mass spectrometer and the total-ion-current collector were measured at the frequency of modulation of the atomic beam. The ratio of these signals to the electron current, considered as a function of the electron energy, gave the energy dependence of the corresponding ionization cross sections (i.e., of the ionization function). Measurements of the total ionization cross sections σ_n were absolute. Therefore, the apparatus included equipment for using several methods to determine the concentration of our atoms in the beam, namely the method of atomic absorption,[8] the method of a quartz crystal resonator,[9] and the method of a condensation target.[4] The absolute values of the multiple ionization cross sections (σ^+ ands σ^{2+}) were determined by a difference method[6] based on the fact that the relationship (1) should be satisfied at all electron energies. In the case of gallium and indium this method did not suffer from significant error of the absolute single and double ionization cross sections, since in the case of these elements there were fairly $\sigma_n=\sigma^+$ and $\sigma_n=\sigma^+ + 2\sigma^{2+}$, and σ^+ and σ^{2+} were comparable in value (Table I). The process of measurements of the ionization cross sections and an analysis of the results were automated employing an HP 9825A minicomputer.

3. The main error in the total ionization cross section was that incurred in the determination of the concentration of atoms in a beam. In the present study these measurements were carried out by the quartz crystal resonator method. The standard substance used in calibration of the quartz resonator was lead for which the ionization cross section was determined earlier with a relative error of $\pm$ 12% using the same apparatus.[3] The degree of condensation of gallium and indium was determined in a special control experiment and direct measurements of the relative number of reflected atoms indicated that this degree differed by less than 1% from unity. The relative errors in the measurements of the ionization functions were 2.5% for σ_n and 2% for σ^+ and σ^{2+}. The total relative error in the absolute measurements of σ_n, σ^+, and σ^{2+} was 17%, 18%, and 22%, respectively. The methods of obtaining the absolute values of the cross sections σ^+

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TABLE I. Ionization cross section of Ga and In ($\times 10^{-16}$ cm^2).

	Ga			In			
E, eV	σ_n	σ^+	σ^{2+}	σ_n	σ^+	σ^{2+}	E, eV
10	2.97	2.98	–	3.79	3.73	–	10
15	4.76	4.78	–	5.85	5.76	–	15
20	5.56	5.56	–	6.96	6.96	–	20
25	5.98	5.98	–	7.38	7.36	–	25
30	6.11	6.05	0.015	7.56	7.45	0.07	30
35	6.04	5.88	0.11	7.57	7.26	0.19	35
40	6.02	5.64	0.20	7.54	6.90	0.31	40
45	5.96	5.40	0.26	7.48	6.57	0.42	45
50	5.88	5.17	0.31	7.40	6.31	0.50	50
55	5.78	4.96	0.35	7.33	6.11	0.63	55
60	5.63	4.78	0.37	7.27	5.91	0.72	60
65	5.45	4.62	0.40	7.18	5.68	0.79	65
70	5.30	4.47	0.42	7.09	5.48	0.84	70
80	5.05	4.18	0.45	6.92	5.07	0.92	80
90	4.87	3.90	0.46	6.75	4.70	0.94	90
100	4.53	3.42	0.47	6.40	4.03	0.86	100
120	4.36	3.19	0.47	6.19	3.79	0.84	120
140	4.05	2.82	0.47	5.78	3.43	0.79	140
160	3.80	2.46	0.47	5.39	3.18	0.74	160
180	3.56	2.21	0.46	5.14	2.93	0.72	180
200	3.37	2.03	0.46	4.85	2.70	0.69	200

and σ^{2+} by the difference method and by calculation of the total errors were similar to the methods described in Ref. 10.

4. Table I and Fig. 1 give the results of measurements of σ_n, σ^+, and σ^{2+} for gallium and indium between the ionization threshold and 200 eV. Figure 1 includes also the total ionization cross sections of gallium and indium taken from Ref. 1. We can see that the maximum values of the total ionization cross sections measured in the present study and those found in Ref. 1 are in good mutual agreement, but the energies corresponding to the maximum of the total cross section are quite different. The observed difference between the total ionization functions is considerably greater than the errors in the determination of these functions. We carried out a search for possible methodological errors by determining the ionization functions of barium using the same apparatus because these functions had a complex structure near the threshold and were determined earlier on several occasions.[4,5,11] The results of all these investigations were mutual agreement and they agreed also with our results within the limits of the errors given in Ref. 3.

