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ELECTRON CAPTURE AND LOSS BY HEAVY IONS PENETRATING THROUGH MATTER

BY

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§ 1. Introduction.

The phenomena connected with the penetration of high speed particles through matter have been a most important source of information about atomic processes. The discovery of nuclear fission, which made it possible to study the behaviour of swift ions with great masses and charges, has in this respect revealed many new interesting features, especially as regards the capture and loss of electrons by such ions. Capture and loss phenomena were, as is well known, first observed for α-rays, and have recently received renewed attention through the study of the tracks in photographic emulsions of highly charged ions of cosmic origin, penetrating into the upper regions of the atmosphere. Still, experiments on the stopping and ionizing effects of fission ions, and especially direct measurements of the charge of the ions during their passage through gases and solids, offer so far the most detailed and varied evidence as regards electron loss and capture by heavy ions.

In a previous treatise[1], a general survey of the theoretical interpretation of the effects accompanying the penetration of atomic particles through matter has been given. In particular, it was attempted to account for the peculiar law which governs the energy loss of fission ions along their path by estimating the ion charge which, on account of the displacement of the balance between electron capture and loss, diminishes gradually with decreasing velocity. While the stopping and ionization effects in the beginning of the path depend primarily on collisions with the electrons in the atoms of the medium penetrated, nuclear collisions become of decisive importance at the end of the path. On the assumption that, irrespective of the substance through

[1] N. Bohr (1948). This paper, in which also a survey of the earlier literature is given, will in the following be referred to as I.

which it passed, the number of electrons carried by a fission ion of given nuclear charge was simply related to its velocity, it seemed possible to account approximately for the experimental evidence then available.

In recent years, however, important new information as regards the charge of fission ions in different materials has been obtained by the continued thorough investigations of N. O. Lassen[1]. Thus, the measurements of deflections of ion beams in magnetic fields disclosed not only a systematic deviation from the charge values previously estimated from stopping and ionization effects in gases (Lassen, 1949), but revealed an unsuspected large difference between the average charge of fission ions when emerging from solids and the charge of the ions of the same velocity when passing through gases. In gases, the magnetic deflections of the ion beams also exhibited a smaller, but distinct increase in the average charge with gas pressure. The detailed study of the gradual adjustment of the charge of the ions emerging from solids into rarefied gases allows, moreover, as shown by Lassen, to derive direct estimates of the cross sections for electron capture in collisions with the gas atoms.

The variation of ion charge with the density of the penetrated material permits several conclusions as to the mechanism of the collision processes determining the balance charge. Thus, the dependence of the average charge of swiftly moving heavy ions on the pressure of the gas through which they pass shows clearly that in the balance between loss and capture we may not, as in previous discussions, consider only processes by which electrons are removed from or captured in the ground state of the ions, but also processes involving excited states of a lifetime comparable with the intervals between successive collisions with the gas atoms. The remarkable difference between the average ion charge in gases and solids further indicates the occurrence of adjustment processes in excited ion states, of lifetimes essentially shorter than those of the radiative transitions.

In the present paper, it is attempted on the basis of simple arguments to give a comprehensive interpretation of the pheno-

[1] A survey of the results of these investigations is given in a dissertation: N. O. Lassen, On the total charges and the ionizing power of fission fragments, Copenhagen 1952.

mena associated with the passage through matter of highly charged ions. To this purpose, we consider first, in § 2, some general features of the balance between the loss and capture processes, with special reference to the fluctuations in ion charge, and the gradual adjustment of the average charge of ions emerging from solids into gases. On the basis of a simplified statistical description of the constitution of ions carrying many electrons we discuss, in § 3, some immediate conclusions to be drawn from the measurements of average ion charge and from stopping and ionization effects. In § 4, main features of the mechanism of electron loss and capture by heavy ions in collisions with atoms are considered, and it is attempted to derive approximate estimates for the cross sections of such processes, especially as regards dependence on ion charge and velocity, and the atomic number of the substance penetrated. On the basis of these estimates, a comparison with experimental evidence as regards the charge of fission ions in gases at low pressures is given in § 5. Finally, in § 6, the effect of the residual ion excitation is considered in connection with the observations on the variation of the ion charge with gas pressure, and its abnormally high value in solids.

On the publication of this paper, which due to various circumstances has been delayed, but parts of which have been reported at various conferences in the last years, we want to acknowledge our indebtedness to Dr. N. O. Lassen for many illuminating discussions during progressive stages of his experimental researches and of our theoretical considerations. We are also indebted to Dr. G. I. Bell, who before publication kindly made the results of his interesting studies of the loss and capture mechanism available to us.

§ 2. General features of the balance between loss and capture by heavy ions.

The problem of electron capture and loss by heavy ions presents features essentially different from those exhibited by swift α-particles or protons where, due to the smallness of the ratio between the cross section for capture by the bare nucleus and the loss cross section for an electron attached to it, the nucleus will carry an electron only during intermediate short intervals which, together, amount to a small fraction of the path. In the case of heavy ions like fission fragments, however, the nucleus will along the whole path carry a large number of electrons which, due to continual capture and loss, fluctuates around an average value determined by the velocity and nuclear charge of the ion and the properties of the medium.

