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EFFECT OF DENSITY AND AGGREGATE STATE OF TARGET ON THE CHARGE COMPOSITION OF LITHIUM, SODIUM, AND POTASSIUM ION BEAMS

L. I. PIVOVAR, L. I. NIKOLAĬCHUK, and F. M. TRUBCHANINOV

Physico-technical Institute, Academy of Sciences, Ukrainian S.S.R.

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We have measured the charge states of beams of lithium, sodium, and potassium ions after passage through vapors of magnesium and cadmium and condensed targets of magnesium, cadmium, silver, and celluloid. The measurements in the vapors were performed in the pressure range from 10^{-5} to 7×10^{-1} mm Hg. This permitted evaluation of the cross section for capture of an electron by Li^+, Na^+, and K^+ ions, and also determination of the contribution of excited atoms to the average charge of the ions. The results obtained confirm the applicability of the theory of Bohr and Lindhard[2] to ions lighter than uranium fission fragments. Comparison of the distributions of charges of ions in vapors and in films indicates a considerable difference in the ratios of the values of the cross sections for capture and loss of electrons as a function of the aggregate state of a given material. The measurements were performed in the energy range 20—150 keV.

INTRODUCTION

THE average charge of fission fragments after passage through gaseous and solid targets was determined for the first time by Lassen[1] in a study of the interaction of uranium fission fragments with matter. He observed that the average equilibrium charge of the fragments in traversal of metallic films turned out to be higher than the average equilibrium charge in passage through gases. Also studying the dependence of the average charge of uranium fission fragments on the gas pressure in the collision chamber, Lassen showed that for an increase of pressure over a certain range the average charge of the fission fragments also increases.

Bohr and Lindhard[2] suggested the following explanation of this effect of the density of the target material on the average equilibrium charge. In rarefied gases, even in cases when a fast particle remains in an excited state after a collision, for the duration of the mean free path it can undergo a radiative transition to the ground state. Such a transition is possible when the duration of the mean free path for the ion is greater than the lifetime of the excited state. In this case, capture and loss of electrons occur in the ground state of the atoms and ions. When the lifetime of the excited state of the particle becomes greater than the duration of its mean free path, the cross sections change in such a way as a result of the excitation energy that the average equilibrium charge of the ions increases. It is obvious that this is due to an increase of the probability of loss of electrons by particles which are in an excited state. As Bohr and Lindhard have pointed out, an increase of the average charge of the ions with increasing pressure can also be observed for just this reason.

Since the mean free path is several orders of magnitude smaller in condensed targets than in gaseous targets, we would naturally expect a corresponding increase in the average charge of the ions in solid targets in comparison with that in gaseous targets. This effect of target density on the average equilibrium charge of the ions should evidently occur for any type of ion, provided only that they do not have such large excited-state lifetimes that they are unable to undergo a radiative transition to the ground state during the mean free path even in rarefied gases.

We will discuss the data existing at the present time on the average equilibrium charge distributions in beams of accelerated particles which have passed through condensed and gaseous targets. In the case of hydrogen ions the difference in the value of average charge in solid and gaseous targets, over a wide range of velocities, does not exceed the difference between different gases.[3] It should be noted that this fact remains true both in cases of a contaminated surface of the film-target and in cases where the distorting effect of the target surface is excluded.[4]

For helium ions, comparison of the data obtained by Dissanaike[5] on films of beryllium, aluminum, and silver, and by Nikolaev et al.[6] in a celluloid film, with those of Allison[3] and Pivovar et al.[7] obtained in gases shows that in solid targets the average equilibrium charge of helium ions has a value intermediate between that for nitrogen and argon gaseous targets. In the energy interval from 130 to 1330 keV the difference in value of the average charge of helium ions in gaseous and solid targets does not exceed 3—6%.

