EQUILIBRIUM DISTRIBUTION OF CHARGES IN A BEAM OF IONS OF LIGHT ELEMENTS

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The equilibrium distribution of ionic charges of light elements with atomic numbers Z between 5 and 10 was studied after passage of the ions through hydrogen, air, argon, and celluloid film with velocities ranging from 3.5 to $8-11 \times 10^8$ cm/sec. The mean ionic charge $\bar{i}$ was found to be from 10 to 30% larger after passage through the film than after passage through air. The values of $\bar{i}$ in gases were found to differ by $10-20\%$. The distribution of the ions in a beam with respect to $\bar{i}$ is approximately Gaussian.

1. INTRODUCTION

It is well known that when accelerated multiply charged ions pass through matter, the exchange of electrons between the ions and the atoms of the medium results in the establishment of an equilibrium distribution of charges which is dependent on the velocity of the particles. The study of the equilibrium distribution of charges is of considerable interest both for practical purposes and for the purpose of improving our understanding of the processes which occur when ions pass through matter in the region for which there is no rigorous theory. The results of such investigations can be used in the study of problems associated with the production of multiply charged ions at high energies, in determining the nature of the particles generated in nuclear reactions, etc.

Thus far only very incomplete experimental data have been available regarding the distribution of the ionic charges of light elements. The most systematic results have been obtained at energies ranging from $20-40$ to 200 kev.[1,2] For ions with nuclear charge $Z \gtrsim 5$ this energy region corresponds to velocities $v < v_0 = e^2/\hbar$, at which the mean ionic charge $\bar{i}$ is many times smaller than Z. For higher energies data on the charge distribution exist for helium ions[1,3,4] passing through various substances, for lithium[5] and nitrogen[6,7] ions after passage through organic films and for individual energies of nitrogen ions in nitrogen,[8] of oxygen and neon ions in argon[9] and of nitrogen, oxygen, and fluorine ions after passage through organic films.[10] Theoretical calculations of the charge distributions and mean charges for ions with $Z > 2$ (Refs. $11-15$) are approximate and are in need of experimental verification.

In the present work we have determined the equilibrium distribution of the ionic charges of light elements with atomic numbers Z from 5 to 10 after their passage through hydrogen, air, argon, and celluloid film with velocities ranging from 3.5 to $8-11 \times 10^8$ cm/sec, i.e., for $v \sim 1.5-5\, v_0$.

2. EXPERIMENTAL METHOD

The source of fast particles was a 72-centimeter cyclotron. In this work we used the following accelerated ions:

$$^{11}B^{+1,+2,+3};\ ^{13}C^{+2,+3};\ ^{14}N^{+2,+3};\ ^{16}O^{+2,+3} \text{ and } ^{20}Ne^{+2,+3}$$

A narrow ion beam extracted from the cyclotron and passing through slit 1 (Fig. 1) was deflected in the magnetic field H_1 and passed through the entrance channel 2, of cross section 1×0.1 cm^2 and length 3 cm, into the charge-exchange chamber of about 4.8 m length. Exit channel 3 had a length of 3 cm and cross section 0.5×0.1 cm^2. The particles passing through the chamber were analyzed by magnet H_2 and were then registered by counters 5, which were described in Ref. 7. The pressure in the charge-exchange chamber during measurements of the equilibrium charge distribution was approximately 70 times greater than the pressure in adjacent portions of the vacuum system and was varied from 5×10^{-3} to 8×10^{-2} mm Hg.

