SEMI-EMPIRICAL METHOD FOR THE CALCULATION OF THE EQUILIBRIUM DISTRIBUTION OF CHARGES IN A FAST-ION BEAM

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Submitted to JETP editor February 21, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 47, 615-623 (August, 1964)

A simple semi-empirical method is proposed for calculating the mean charge and equilibrium distribution of the charges of fast ions moving in solid and gaseous media. In agreement with the experimental data, the number of ions in a beam containing different charges is assumed to depend on two parameters and to be described by a Gaussian curve. To evaluate the mean charge and width of the equilibrium distribution, the generalized Bohr criterion is employed in conjunction with the statistical model of the ion and with some concrete experimental data.

1. INTRODUCTION

WHEN fast multiply-charged ions pass through matter, their charge fluctuates as a result of electron loss and capture, and a dynamically stable charge distribution is established in the ion beam even before a noticeable slowing down of the fast particle takes place. This distribution does not depend on the initial ion charge and is determined solely by the cross sections for the loss and capture of electrons.

Information on the equilibrium charge distribution in ion beams is needed for the analysis of heavy particles produced in nuclear reactions, to solve problems involved in the production of fast multiply-charged ions in accelerators, and for other problems in nuclear physics. However, a consistent theoretical derivation of the equilibrium charged state of ion beams from the cross sections for the loss and capture of electrons is quite cumbersome, and at the present time it apparently cannot give sufficiently accurate results, owing to the difficulties involved in the calculation of the cross sections. Consequently, great importance is attached to approximate methods of solving the equilibrium distribution, which do not require calculations of the cross section.

We propose here a simple semi-empirical method, based on established laws governing the equilibrium charge distribution and using concrete experimental data. This method can be used to calculate the charge composition of a beam of ions of arbitrary elements with atomic numbers $Z > 2$, at ion velocities $v > v_0 = e^2/\hbar = 2.19 \times 10^8$ cm/sec.

2. FORM OF EQUILIBRIUM CHARGE DISTRIBUTION

In view of the relative smallness of the cross sections for simultaneous loss and capture of several electrons, the distribution of the ions among the most intense charge groups is determined essentially by the cross sections for the loss and capture of a single electron [1,2], so that if the relative number of ions with charge i is denoted by F_i ($\Sigma_i F_i = 1$), then

$$F_{i+1} / F_i \approx \sigma_{i,\,i+1} / \sigma_{i+1,\,i}, \qquad (1)$$

where σ_{ik}—cross section of the process whereby an ion with charge i is converted into an ion with charge k. Inasmuch as the dependence of the ratios $\sigma_{i,i+1}/\sigma_{i+1,i}$ on i is nearly exponential, we have

$$F_{i+1} / F_i \approx \exp(a - bi). \qquad (2)$$

The exponential dependence of the ratio F_{i+1}/F_i on i for the most intense charge groups is confirmed by practically all the experimental investigations of the equilibrium charge distribution. When the average ion charge $\bar{i} = \Sigma_i i F_i$ satisfies the condition $1 \lesssim \bar{i} \lesssim Z - 1$, the equilibrium charge distribution is therefore close to Gaussian:

$$F_i \approx (2\pi d^2)^{-1/2} \exp[-(i - \bar{i})^2 / 2d^2] \qquad (3)$$

and is characterized essentially by two parameters: the average charge $\bar{i}$ and the half-width of the distribution $d = [\Sigma_i (i - \bar{i})^2 F_i]^{1/2}$, which are connected with the quantities a and b of (2) by the approximate relations:

$$\bar{i} \approx a/b + 1/2, \qquad d^2 \approx 1/b. \qquad (4)$$

The number of ions constituting low-intensity charge groups is decisively influenced by processes involving simultaneous loss and capture of several electrons [1,2]. As a result, the values of F_i can be larger than Gaussian for $i \ll \bar{i}$, and particularly for $i \gg \bar{i}$ [3]. We present below methods for calculating the values of $\bar{i}$ and d, from which we can obtain the Gaussian distribution of the charges.