In the case of lithium ions with energies of 20—150 keV the average charge of the ions after passage through carbon and celluloid films turned out to be even lower than for gaseous targets.[8] With increasing energy this difference decreases and in the region between 0.5 and 5 MeV the average charges of lithium ions in a celluloid film and in argon become practically the same.[9] However, for ions with atomic number $Z \geq 4$ on passage through a celluloid film the average charge of the ions over a wide range of velocities turned out to be already higher than in gases. This difference does not exceed 30% for ions with atomic number $Z < 10$. For ions with atomic number $Z = 10—36$ the difference in the values of the average charge in a celluloid film and in gases amounts to 60—130%, depending on the atomic number.[10]

A small excess in the value of effective charge has been observed in particular for fluorine ions in nickel and aluminum foils in comparison with gaseous targets by Roll and Steigert.[11]

Attempts to interpret uniquely the facts outlined above from the point of view of effect of target density on average ion charge encounter definite difficulties. In the first place it must be kept in mind that in all the cases described the phenomena being considered could be strongly distorted by the difference in chemical nature of the solid and gaseous targets used, and also by the effect of the surface of the films on the side from which the ion beam leaves the film. The latter fact may be the result primarily of contamination of the film surface.

With the aim of removing these distortions, Meckbach and Allison[12] performed measurements of the ratios of effective charges of helium ions on passage through gaseous and metallic cadmium targets. As is turned out, over the entire energy region studied from 150 to 920 keV the greatest difference in the value of effective charge between solid and vapor cadmium targets did not exceed 8%, i.e., did not exceed the difference between different gases or films. Furthermore, in the energy region above 600 keV the effective charge in cadmium vapor turned out to be somewhat higher than in the film. Although such distorting factors as difference in the chemical nature of the targets and the effect of the film surface were removed in this experiment, a possible difference in the energy states of the particles in the solid and gaseous phases still remains, and correspondingly a difference in the cross sections for capture and loss of electrons by fast ions.

An attempt to estimate the dependence of the average ion charge of nitrogen on the density of a gaseous target in the pressure range $4 \times 10^{-5}—5 \times 10^{-2}$ mm Hg was made recently by Nikolaev et al.[13] These authors made a comparison of the average charge of ions computed from the cross sections for capture and loss of electrons with the measured values, in which some increase in the average charge was observed with increase of target density. The difference obtained does not, however, exceed the experimental errors.

In the present work we describe the results of measurements of the charge distribution in beams of lithium, sodium, and potassium particles after passage through magnesium and cadmium vapors and thin films of magnesium, cadmium, silver, and celluloid. The pressure of the metallic vapors in the collision chamber was varied continuously over the pressure range $10^{-5}—7 \times 10^{-1}$ mm Hg. This broad range of pressure variation allowed us to make rough measurements of the cross sections for electron capture σ_{10} by singly charged ions of Li^+, Na^+, and K^+, and also to measure the values of the charge fractions in the pressure regions providing those concentrations of target atoms for which the average time between two collisions is much less than the time of a radiative transition.

The data of the present work, together with data obtained by us previously[14] in a measurement of the equilibrium charge fractions of beams of lithium, sodium, and potassium particles after passage through vapors of magnesium, cadmium, and zinc, permit a unique conclusion on the effect of density and aggregate state of the target on the values of the charge fractions of the ion beam and correspondingly on the ratio of cross sections for capture and loss of an electron by fast atoms and ions.

2. APPARATUS AND EXPERIMENTAL TECHNIQUE

The measurements were performed with an experimental setup described by us previously.[8,14]

Here we will dwell only on the changes and additions which were made to the equipment. In the first place, to improve the vacuum in the apparatus we installed between the vacuum chamber and the M-1000 oil diffusion pump a baffle trap cooled by liquid nitrogen. This allowed operation at residual gas pressures in the apparatus not exceeding 1×10^{-6} mm Hg. In the second place, in the vacuum chamber we installed tungsten evaporators with which we could evaporate fresh layers of the metals being investigated onto an exposed film directly before the measurement.