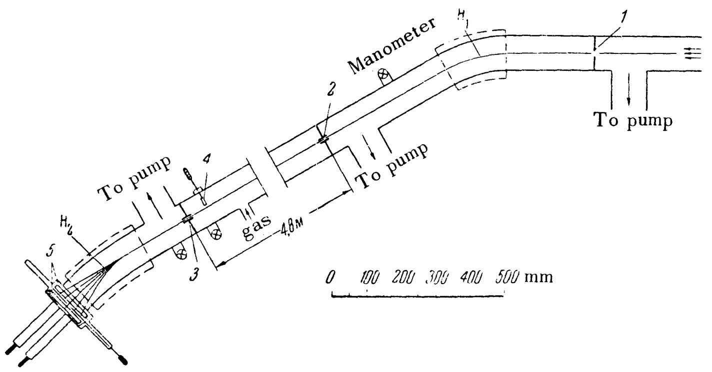

FIG. 1. Diagram of the apparatus

In the majority of cases the charge of the accelerated ions was smaller than the mean charge $\bar{i}$ of the equilibrium distribution. Therefore, when the pressure was increased, the mean charge at first increased and then began to decrease because the particles were decelerated by the gas. The maximum mean charge corresponded to the pressure at which the velocity loss of the particles passing through the charge-exchange chamber was about 2%. At this and higher pressures, the charges had practically an equilibrium distribution. The relative intensity Φ_i of any individual charge group in the ionic charge distribution (i) was determined by the velocity of the ions and varied little with the pressure. In most instances the error in values of Φ_i (Table I) was the possible result of ion scattering in the gas. The magnitude of this error was estimated from the variations of Φ_i with pressure. The statistical error was important only for $\Phi_i \leq 0.02$. Errors due to other causes were insignificant.

In order to determine the equilibrium charge distribution after the passage of ions through an organic film, film 4 of thickness $\sim 10 \mu g/cm^2$ was placed in the path of the beam, 2 cm in front of channel 3. After passage through this film the ions had an equilibrium distribution, as was shown by measurements with a film of twice the thickness. During the measurements with the film the pressure along the entire trajectory of the beam was $\sim 1-2 \times 10^{-5}$ mm Hg. The width of channel 3 remained 1 mm, as during the determination of charge distribution in gases. In some instances the distributions were compared as the width of the channel was varied from 0.3 to 3 mm. The variation of the relative intensities of individual charge groups then did not exceed in absolute value the variations which resulted from a 1% change of velocity.

TABLE I. Relative errors of experimental values of Φ

Φ	0.4	0.1	0.02	0.003
$\Delta\Phi/\Phi$	0.02	0.04	0.010	0.20

The velocity of the particles was calculated from the field strength of the deflecting magnet, which was calibrated with deuterons of known energy, making allowance for energy losses in the gases and film. The error in the velocity determinations was due mainly to inaccuracy in the calibration of the magnet and amounted to $\sim 2\%$. The error in the velocity ratio did not exceed 1% and resulted from variation of the ion trajectories in the magnetic field of the cyclotron and from errors in determining the field of the deflecting magnet. In each individual experiment the velocity spread of the ions traversing channel 2 into the charge exchange chamber did not exceed 0.5%.

3. EXPERIMENTAL RESULTS

The measurements of the equilibrium distribution of boron, nitrogen, and neon ion charges are represented in Figs. 2 — 4. The values of Φ_i between the experimental points were taken from curves representing the dependence of $\log(\Phi_{i+1}/\Phi_i)$ on log v, which were plotted from the experimental values of Φ_{i+1}/Φ_i; this is simpler than the dependence of Φ_i on v and permits more reliable interpolation. As an example Fig. 5 gives curves showing the dependence of $\log(\Phi_{i+1}/\Phi_i)$ on log v for oxygen ions. In order to obtain the dependence of Φ_i on v at low velocities for nitrogen and neon ions we used the experimental values of Φ_i in Ref. 2. For nitrogen, oxygen, and neon ions the error in the values of Φ_i between our experimental points does not exceed the errors in the measurements of Φ_i which were mentioned above. For boron ions, and also for nitrogen and neon ions at low velocities, the interpolation is less reliable and the error is apparently 2 — 3 times greater.

The velocity dependence of the degree of ionization $\bar{i}/Z$ in the different media is given in Fig. 6. The error in $\bar{i}/Z$ is about 1%.

In addition to the results already mentioned, we obtained the equilibrium distribution of helium ion charges at three different velocities, after passage through a celluloid film. These results are given in Table II.