3. AVERAGE ION CHARGE

In the calculation of the average ion charge $\bar{i}$ we shall make use of experimental information. The most complete data on the value of $\bar{i}$ in solid and gaseous media, over a sufficiently wide velocity interval, is available at present for ions with $Z \le 10$ [3-15]. For heavier ions with $Z \lesssim 50$, the data on the charge distributions have been obtained in a relatively small velocity interval, and have no systematic character. For sodium, magnesium, aluminum, and krypton ions the measurements of the equilibrium charge distribution in different media were made at $v \approx 2.6 \times 10^8$ cm/sec, while for phosphorus and argon ions also at $v \approx 4.1 \times 10^8$ cm/sec [4,11]. The average degree of the ionization $\bar{i}/Z$ was of the order of 0.2—0.3.

The equilibrium charge distribution in carbon is known for chlorine ions at $v \approx (6-13) \times 10^8$ cm/sec ($\bar{i}/Z \sim 0.4-0.6$) and for bromine ions at $v \approx (6-9) \times 10^8$ cm/sec ($\bar{i}/Z \sim 0.3-0.4$) [14]. Measurements were also made of the average charge of uranium fission fragments [16,17]. In gases these measurements were made for the light group of fragments ($Z \sim 38$) at $v \approx (5.5-14) \times 10^8$ cm/sec, when $\bar{i}/Z \sim 0.2-0.4$, and for the heavy group ($Z \sim 54$) at $v \approx (4.4-11) \times 10^8$ cm/sec, when $\bar{i}/Z \sim 0.1-0.3$ [16]. The average charge in solid substances is known only for velocities close to maximal [17], when $\bar{i}/Z \sim 0.5$ for the light group and ~ 0.4 for the heavy group.

To interpolate and extrapolate the values of $\bar{i}$ to the region of v and Z for which there are no direct experimental data, we make use of the Bohr criterion [18,19], according to which the ion retains only those electrons whose orbital velocity v_e exceeds the ion velocity v. As applied to real atoms and ions, this criterion leads, however, to results that differ appreciably from the experimental data [16,20]. Thus, if the values of $\bar{i}$ were to be determined only by v_e, then a sharp decrease in the dependence of $\bar{i}$ on v would occur on going from the ionization of one electron shell to the ionization of another shell, and minima would appear on the $\bar{i}$ vs. Z curve. Actually, the shell structure is not so distinctly manifest and the dependence of $\bar{i}$ on v and Z is smoother. This is connected with the fact that the electron loss and capture cross sections, and consequently also the values of the average charge, depend not only on v_e but also on the number of electrons in the outer shell of the atom, on the degree of its filling, and on the effective charge of the ion [1,2,21,22]. The action of each of these factors leads to a smoothing of the curves of $\bar{i}$ against v and against Z. Consequently, the Bohr criterion agrees best with experiment if the statistical model of the atomic particle is used for the description of the ion; according to this model [23,4]

$$v_e \approx Z^{\alpha} f(i/Z), \qquad (5)$$

where $f(i/Z)$ is a monotonically increasing function of i/Z, and the exponent α can range from $1/3$ to $2/3$, depending on the assumptions made.

Since the Bohr criterion should similarly yield the ion velocity v at which the average degree of ionization $\bar{i}/Z$ assumes a specified value, it follows from (5) that log v is a linear function of log Z:

$$\lg v = \alpha \lg Z + \lg f(\bar{i}/Z). \qquad (6)^*$$

The experimental values of log v as functions of log Z for a series of equidistant values of $\bar{i}/Z$ from 0.2 to 0.9, obtained by measuring the average charge of ions in solids, are shown in Fig. 1. Since the values of $\bar{i}/Z$ for $Z > 3$ in different solids with atomic numbers $Z \sim 5-50$ differ by not more than 2—5% [8,17], Fig. 1 shows the results of measurements for all solids. Figure 2 gives analogous values for nitrogen and argon—gases for which the most data are available.