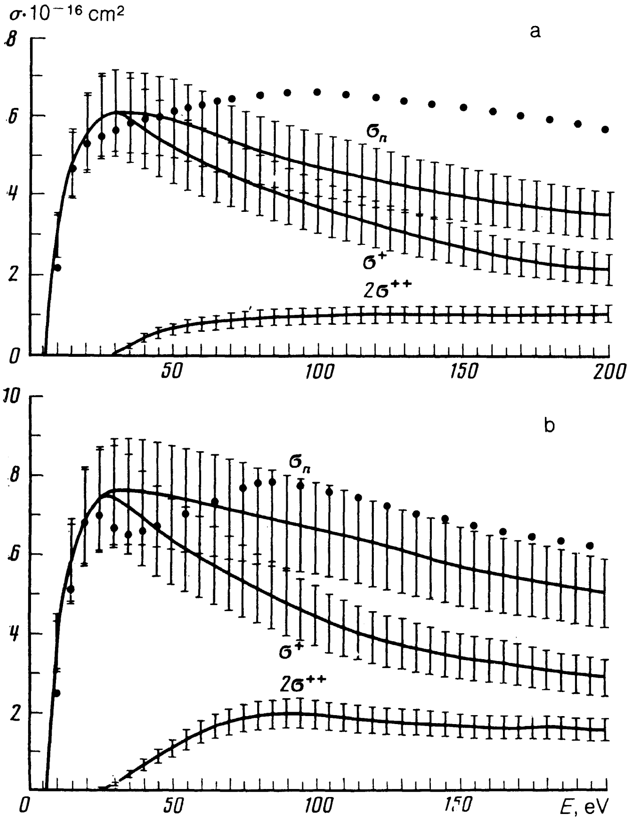

FIG. 1. Ionization cross sections of gallium (a) and indium (b) by electron impact: single (σ^+), double (σ^{2+}), and total (σ_n) ionization cross sections representing the results of our investigation; the dots represent the total ionization cross section taken from Ref. 1.

5. The experimental results were then compared with the calculations of the ionization cross sections carried out by several different methods: the Born method, the Born method including exchange, and the semiclassical variant of the distorted wave method. We allowed for the np and ns^2, and partly for the $(n-1)d^{10}$ atomic shells. The following comments should be made about the ionization energy of the last shell. The available published data are slightly contradictory. According to Ref. 12, at least a large number of the d^9ns^2np terms are characterized by an LS energy exceeding double the ionization energy of the atom. In other words, these terms experience preionization decay and contribute to the double ionization cross section σ^{2+}, which we determined independently. In the treatment given below the contribution of the $(n-1)d^{10}$ shell refers entirely to σ^{2+}, but this aspect requires careful study.

The single ionization cross section is defined as follows:

$$\sigma^+=\sigma(np)+2\sigma(ns), \qquad (2)$$

where

$$\sigma(nl)=\sum_{l_1=0}^{l_m}\int_0^{\varepsilon_m} d\varepsilon_1\sigma(nl,\varepsilon_1 l_1), \qquad \varepsilon_m=(E-\varepsilon_{nl})/2. \qquad (3)$$

Here, E is the energy of an incident electron; ε_1 and l_1 are the energy and momentum of a knocked-out electron; $\varepsilon_{nl}>0$ is the binding energy for the nl shell.

The upper integration limit in Eq. (3) is selected in accordance with the general theory of ionization allowing for the identity of the particles. Therefore, in accordance with our terminology, the Born method and the Born method with exchange differ by the presence or absence of the exchange term but the integration limits are the same. This definition of the Born cross section in the problem of ionization is reasonable because the ignored contribution of all those collisions which are characterized by $(E-\varepsilon_{nl})/2<\varepsilon_1<E-\varepsilon_{nl}$ is still estimated incorrectly in the Born approximation since the primary electron is now too slow.

Calculations of the Born type were carried out using the "Atom program (written at the Lebedev Physics Institute).[13] Calculations indicated that in the summation with respect to l_1 in Eq. (3) it is sufficient to limit the process to $l_m=7$. The values of $\sigma(nl,\varepsilon_1 l_1)$ were calculated in the q representation[14]:

$$\sigma(nl,\varepsilon_1 l_1)=\frac{8p}{E}\sum_{\varkappa}\int_{K-K'}^{K+K'}\frac{dq}{q^3}|R_\varkappa(q)|^2\quad[\pi a_0^2],\qquad(4)$$

where $p=1$ for the np shell, $p=2$ for the ns^2 and $(n-1)d^{10}$ shells,

$$K=E^{1/2},\qquad K'=(E-\varepsilon_{nl}-\varepsilon_1)^{1/2},$$

and $R_\varkappa(q)$ is the radial matrix element (see Ref. 14). The radial wave functions of an atom were calculated using the same program employing a semiempirical method and the experimental value of the energy ε_{nl}. The Born approximation with exchange requires the use of partial waves instead of Eq. (4), which increases considerably the volume of the calculations. The method of orthogonalized functions was used to avoid the shortcomings of the Born-Oppenheimer approximation.