Figures 2 — 4 show that the charge distribution in an ionic beam generally differs after passage through

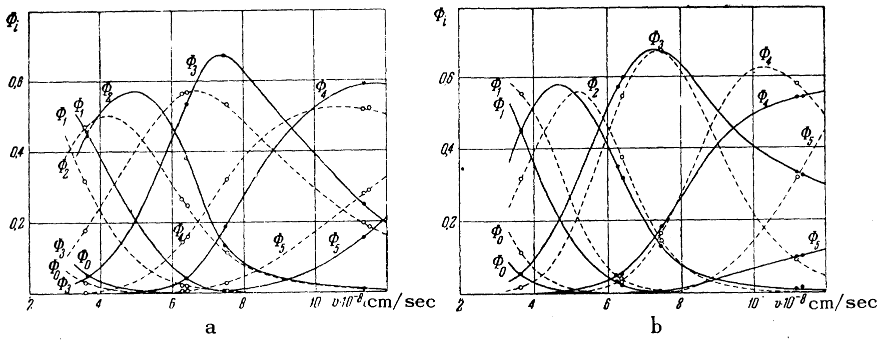

FIG. 2. Equilibrium distribution of boron ion charges after passage through a: ● — air, ○ — celluloid film; b: ● — argon, ○ — hydrogen

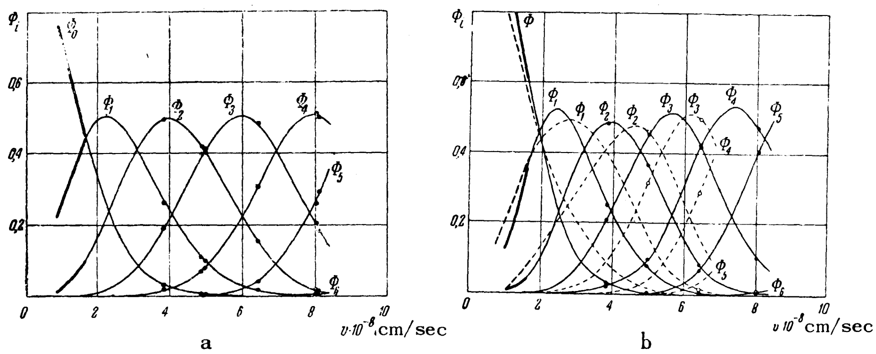

FIG. 3. Equilibrium distribution of nitrogen ion charges after passage through a: air; b: ● — argon, ○ — hydrogen. The heavier lines at low velocities represent results taken from Ref. 2

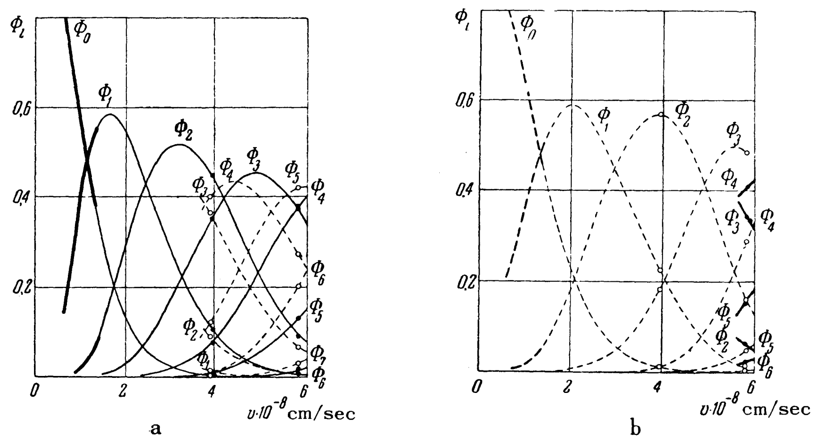

FIG. 4. Equilibrium distribution of neon ion charges after passage through a: ● — air, ○ — celluloid film; b: ● — argon, ○ — hydrogen. The heavier lines at low velocities represent results taken from Ref. 2