As can be seen from Figs. 1 and 2, for ions of different elements with $Z \lesssim 50$ the experimental values of log v actually lie near straight lines. However, the slopes of the lines are not constant, and decrease monotonically with decreasing $\bar{i}/Z$. In gases this effect is small: when $\bar{i}/Z$ changes from 0.8 to 0.3 the exponent α decreases from 0.6 to 0.5. On the other hand, when the gases pass through solids, the values of α decrease rapidly with increasing $\bar{i}/Z$, and become smaller than $1/3$ when $\bar{i}/Z < 0.3$. This is in all probability connected with the fact that the ions passing through

*lg = log.

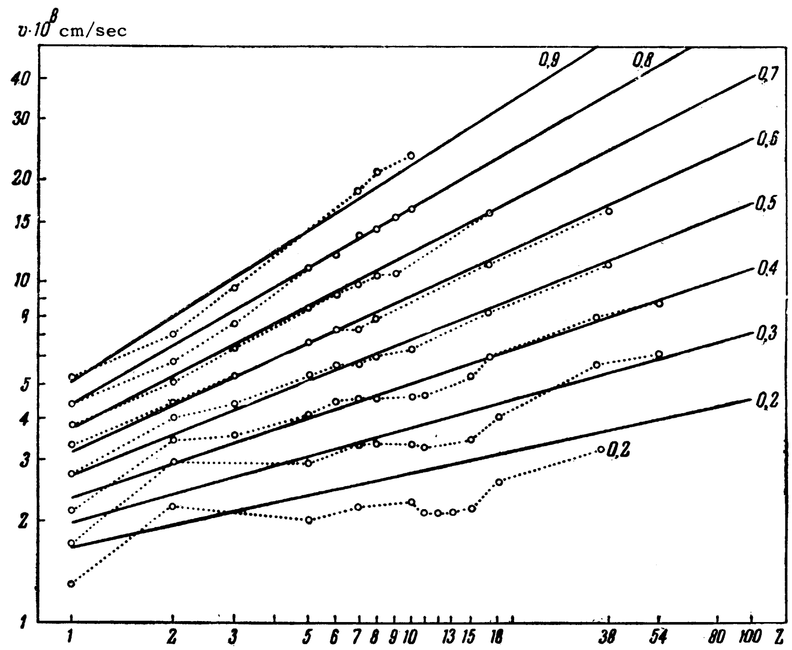

FIG. 1. Ion velocity v at which $\bar{i}/Z$ in solids reaches a specified value as a function of Z, in accordance with data from[3–6,8–10, 13–15, 17]. The lines are drawn in accordance with formulas (7) with the coefficients taken from the table. The values of $\bar{i}/Z$ are marked next to the lines. The experimental points are joined here and in other figures by dotted lines.