A theory of the semiclassical calculation of the cross sections and the details of the calculation procedure for gallium and indium were described in detail in Ref. 15. The essence of the method is the use of semiclassical wave functions for the atomic and external electron beams. Since these functions are defined in the same field, the problem of orthogonalization does not arise and the exchange term is obtained by a simple transposition of the momenta of scattered and knocked-out electrons. The radial integral was calculated by the stationary phase method. This approach was analogous to the usual distorted wave method with exchange. However, additional simplifications reduced considerably the time needed for the calculations.

6. The results of the calculations are compared in Fig. 2 with the experimental data obtained in the present study. We can see that the Born cross sections are in good agreement with the experimental results. If the exchange interaction is allowed for, the cross section rises more steeply at the threshold and the curve begins to bend at 10–15 eV. The experimental results do not confirm these predictions. It should be pointed out that although the exchange effect is quantitatively small, it exceeds the experimental error. The cross section found by the semiclassical method in the vicinity of the threshold is similar to the Born cross section with exchange. After attainment of the maximum this cross section falls too rapidly. Clearly, this is due to the insufficient number of the partial waves included in the calculations, since the sum over the partial waves converges slowly.

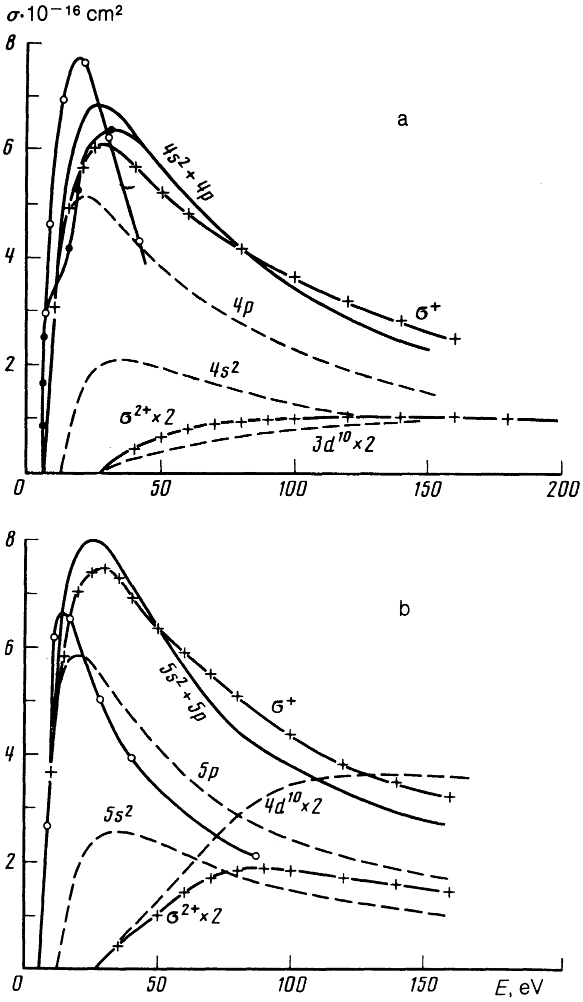

FIG. 2. Ionization cross sections of gallium (a) and indium (b) by electron impact: +) experimental results obtained in the present study; O) calculation of $(4s^2+4p)$ using the semiclassical variant of the distorted wave method; the continuous and dashed curves are calculations by the Born method; the continuous curve with the black dots is a calculation of $(4s^2+4p)$ by the Born method with exchange.

As pointed out earlier, the ionization of the d^{10} shell is not included in the total single ionization cross section σ^+ in Fig. 2. The bulk of the ionization events d^{10} is accompanied by preionization, i.e., it contributes to σ^{2+}. It is clear from Fig. 2 that $\sigma(d^{10})$ agrees well with σ^{2+} in the case of gallium and it is double the value of σ^{2+} in the case of indium. Clearly, in the latter case, ~50% of the ionization events are not accompanied by preionization and do not contribute to σ^+. This is in agreement with the observation that the experimental cross section σ^+ exceeds considerably the calculated cross section $\sigma(4p+4s^2)$ at high energies. It should be pointed out that Fig. 2 gives the doubled values of the cross sections σ^{2+} and $\sigma(d^{10})$.

We shall note in conclusion that the gallium and indium ionization cross sections obtained in the present study are in good agreement with the calculations and with the Born approximation. However, a reliable separation of the calculated values of σ^+ and σ^{2+} requires additional data on the energies of the $(n-1)d^{10}ns^2np$ terms.

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