different media. There is a striking difference in the character of the velocity dependence of Φ_{i+1}/Φ_i after passage of the ions through solid and gaseous matter (Fig. 5). When boron and oxygen ions pass through a celluloid film, just as for ions of helium,[4] lithium,[5] and nitrogen,[7] the values of

$$k_i = \frac{d \log (\Phi_{i+1}/\Phi_i)}{d \log v}$$

vary little for velocities from 4 to 8×10^8 cm/sec, so that Φ_{i+1}/Φ_i shows a nearly power-law dependence on the velocity ($\Phi_{i+1}/\Phi_i \approx C_i v^{k_i}$). For all of the indicated ions $k_0 \sim 2.5-3$; for $i \sim Z/2$ the value of k_i is close to 4 and k_{Z-1} is around $4.5-5$. When boron, nitrogen, and oxygen ions pass through gases the values of k_i in the same velocity range are greatly increased. For example, in the case of oxygen in argon k_2, k_3, and k_4 increased from $3-4$ to $6-7$, which is almost double. At high velocities k_i is again reduced, as is shown by the data for boron ions.

$\mathrm{Log}(\Phi_{i+1}/\Phi_i)$ has different values for different gases at the same ion velocity, but in most cases the differences $\log(\Phi_i/\Phi_{i-1}) - \log(\Phi_{i+1}/\Phi_i) = 0.434/\sigma_i^2$ are identical within the limits of error. σ_i does not change greatly as the charge and velocity are varied. The dependence of σ_i on v and i is found to be especially slight for $i < Z-2$ in the case of nitrogen and oxygen ions. In the velocity range from 3.5 to 8×10^8 cm/sec the variation of σ_i for these ions is $\pm 7-10\%$. For $\sigma_i = \sigma =$ const we have the relation

$$\Phi_i = A \exp [-(i-i_0)^2/2\sigma^2],$$

where

$$i_0 = \bar{i} + \frac{\sigma^2}{2} \ln (\Phi_{i+1}/\Phi_{i-1}).$$

For not very small values of the mean charge $\bar{i}$ the value of A is close to $1/\sigma\sqrt{2\pi}$, while i_0 is close to $\bar{i}$ [when $\bar{i} > 1.5$ and $\sigma \approx 0.7$, we have $|\bar{i} - i_0| < 0.003$ and $|(1/\sigma\sqrt{2\pi}) - A| < 0.002$]. Therefore

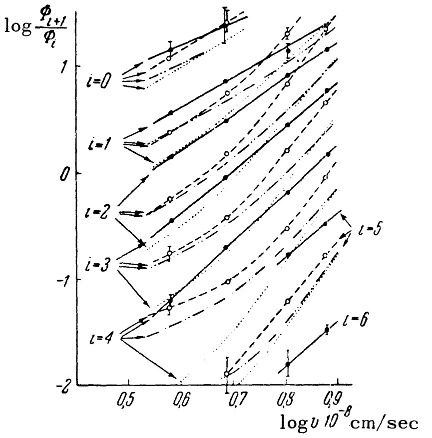

FIG. 5. Dependence of $\log(\Phi_{i+1}/\Phi_i)$ on log v for oxygen ions after passage through hydrogen (dots), air (dots and dashes), argon (dashes and open circles), celluloid film (solid lines and heavy dots).

our values of Φ_i can be represented approximately by the Gaussian function

$$\Phi_i(i-\bar{i}) \approx \Phi^0(i-\bar{i}) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(i-\bar{i})^2}{2\sigma^2}\right].$$

For boron, nitrogen, oxygen, and neon ions σ is 0.65, 0.80, 0.83, and 0.76, respectively. For nitrogen and oxygen ions with velocities from 3.5 to 7.5 × 10^8 cm/sec, the experimental values of $\Phi_i(i-\bar{i})$ do not differ from $\Phi^0(i-\bar{i})$ by more than 5% when $\Phi_i \geq 0.2$ (Fig. 7); when $\Phi_i \approx 0.1$ the difference is not greater than 15% and for $\Phi_i \sim 0.02$ it is about 30%. For boron ions in the velocity range 3.5 — 11 × 10^8 cm/sec the corresponding difference is 1.5 — 2 times greater and for neon ions it is 3 times greater than for oxygen ions. The values of Φ_i, after passage through a celluloid film, can also be represented approximately by a Gaussian function with