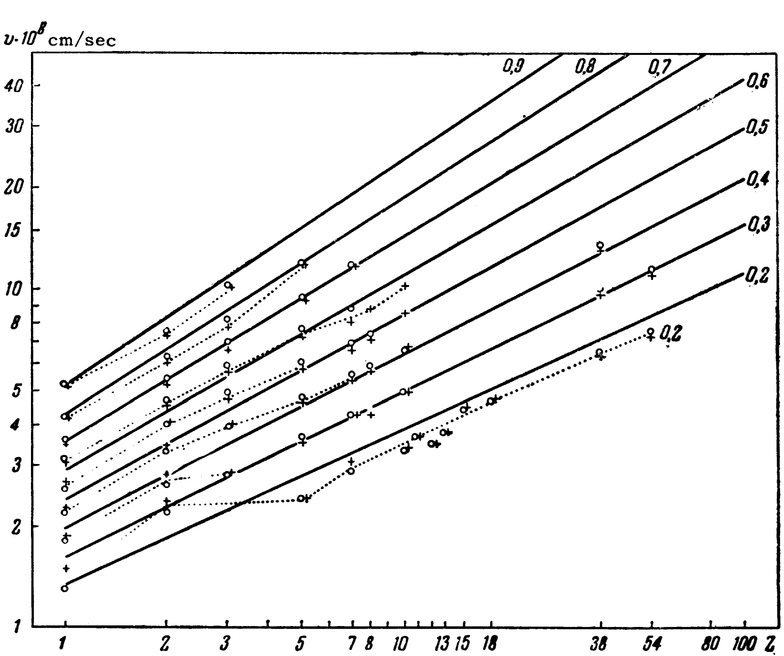

FIG. 2. Ion velocity v at which $\bar{i}/Z$ in nitrogen (o) and in argon (+) reaches a specified value, as a function of Z, in accordance with data from[3,7,10,12,16]. The lines are plotted in accordance with formulas (7) with coefficients from the table. The values of $\bar{i}/Z$ are indicated near the lines.

solids are excited and consequently cannot be described by the statistical model of an atomic particle in the ground state.

The experimental dependence of v on $\bar{i}/Z$ differs from the statistical-model dependence of v_e on $\bar{i}/Z$. This difference, together with the dependence of the exponent α on $\bar{i}/Z$, signifies that the experimental values of $\bar{i}/Z$ satisfy not the simple Bohr criterion, but the generalized criterion[23,24], according to which the average ion charge is determined by the condition $v_e = \gamma v$, where γ—slowly varying quantity on the order of unity. From the experimental data it follows that γ is a function of $\bar{i}/Z$, of Z, and also of the medium in which the ions move.

The introduction of γ takes into account the effect exerted on the value of $\bar{i}$ by the number of external electrons in the ion, by the degree of filling of the external shell, and by the effective charge of the ions. The assumption made in this case is that the factors that determine γ are assumed to be monotonically dependent on v and Z. By the same token, no account is taken of the weak dependence of $\bar{i}$ on the singularities of the structure of the electron shell of the ion, a dependence which exists all the same and which causes, as can be seen from Figs. 1 and 2, deviations from the linearity of log v against log Z. The greatest deviations were obtained for small values of $\bar{i}/Z$, for the ions with $Z \leq 3$ and $Z \sim 10-13$, i.e., in regions of Z where a transition takes place from the filling or ionization of one electron shell to the filling or ionization of the other. In the first case these deviations are larger than in the second, so that the deviations of the actual values of log v from the values given by the averaged straight lines become lower with increasing Z, when the number of electrons that get involved in the change of the ionic charge increases.

The lines drawn in Figs. 1 and 2 correspond, for $\bar{i}/Z$ between 0.3 and 0.9, to formula (6) with

$$\alpha = \alpha_1 + \bar{i}Z^{-1}\alpha_2, \quad \lg f = \lg m + \bar{i}Z^{-1} \lg n. \qquad (7)$$

The values of the coefficients α_1, α_2, m, and n are listed in the table. The ion velocity v is expressed here in units of 10^8 cm/sec.

Values of coefficients used in (8) and (11) for $\bar{i}$ and d in gases and in a solid substance (T).

Medium	α_1	α_2	m	n	$\varkappa$	d_0
H_2	0.4	0.3	1.2	4.0	0.43	—
He	0.4	0.3	1.3	4.5	0.43	0.35
N_2	0.4	0.3	0.9	7.0	0.45	0.32
Ar	0.4	0.3	0.9	7.0	0.45	0.32
T	0.1	0.6	1.2	5.0	0.40	0.38

It follows from (6) and (7) that in the region from 0.3 to 0.9 the degree of ionization $\bar{i}/Z$ assumes the form

$$\bar{i}/Z = \lg (vZ^{\alpha_1}/m)/\lg nZ^{\alpha_2}, \qquad (8)$$

i.e., the average charge $\bar{i}$ is proportional to log v. In the region $\bar{i}/Z < 0.3$, the quantity $\bar{i}$ can be assumed to be proportional to the velocity v, i.e.,

$$\bar{i}/Z = AZ^{-1/2}v. \qquad (9)$$

The proportionality coefficient A is equal to approximately 0.16 in helium and 0.18 in nitrogen and argon.