The substrates used were celluloid films 2—3 μg/cm^2 thick obtained by spreading a solution of celluloid in butyl acetate on a water surface. Deposition of thin films of magnesium and cadmium on celluloid substrates was performed in a separate apparatus. To obtain uniform and oxide-free films of these metals, we followed the well developed technique, which requires the preliminary deposition of a very thin layer of silver.[15] The celluloid substrates with a deposited layer of magnesium or cadmium were fastened to a drum along its periphery by means of ring mountings. By turning this drum we could place in the beam any of the films fastened to it, without destroying the vacuum. The targets were mounted in such a way that the metallic coverings were on the side from which the beam left the film.

The study of the effect of density of magnesium and cadmium vapors on the ratio of the charge fractions in the ion beams, and also the measurement of the electron capture cross sections by ions of Li^+, Na^+, and K^+ were performed in the collision chamber described previously.[14] The technique used to measure the values of the charge fractions was the same as that described previously.[14]

Special attention was devoted to measurement of the collision chamber temperature, since the temperature determines the pressure and hence the concentration of the metal vapor. The temperature was measured with chromel-alumel thermocouples calibrated with an accuracy of ±0.1°C. The coolest part of the collision chamber was determined from the temperature distribution over the chamber.

In the energy range investigated, we can neglect without large error the effect of loss and capture of two or more electrons and consider a system consisting essentially of two components resulting from neutral and singly charged particles. Therefore the cross sections for electron capture by singly charged ions were calculated according to the formula[3]

$$\sigma_{10} = \frac{F_{0\infty}}{nl} \ln \frac{F_{0\infty}}{F_{0\infty} - F_0}, \qquad (1)$$

where F_0 is the relative number of neutral particles for a given nl, n is the concentration of target atoms, l is the effective length of the collision chamber, and $F_{0\infty}$ is the relative number of neutral particles in the beam of equilibrium composition. The cross sections for electron loss σ_{01} by fast atoms of lithium, sodium, and potassium can obviously be determined in the same way or from the ratio between the equilibrium fractions $F_{0\infty}$ and $F_{1\infty}$.

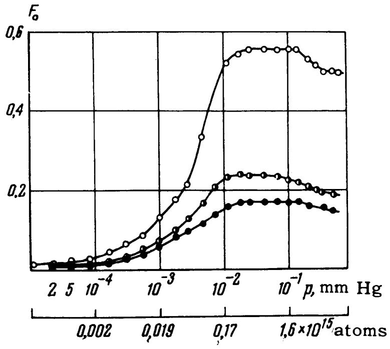

FIG. 1. Neutral component of the particle beam as a function of cadmium vapor pressure: ○ – Li ions, 96 keV; ◑ – Na ions, 96 keV; ● – K ions, 80 keV.

Although chemically pure metals were used as targets in this work, it is known that in evaporation of metals in vacuum, oxide films can be formed on the surface which appreciably reduce the rate of evaporation.[16] Therefore we must use the cross sections determined from the collision chamber temperature with a certain degree of caution. However, the results of studies made very recently by Greenbank and Argent[17] show that, at least at temperatures above 280°C, the vapor pressure over a cadmium surface always corresponds to the saturated vapor pressure, and in the case of magnesium the vapor pressure can be below the saturated vapor pressure by 1.6—2 times. Therefore the cross section values σ_{10} and σ_{01} measured in magnesium are too low by a factor of 1.6—2. In the measurements of charge fractions in the region of high concentrations of magnesium and cadmium vapor and after traversal of condensed targets, we always made observations of the energy loss of the primary ions. As a rule these losses did not exceed 1—3% in the vapors and 3—5% in the solid targets. The random errors in the charge fraction measurements are ±5%.