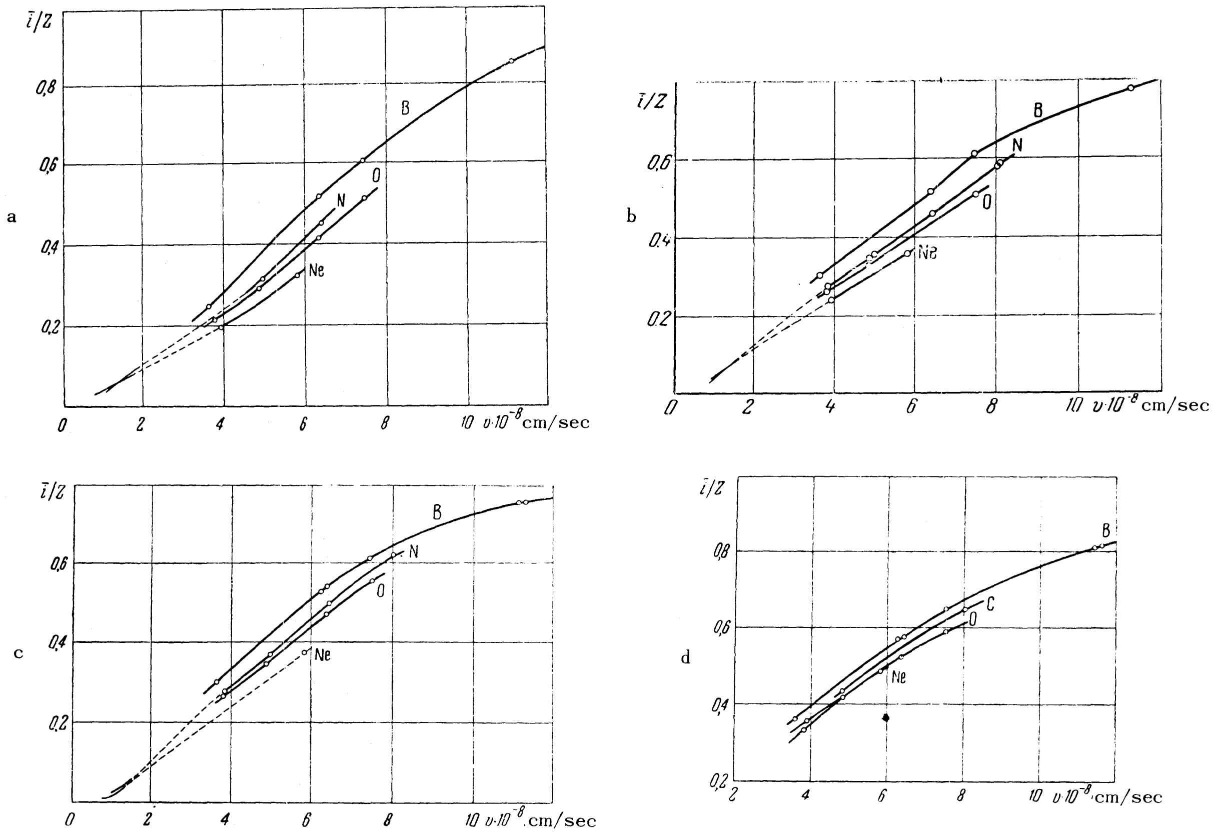

FIG. 6. The velocity dependence of the degree of ionization $\bar{i}/Z$ after the passage of ions through a — hydrogen, b — air, c — argon, d — celluloid film. The solid lines at low velocities represent results taken from Ref. 2

the same values of σ as for the passage of the ions through gases (for carbon ions $\sigma \approx 0.73$). But the difference between $\Phi_i(i-\bar{i})$ and $\Phi^0(i-\bar{i})$ in this case is 1.5 — 2 times greater than for the passage of the same ions through gases.