Figure 3 shows plots of $\bar{i}/Z$ vs. v, calculated from (8), for boron, neon, chlorine, bromine, and mercury ions in solids, together with the available experimental data. Analogous curves for the dependence of $\bar{i}/Z$ on v in nitrogen and argon, calculated from formulas (8) and (9), are shown in Fig. 4. We see from the figures that the deviation of the calculated values of $\bar{i}/Z$ from the measured ones does not exceed 5% as a rule.

In other gases the experimental values of the average ion charge are much scantier. Therefore the coefficients α_1, α_2, m, and n listed in the table for hydrogen and helium are tentative. To estimate $\bar{i}/Z$ in gaseous media, where there are no direct measurements, we can make use of the fact that the difference between the values of $\bar{i}/Z$ in different media is of the same order of magnitude as the deviation of $\bar{i}/Z$ in a given medium from the values obtained from (8) and (9). We can therefore obtain the values of $\bar{i}$ in arbitrary gaseous media from the data shown in Fig. 2 and in the table, accurate within 10–20%.

4. WIDTH OF EQUILIBRIUM CHARGE DISTRIBUTION

The presence of a noticeable number of ions with different charges in an ion beam having an equilibrium charge composition signifies that some of the ions retain electrons with orbital velocity v_e considerably lower than γv, whereas other ions lose electrons with $v_e \gg \gamma v$. This indicates that the Bohr criterion is satisfied only in the mean, and that when the ion velocity changes the probability of removing an electron with a given velocity v_e does not vary abruptly, and never

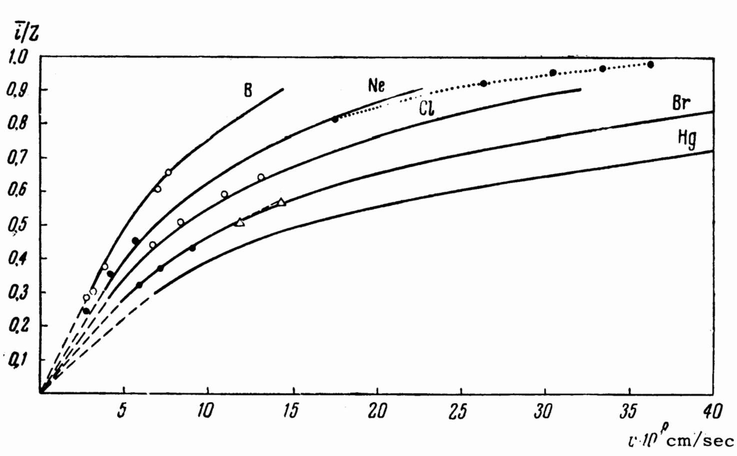

FIG. 3. Values of $\bar{i}/Z$ for boron, neon, chlorine, bromine, and mercury ions as functions of v in a solid, obtained from formula (8). The experimental values for the boron ions are taken from[3], for neon ions at $v = (2.6 - 5.5) \times 10^8$ cm/sec from[3] and at $v \geq 17 \times 10^8$ cm/sec from[15], and for chlorine and bromine ions from[14]; Δ---Δ – values of $\bar{i}/Z$ for the light group of uranium fission fragments, from[17].