3. EXPERIMENTAL RESULTS AND DISCUSSION

A. Vapor targets. Figure 1 shows typical curves of the magnitude of the neutral component of the particle beam as a function of the pressure p of

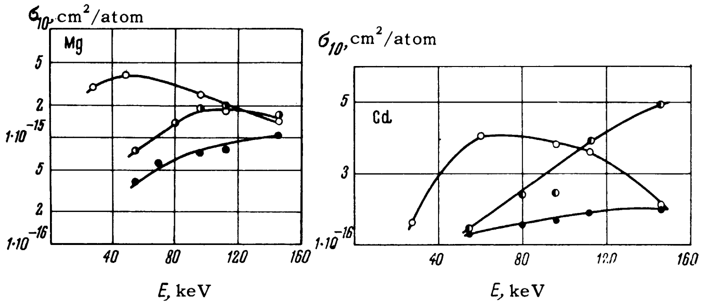

FIG. 2. Cross sections for electron capture by Li, Na, and K ions in magnesium and cadmium vapor as a function of energy: ○ – Li^+, ◑ – Na^+, ● – K^+.

cadmium vapor in the collision chamber. The same form of dependence $F_0 = f(p)$ was obtained for particles of lithium, sodium, and potassium in magnesium and cadmium vapors for a number of fixed values of ion energy. From these curves for several pressures we determined according to formula (1) the cross section σ_{10} for electron capture by singly charged ions. As it turned out, the spread in the values of σ_{10} determined for different pressures was small. The corresponding energy dependence of the cross sections for electron capture by Li^+, Na^+, and K^+ ions in magnesium and cadmium vapor is shown in Fig. 2.

As we can see from Fig. 1, the neutral component of the beam decreases in the high pressure region, i.e., the average ionic charge increases. This increase in average ionic charge with increasing vapor density obviously agrees with the suggestion of Bohr and Lindhard[2] of an increased probability of electron loss by an atom in an excited state, in proportion to the decrease in the average time of flight between two collisions. Let us consider the relations determining the target-particle concentrations at which the effect of density should be felt on the equilibrium charge distributions in the ion beam.

After a collision a fast particle may remain in an excited state. The reduction in the number of excited particles with time t due to radiative transitions is determined by the law of probability. We will set the time t equal to the mean time between two successive collisions of a fast particle; then the number of particles remaining in an excited state is determined by the expression

$$N_t = N_0 e^{-tv/\tau v} = N_0 e^{-\lambda/\tau v}, \qquad (2)$$

where v is the ion velocity, λ is the mean free path of the particle, and τv is the path length of the particle in an excited state.

In the case when $\lambda \gg \tau v$, the ratio N_t/N_0 is very small, i.e., during the time between two successive collisions a large fraction of the excited particles succeed in dropping to the ground state as the result of radiative transitions.

In cases when $\lambda \ll \tau v$, the approximate equality $e^{-\lambda/\tau v} \approx 1 - \lambda/\tau v$ is valid. Assuming that collision of a fast excited atom is accompanied by loss of one electron, we can write $\lambda = 1/n\sigma_{01}$, where n is the concentration of target atoms and σ_{01} is the cross section for electron loss by the excited atom. After substitution we obtain

$$N_t = N_0(1 - 1/n\sigma_{01}\tau v), \qquad (3)$$

where

$$n\sigma_{01}\tau v \gg 1. \qquad (4)$$

Thus, as can be seen from formula (3), for target-atom concentrations n satisfying the condition (4), the number of particles N_t remaining in an excited state between two successive collisions approaches the initial number N_0.

Let us consider as an example the change of N_t for an increase of $n\sigma_{01}\tau v$ from 0.2 to 10. For these two extreme values the quantity N_t is determined respectively by formulas (2) and (3) and varies from $0.12N_0$ to $0.9N_0$. It is evident that further increase in target density can affect N_t only very insignificantly. Therefore we can assume without great error that for $n\sigma_{01}\tau v = 10$ essentially the maximum contribution is reached to the process of electron loss as the result of excitation energy of the atoms.

As a check showed, the effect of target density on the formation of doubly charged ions in the equilibrium beam was absent in the energy and pressure regions investigated by us. Therefore the increase in the average equilibrium charge with increasing vapor concentration is due only to the change in ratio between the neutral and singly charged components of the beam.