The following characteristics of $\bar{i}/Z$ can be noted for the passage of ions through different media:

1. For values of $\bar{i}/Z$ from 0.2 to 0.6 the mean charge in argon, for all of the ions investigated, is greater than the mean charge in hydrogen (by about 10 — 20%). The mean charge in air depends less strongly on velocity than the mean charge in hydrogen and argon. When $\bar{i}/Z \sim 0.3$ it is close to the

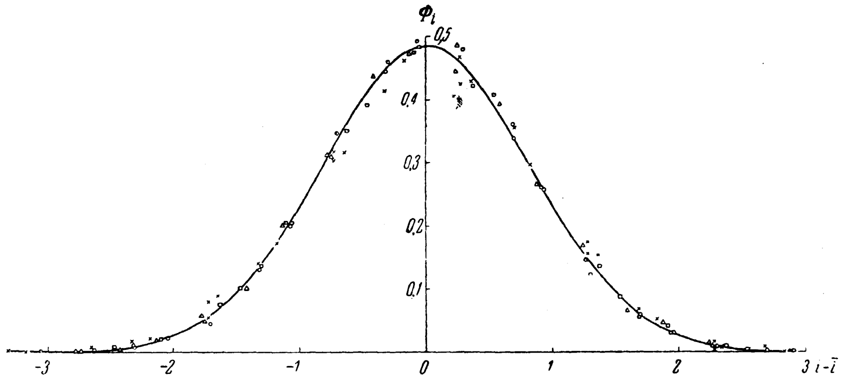

FIG. 7. Dependence of Φ_i on $i - \bar{i}$ for oxygen ions: ○ — in hydrogen, □ — in air, Δ — in argon, and × — in celluloid film. The solid line is a Gaussian curve with $\sigma = 0.83$

TABLE II. Φ_i and $\bar{i}$ for helium ions passing through celluloid film ~ 20 $\mu g/cm^2$ thick

$v \cdot 10^{-8}$ cm/sec	Φ_0	Φ_1	Φ_2	$\bar{i}$
6.0	0.017	0.368	0.615	1.60
8.0	0.002	0.145	0.853	1.85
18.3	~0.0001	0.005	0.995	1.995

mean charge in argon, but when $\bar{i}/Z \sim 0.5$ it approaches the value of $\bar{i}$ in hydrogen. For $\bar{i}/Z \sim 0.8$, the mean charge of boron ions is greatest in hydrogen, and smallest in argon. In hydrogen $\bar{i}$ is 10% greater than in air and 13% greater than in argon.

2. The mean charge of ions after passage through celluloid film in a broad range of velocities is greater than the mean charge of the ions in gases, and with increasing nuclear charge Z of the ions the excess of the mean ionic charge after passage through the film over the mean charge in air increases quite rapidly. While for boron ions with $\bar{i}/Z \sim 0.4$ this excess is ~ 15%, for neon ions it is ~ 30%. For the light group of uranium fission fragments ($Z = 38$) this difference is known[16] to be 40% and for the heavy fragments ($Z = 54$) it is 50%. Only at relatively high velocities with $\bar{i}/Z \sim 0.8$, for which data on boron ions are available, is the mean charge after passage through the film found to be 4% smaller than the mean charge in hydrogen.

3. The degree of ionization of the investigated ions in air, hydrogen, and argon can be represented within the range $\bar{i}/Z \sim 0.2 - 0.6$ as a different function of the parameter $v/v_0 Z^{\alpha}$ with $\alpha \approx 0.4$ for each gas. When $\bar{i}/Z$ is represented in the same form for the passage of ions through a celluloid film, the exponent α is not even approximately constant: for $Z \sim 5 - 6$ the value of α is close to 0.35, while for $Z \sim 8 - 10$ we have $\alpha \sim 0$.

4. DISCUSSION OF RESULTS

Our values for the mean charge of ions which have traversed a celluloid film are in good agreement with the mean charge of nitrogen ions given in Refs. 6 and 7.