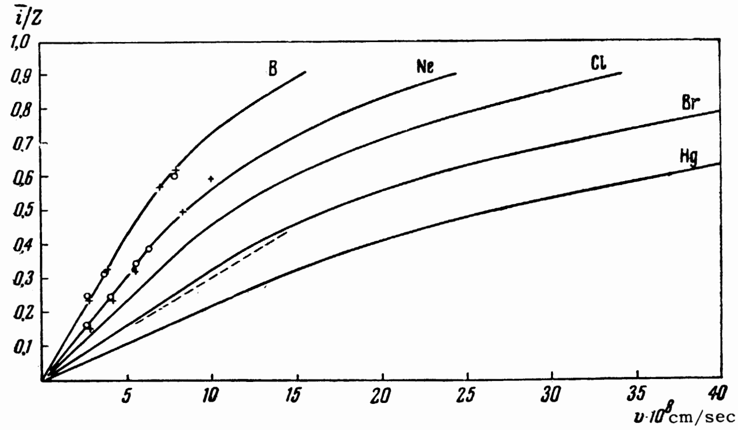

FIG. 4. Values of $\bar{i}/Z$ for boron, neon, chlorine, bromine, and mercury ions as functions of v in nitrogen and argon, calculated by formulas (8) and (9). Experimental values in nitrogen (o) and argon (+) were obtained as follows: for boron ions from[3], for neon from[3, 7]; dashed line – values of $\bar{i}/Z$ for the light group of uranium fission fragments ($Z \sim 38$) from[16].

reaches the limiting values zero and unity.

As a measure of the probability of removal of the next $(i + 1)$-st electron from an ion with charge i we can use the quantity

$$w = \frac{F_{i+1}}{F_i + F_{i+1}} = \left[1 + \frac{F_i}{F_{i+1}}\right]^{-1}.$$

Inasmuch as the average charge of the ion is determined, in accordance with the generalized Bohr criterion, by the equation $v_e = \gamma v$, it is natural to regard w, and consequently also the ratio F_{i+1}/F_i, as a function of $x = \gamma v/v_e$, with $F_{i+1}/F_i = 1$ for $x = 1$. Expanding the quantity log $(F_{i+1}/F_i) = \varphi(x)$ in a series in the vicinity of $x = 1$, i.e., near $i = \bar{i}$, and recognizing that in accordance with (8) we have $v_e/\gamma = mZ^{\alpha_1}(nZ^{\alpha_2})^{\bar{i}/Z}$, i.e., $x = v/mZ^{\alpha_1}(nZ^{\alpha_2})^{\bar{i}/Z}$, we obtain

$$\varphi(x) = \varphi(1) + \varphi_x'(1)(i - \bar{i})\partial x / \partial i_{i=\bar{i}}$$

$$= k[\ln(nZ^{\alpha_2})(i - \bar{i}) / Z]. \quad (10)$$

By comparing (10) with (2) and (4), we see that we can obtain from the Bohr criterion a Gaussian charge distribution with half-width

$$d = d_0[Z / \ln(nZ^{\alpha_2})]^{1/2} = d_0 Z^{\varkappa},$$

$$\varkappa = {}^1/_2\{1 - d \lg [\ln(nZ^{\alpha_2})] / d \lg Z\}$$

$$= {}^1/_2(1 - \alpha_2 / \ln(nZ^{\alpha_2})). \quad (11)$$

Using the experimental data for the coefficients n and α_2, we get $\kappa \sim 0.45$ for gases and $\kappa \sim 0.4$ for solids (see the table).

According to the available experimental data, in the region of velocities where $\bar{i}/Z$ varies for ions of a given element from 0.3 to 0.7, the half-width d of the equilibrium charge distribution in solids remains approximately constant, decreasing for $\bar{i}/Z < 0.3$ and for $\bar{i}/Z > 0.7$ (see Fig. 5). For ions with $Z \geq 5$ in solids, as can be seen from Fig. 6, the value of d in this region of $\bar{i}/Z$ increases in the mean like $Z^{0.4}$, in accordance with (11), whereas d_0 can be assumed equal to 0.38. Thus, from the generalized Bohr criterion and from the experimental values of the parameters α_2 and n, which characterize the average charge, we obtain the correct Z-dependence of the equilibrium distribution width d.

In gases, for equal values of $\bar{i}/Z$, the equilibrium distribution is somewhat narrower than in solids. However, these data pertain essentially to the region $\bar{i}/Z < 0.3$. The value of d for gaseous media with $\bar{i}/Z \geq 0.3$ is available only for ions with $Z \leq 10$. The value of d in gases with the same region of $\bar{i}/Z$ can be represented in the form (11). In this case the values of d_0 are close to 0.3 and differ in different gases by not more than 10% (see the table).