Let us make a quantitative estimate of the values of target-atom concentrations at which the increase in average charge as the result of the excited states approaches the maximum possible value. We will proceed from the condition $n\sigma_{01}\tau v = 10$, i.e., $N_t = 0.9\ N_0$.

For the most intense spectral lines of lithium, sodium, and potassium atoms, the average lifetimes of the excited states are

$$\tau_{Li} = 6\cdot 10^{-8}\,\text{sec},\quad \tau_{Na} = 1{,}5\cdot 10^{-8}\,\text{sec},\quad \tau_K = 2{,}7\cdot 10^{-8}\,\text{sec}.$$

For the cases shown in Fig. 1, the cross sections for electron loss by fast atoms of lithium, sodium, and potassium in cadmium are

$$(\sigma_{01})_{Li} = 3{,}2\cdot 10^{-16}\,\text{cm}^2/\text{atom},\ (\sigma_{01})_{Na} = 8\cdot 10^{-16}\,\text{cm}^2/\text{atom},$$

$$(\sigma_{01})_K = 8\cdot 10^{-16}\,\text{cm}^2/\text{atom}.$$

(The cross sections for electron loss were determined from the electron capture cross sections σ_{10} and from the ratio of the equilibrium fractions $F_{0\infty}$ and $F_{1\infty}$.) Substituting the values of σ_{01}, τ, and v into the formula $n = 10/\sigma_{01}\tau v$, we obtain the cadmium atom concentrations n in the cases considered for lithium, sodium, and potassium, respectively:

$$n(\text{Li}) = 3\cdot 10^{15}\,\text{atom/cm}^3,\quad n(\text{Na}) = 9\cdot 10^{15}\,\text{atom/cm}^3,$$

$$n(\text{K}) = 7{,}4\cdot 10^{15}\,\text{atom/cm}^3.$$

It can be seen from Fig. 1 that the maximum contribution of excited particles to the average charge was clearly reached for lithium ions. The situation is not as satisfactory with sodium and potassium ions. Further increase of cadmium atom concentration was hindered by scattering of the fast particles and sharply decreasing intensities of the beams at the exit of the collision chamber. This fact led to additional errors in determination of the size of the charge components of sodium and potassium at high cadmium concentrations.

Figure 3 shows the relative increases in the singly charged components $\Delta F_1/F_{1\infty} = (F_1 - F_{1\infty})/F_{1\infty}$, measured for the maximum concentrations achieved by us of magnesium and cadmium atoms. It can be seen from the figure that the relative increase in the average ionic charge with increasing target-atom concentration depends strongly on the nature of these ions and atoms, and also on the energy of the fast particles.

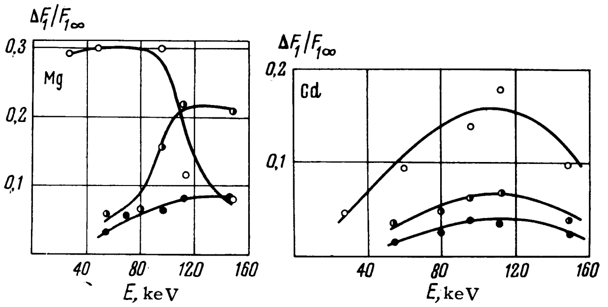

FIG. 3. Relative increase in the singly charged component as a function of ion energy in magnesium and cadmium vapors: ○ – Li ions, ◑ – Na ions, ● – K ions.

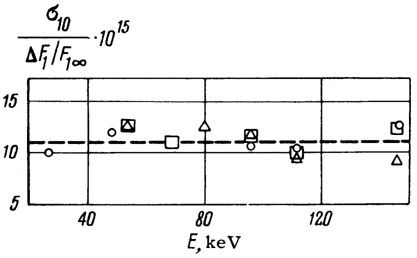

FIG. 4. The ratio $\sigma_{10}/(\Delta F_1/F_{1\infty})$ as a function of ion energy in magnesium vapor: ○ – Li ions, △ – Na ions, □ – K ions.