The distributions of nitrogen and oxygen ion charges which are given by Stephens and Walker[10] can be compared with the corresponding distributions in the present work and in Refs. 6 and 7 if the ion velocities given in Ref. 10 are reduced by 8%. In view of this discrepancy, Reynolds et al. checked their ion velocity measurements by measuring the energy of recoil protons. In the present work and in Ref. 7, ion velocities were determined from the curvatures of particle trajectories in the magnetic field as calibrated with deuterons, and they cannot contain such a large error. In Ref. 10 the velocities were calculated from the frequency of the accelerating electric field between the dees and from the final trajectory radius, and may therefore be too high.

The mean charge of oxygen and neon ions in argon, as given in Ref. 9, is 5 — 7% below the mean charge of these ions in argon as shown in Fig. 6c, and is about 7% greater than the mean charge in hydrogen. In Ref. 9 no essential difference was found in the values of $\bar{i}$ for different gases, but the authors do not exclude the possibility of a difference on the order of 10%. Within this limit of accuracy, the results of Ref. 9 agree with ours.

The article by Korsunskii et al.[8] gives the charge distribution only for the charge portion of a beam of nitrogen ions passing through nitrogen. Since the values of Φ_0 are not given, it is best to compare values of Φ_{i+1}/Φ_i. The values of Φ_2/Φ_1 for ion energies of 0.8 — 1.0 Mev, are approximately 1.5 times lower than in our present work. This discrepancy may possibly result from the fact that in Ref. 8 the beam passed through a layer of gas only about one tenth as thick as ours. Under such conditions the charge distribution can differ appreciably from equilibrium, as shown by our measurements. The values of Φ_3/Φ_2

and Φ_4/Φ_3 agree with ours; the explanation for this may be that for charges which differ more from the mean charge the equilibrium value of Φ_{i+1}/Φ_i is reached in a thinner layer of gas. In both investigations the values of Φ_5/Φ_4 are apparently not very accurate, but agree in order of magnitude.

Existing theoretical calculations of the equilibrium distribution and mean charge yield values of Φ_i and $\bar{i}$ which differ considerably from the experimental results. Calculations of the equilibrium charge distribution in Refs. 11 and 12 give lower values of k_i (around 3). The calculated values of Φ_i are at the maximum ~ 20% below experimental values. The mean charge as calculated in Refs. 11 — 14 differs by 10 — 30% from experiment, and none of the methods used agrees better than the others with experiment. Bohr's formula $\bar{i} = Z^{1/3}v/v_0$ in Ref. 17 gives values of $\bar{i}$ which are 1.5 — 2 times higher than experiment.

Brunings, Knipp, and Teller,[15] in attempting to improve Bohr's approximate formula, assumed that an ion traversing matter retains electrons with orbital velocities that satisfy $v_e > \gamma v$. For the calculation of v_e, they used the statistical model of the atom; in one calculation they separated from the electronic cloud of the ion the energetically most easily removable electron, and in another calculation they separated the outermost electron. The two different calculations are represented by the coefficients γ_1 and γ_2, respectively. For γ_1 = const the dependence of $\bar{i}/Z$ on v in the interval $\bar{i}/Z \sim 0.3 - 0.6$ is stronger, and for γ_2 = const it is weaker than the experimental dependence for all of the substances investigated. With increasing velocity, the experimental coefficient γ_1 is reduced by 10 — 30% while γ_2 increases by 10 — 30%. According to Ref. 15, the degree of ionization $\bar{i}/Z$ in the same range must be a function of v/v_0Z^{α}, where for $Z \sim 6 - 10$ with γ_1 = const $\alpha \approx 0.55$ and with γ_2 = const $\alpha = 0.33$. Experiments show that for ions passing through gases $\bar{i}/Z$ actually can be represented as a function of v/v_0Z^{α}, and although the form of the function differs with the substance we have the approximately constant value $\alpha \approx 0.4$ within the indicated limits. Bohr's formula gives $\alpha = 2/3$. Thus the consistent use by Brunings et al. of the statistical atomic model gives a more nearly correct relation between $\bar{i}/Z$ and Z than does Bohr's formula.