Formula (11) corresponds to the statistical

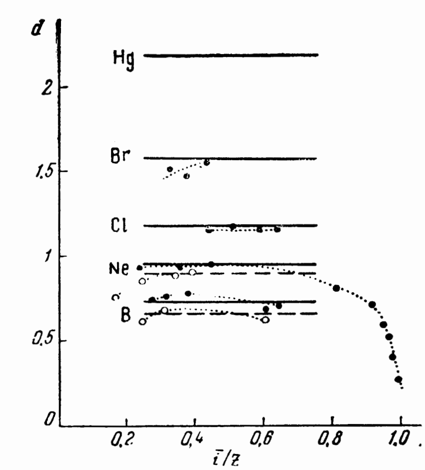

FIG. 5. Dependence of the equilibrium distribution half-width d on Z in solids (filled circles and solid lines) and in nitrogen (light circles and dashed lines). For boron, neon, chlorine, bromine, and mercury ions. Experimental values of v are taken from[3] for boron and neon ions and from[14] for the chlorine and boron ions. The solid and dashed lines are plotted in accordance with (11).

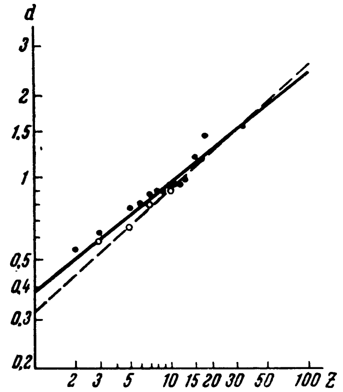

FIG. 6. Dependence of d on Z for $\bar{i}/Z \sim 0.3 - 0.7$ in solids (filled circles and solid line) and in nitrogen (light circles and dashed line). The experimental values are taken from[3, 4, 9, 10, 14]. The solid and dashed lines are plotted in accordance with (11).

model and does not reflect the singularities of the structure of the electron shells of individual atoms and ions. The shell structure is manifest, in particular, in the fact that no increase in d is observed in solids for Z in the region 10—13, i.e., on going from ionization or filling of the L shell to ionization of filling of the M shell (see Fig. 6). In spite of these deviations, the experimental values of d differ from those obtained by formula (11) by not more than 10%.

From (3) and from (8), (9), and (11) we can obtain the equilibrium distribution for arbitrary ions in different media. Figure 7 shows by way of an example the calculated values of F_i in solids, as functions of $i - \bar{i}$, for boron, neon, chlorine, argon, and mercury ions. The figure shows also the experimental data. As can be seen from the figure, for $F_i \gtrsim 0.1$ the calculated values of F_i differ from the measured ones by not more than 5—10% as a rule.

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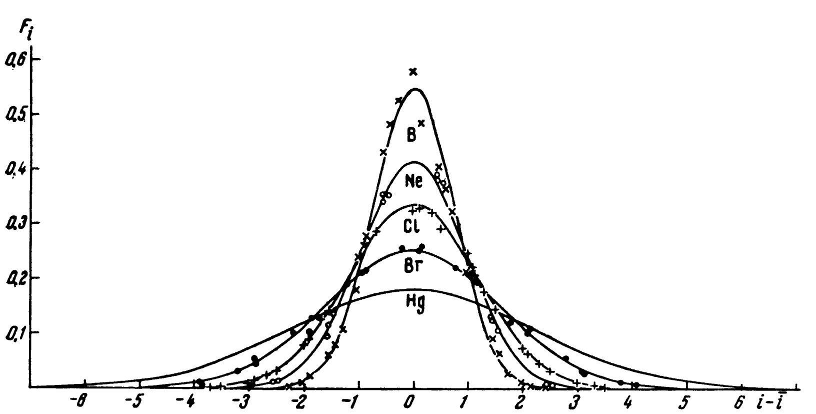

FIG. 7. Values of F_i in solid substance for boron, neon, chlorine, bromine, and mercury ions, calculated as functions of $i - \bar{i}$. The experimental values of F_i for boron (×) and neon (o) ions are taken from[3], while for the chlorine (+) and bromine (•) – from[14].

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Translated by J. G. Adashko
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