Comparison of the data given in Figs. 2 and 3 for magnesium allows us to draw the conclusion that there is a unique correspondence between the value of the electron capture cross section σ_{10} and the relative increase in the average charge $\Delta F_1/F_{1\infty}$ in the energy region studied. In fact, from Fig. 4 which shows the ratio $\sigma_{10}/(\Delta F_1/F_{1\infty})$ for lithium, sodium, and potassium ions in magnesium, it is evident that within the experimental errors this ratio remains constant and independent of the nature of the ions and their energy. This connection between the capture cross section and the average charge may be the result either of the fact that the fast excited atoms participating in the collisions are produced by capture of an electron by an ion in an excited state, or that the dependence of the excitation cross sections on ion energy is very weak in the range of velocities being considered.

The same tendency is observed also in the interaction of the ions studied with cadmium atoms, but as the consequence of large errors in determination of ΔF_1 the spread in the value of the ratio $\sigma_{10}/(\Delta F_1/F_{1\infty})$ is very large. The data obtained in this work on the dependence of the average charge on the density of the target gas unambiguously confirm the applicability of the theory of Bohr and Lindhard[2] to ions lighter than uranium fission fragments.

B. Condensed targets. To compare the equilibrium distribution of ionic charges in vapor and condensed targets having the same chemical nature, measurements were made in the present work of the charge fractions of lithium, sodium, and potassium ions after passage through films of magnesium and cadmium. To evaluate the effect of oxidation of the film surfaces and for comparison with previously obtained results, films of silver and celluloid were also used as targets.

Since films a few atomic layers thick already provide an equilibrium charge distribution, it is obvious that a layer of impurities on the film surface

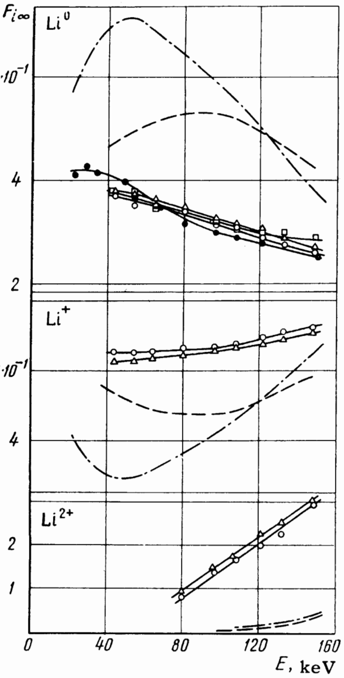

FIG. 5. Equilibrium distribution of charges in a beam of Li ions as a function of energy: △ – in magnesium film, ○ – in cadmium film, □ – in silver film, ● – in celluloid film; dot-dash curve – in magnesium vapor according to our previous work, [14] dashed curve – in cadmium vapor according to our previous work [14] (for Li^{2+} the ordinate scale is multiplied by 1×10^{-2}).

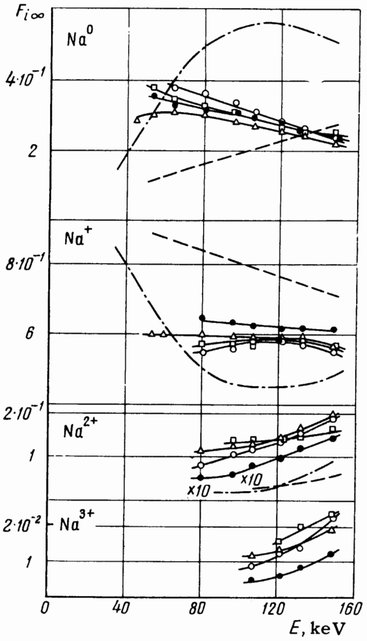

FIG. 6. Equilibrium distributions of charges in a beam of Na ions as a function of energy: △ – in magnesium film, ○ – in cadmium film, □ – in silver film, ● – in celluloid film; dot-dash curve – in magnesium vapor according to our previous work, [14] dashed curve – in cadmium vapor according to our previous work. [14]

may have a distorting effect. Therefore we took measures to avoid these impurities. Following Phillips,[4] we evaporated a clean layer of the metal being studied onto the substrate immediately before measurement of the charge fractions.