The method of Brunings, Knipp, and Teller does not permit us to draw any conclusions regarding the different values of $\bar{i}$ in various media. Livesey's calculation[13] of the mean charge of nitrogen ions in air and solid matter does not give the correct ratio between the values of $\bar{i}$ in these media. According to these calculations, the mean charge in a solid is smaller than the mean charge in air. A qualitatively correct ratio between the values of the mean charge in different gases with $\bar{i}/Z \sim 0.5$ was obtained by Gluckstern,[11] who calculated $\bar{i}$ from the cross sections for electron loss and capture. However, the absolute values which he obtained for $\bar{i}$ are 20 — 30% below the experimental values.

Our relations between the values of the mean ionic charge in different media reflect the behavior of the cross sections for electron capture σ_c and loss σ_ℓ in these substances. The cross sections σ_c and σ_ℓ increase with the atomic number Z_m of the medium. Larger values of $\bar{i}$ in heavier gases for $\bar{i}/Z \sim 0.3 - 0.5$ ($v \sim 4 - 7 \times 10^8$ cm/sec) show that σ_ℓ increases with Z_m by a higher power than σ_c, and this is a qualitative confirmation of the approximate determination of σ_ℓ and σ_c based on the statistical model. According to these calculations[17,18] σ_ℓ is proportional to $Z_m^{2/3}$ and σ_c is proportional to $Z_m^{1/3}$. At high velocities σ_ℓ must be proportional to Z_m^2 and σ_c must be proportional to Z_m^5, so that in the lightest elements the mean charge must be larger. We obtained the corresponding ratio between the values of $\bar{i}$ in gases for boron ions with $v \sim 11 \times 10^8$ cm/sec. The relative increase of the mean ionic charge in air compared with hydrogen and argon for $v \le 4 \times 10^8$ cm/sec cannot be explained by the Thomas-Fermi statistical model and is apparently associated with the structural characteristics of atoms with unfilled shells, especially in the case of relatively large atomic dimensions, which must result in some increase of σ_ℓ.

The large value of $\bar{i}$ in solid matter compared with its values in gases for uranium fission fragments has been attributed by Bohr and Lindhard[19] to changes in σ_ℓ and σ_c resulting from ionic excitation. An analogous difference between values of $\bar{i}$ for lighter elements evidently has the same causes. A reduction of the mean electronic binding energy in an ion traversing solid substances must result not only in an increase of σ_ℓ but also evidently in a shift of the maximum of σ_ℓ as a function of v at low velocities. Since the cross sections for the simultaneous loss and capture of two electrons are considerably smaller than the corresponding cross sections for a single electron[18,20], for the most intense charge groups, at least, $\Phi_{i+1}/\Phi_i \approx \sigma_\ell(i)/\sigma_c(i+1)$, where $\sigma_\ell(i)$ and $\sigma_c(i+1)$ are the single-electron loss and capture cross sections for ions with charges i and i + 1, respectively. Thus

$$k_i = \frac{d \log (\Phi_{i+1}/\Phi_i)}{d \log v} \approx k_l(i) - k_c(i+1),$$

where

$$k_l(i) = d\log\sigma_l(i)/d\log v, \quad k_c(i+1) = d\log\sigma_c(i+1)/d\log v.$$

The shift of the maximum of σ_l towards lower velocities when ions pass through solids must lead to a reduction of k_l and consequently of k_i; this is observed experimentally. Thus the difference in the equilibrium charge distribution after ion passage through gases and film can be explained qualitatively by excitation of the ions. A more thorough discussion of this question requires more complete information concerning the cross sections for electron loss and capture.

In conclusion we consider it our duty to thank S. S. Vasil'ev for his comments during the discussion of our results, and to the cyclotron crew, especially G. V. Kosheliaev, A. A. Danilov, and V. P. Khlapov, for their assistance in the experimental work.

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