Since the evaporation of the metals was carried out at residual gas pressures up to 10^{-6} mm Hg and in the experimental apparatus proper, it was necessary to convince ourselves that the freshly evaporated films of magnesium and cadmium were preserved in a sufficiently pure state at least for the length of time necessary for making the measurements. In this connection we measured the dependence of the charge composition of the beam on time, starting at the time of evaporation of the metallic film. On aging of the freshly evaporated layers of magnesium and cadmium for 48 hours in vacuum, we observed changes in the fractions $F_{0\infty}$, $F_{1\infty}$, and $F_{2\infty}$ of no more than 3%. Here the fractions $F_{0\infty}$ and $F_{2\infty}$ were greater for the freshly evaporated films than in the case of long exposure. The changes in charge composition with time of aging were observed for all ions. The results of measurements of equilibrium charge fractions as a function of the energy of Li^+, Na^+, and K^+ ions for solid targets of magnesium, cadmium, silver, and celluloid are shown in Figs. 5—7. These same figures show for comparison the results of measurements of charge fractions in magnesium and cadmium vapors taken from our previous work.[14] It can be seen from the figures that the values of the corresponding fractions in the different solid targets are not greatly different. However, in the vapor targets this difference is considerable and depends on the energy and nature of the ions.

In the region of velocity studied for sodium and potassium ions, the magnitudes of the singly charged components in the solid targets are distinctly lower than in the vapors of the same materials. This, it would appear, is inconsistent with our ideas about the effect of density on the charge composition of the beam. However, this result becomes understandable if we proceed from the fact that the solid and gaseous states of the same material have considerably different values of electron capture and loss cross sections. Comparison of the data shown in Figs. 5—7 shows that with increasing ion velocity the ratios of electron loss and capture cross sections change in different ways in the solid and vapor targets. There are regions of ion velocities where the fractions $F_{1\infty}$ in solid targets are larger than in vapor targets (Figs. 5, 6).

Even more evident is the difference in the cross sections for different aggregate states in comparison of the charge fractions corresponding to a

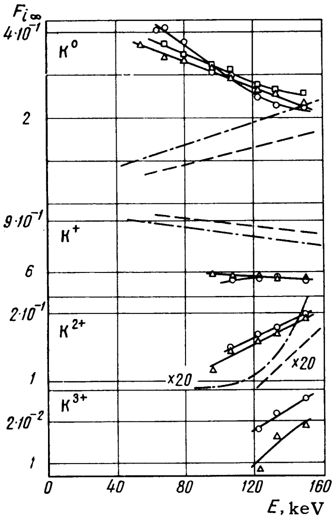

FIG. 7. Equilibrium charge distributions in a beam of K ions as a function of energy: △ – in magnesium film, ○ – in cadmium film, □ – in silver film; dot-dash curve – in magnesium vapor according to our previous work, [14] dashed curve – in cadmium vapor according to our previous work. [14]

higher multiplicity of ionization. For example, the fraction $F_{2\infty}$ in the solid targets is greater than in the vapor targets by factors of ten. This large difference cannot possibly be explained only by the effect of target density. As follows from the theory of Bohr and Lindhard[2] and from the results of the present work presented above, the limiting value of the charge fractions with increasing density of the medium cannot exceed a few tens of percent.

In conclusion it should be noted that the data obtained allow us to draw conclusions only on the difference in the ratios between the electron loss and capture cross sections by ions in solid and gaseous targets. However, the interesting question of the magnitude of these cross sections in solid targets remains completely open at the present time.

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