Let us, for simplicity, consider a beam of ions penetrating through a gaseous medium of a density so low that the ions between collisions will practically all have returned to their ground state. The state of the beam as regards the effects of the collisions is therefore fully specified by the number $N(\tau)$ of ions carrying τ electrons. Disregarding, for the moment, loss and capture processes in which more than one electron is involved, we find thus, for the rate of change of $N(\tau)$, within an interval of the path where the velocity may be regarded as constant,

$$\left.\begin{aligned}\frac{dN(\tau)}{dx} &= \varrho\{N(\tau-1)\cdot\sigma_c(\tau-1)\\ &\quad + N(\tau+1)\cdot\sigma_l(\tau+1) - N(\tau)\cdot(\sigma_c(\tau)+\sigma_l(\tau))\},\end{aligned}\right\} \qquad (2.1)$$

where ϱ is the number of gas atoms per unit volume, $\sigma_c(\tau)$ the cross section for capture of an electron by an ion carrying τ electrons before the collision, and $\sigma_l(\tau)$ is the cross section for loss of an electron by such an ion. For the rate of change of the average number of electrons, $\bar{\tau} = \bar{\tau}(x)$, carried by the ions, we get from (2.1) by simple summation

$$\frac{d\bar{\tau}}{dx} = \frac{d}{dx}\cdot\frac{\sum\tau N(\tau)}{N} = \frac{\varrho}{N}\cdot\sum N(\tau)\cdot(\sigma_c(\tau)-\sigma_l(\tau)), \qquad (2.2)$$

where N is the total number of ions in the beam.

In a beam of heavy ions carrying many electrons the distribution of τ around the mean value will extend over several units and, therefore, a strict application of (2.2) demands a detailed knowledge of the dependence of the cross sections on the number of electrons in the ion. However, the summation in (2.2) is easily performed on the assumption that, in the interval in question, both σ_l and σ_c vary slowly and linearly with τ. We may then write

$$\left.\begin{aligned}\sigma_c(\tau) &= \Omega\cdot(1+\alpha_c\cdot(\tau-\omega)),\\ \sigma_l(\tau) &= \Omega\cdot(1+\alpha_l\cdot(\tau-\omega)),\end{aligned}\right\} \tag{2.3}$$

where α_c and α_l are constants small compared with unity, and ω is the value of τ for which the capture and loss cross sections have equal magnitude, Ω. Introducing the expressions (2.3) into (2.2) we thus get

$$\frac{d\bar{\tau}}{dx} = -\varrho\Omega\cdot(\alpha_l-\alpha_c)\cdot(\bar{\tau}-\omega), \tag{2.4}$$

and by integration

$$\bar{\tau}(x) = \omega + (\bar{\tau}(x_o)-\omega)\cdot\exp(-\varrho\Omega\cdot(\alpha_l-\alpha_c)\cdot(x-x_o)) \tag{2.5}$$

for the average electron number $\bar{\tau}(x)$ at the point x in a beam with a given value for $\bar{\tau}$ at the point x_o.

In a corresponding way, we derive from (2.1) and (2.3)

$$\left.\begin{aligned}\overline{\Delta\tau^2}(x) = \frac{1}{\alpha_l-\alpha_c} + \left\{\overline{\Delta\tau^2}(x_o) - \frac{1}{\alpha_l-\alpha_c}\right\}\\ \cdot\exp(-2\,\varrho\Omega\cdot(\alpha_l-\alpha_c)\cdot(x-x_o))\end{aligned}\right\} \tag{2.6}$$

for the average square fluctuation of the electron number at the point x. For large values of $(x-x_o)$, where the second term in (2.6) vanishes, the fluctuations will thus depend only on $\alpha_l-\alpha_c$, and the distribution around the average will be Gaussian with a width at half maximum equal to $2.35\cdot(\alpha_l-\alpha_c)^{-\frac{1}{2}}$.

In these simple calculations it is assumed that in every capture and loss process only one electron is removed from or transferred to the ion. Still, in the actual cases, especially in encounters with heavy atoms, there is a considerable probability that several electrons are lost or captured by the ion. However, such effects can easily be included in the above description by introducing in (2.1) further terms corresponding to cross sections $\overset{n}{\sigma}_l(\tau)$ and $\overset{n}{\sigma}_c(\tau)$ for collisions by which the electron number τ is changed by n units. Writing thus, with the same approximation as in (2.3),

$$\left.\begin{aligned} \overset{n}{\sigma_l}(\tau) &= \Omega_n \cdot (1 + \overset{n}{\alpha_l} \cdot (\tau - \omega_n)), \\ \overset{n}{\sigma_c}(\tau) &= \Omega_n \cdot (1 + \overset{n}{\alpha_c} \cdot (\tau - \omega_n)), \end{aligned}\right\} \quad (2.7)$$

we find by the same procedure formulas for the average charge and for the fluctuations corresponding to (2.5) and (2.6) if only Ω, $\Omega \cdot (\alpha_l - \alpha_c)$, and $\Omega \cdot (\alpha_l - \alpha_c) \cdot \omega$ are replaced by $\sum_n \Omega_n \cdot n^2$, $\sum_n \Omega_n \cdot n \cdot (\overset{n}{\alpha_l} - \overset{n}{\alpha_c})$, and $\sum_n \Omega_n \cdot n \cdot (\overset{n}{\alpha_l} - \overset{n}{\alpha_c}) \cdot \omega_n$, respectively. Thus, collisions involving a change of the electron number by several units may in particular influence the fluctuations, but as long as the value of n in the frequent collisions remains small compared with the average fluctuations, the equilibrium distribution will still be of approximately Gaussian type.

When considering the balance and fluctuations of the ion charge in media of greater density, where a considerable part of the ions, if not all, will remain in excited states between collisions, further considerations are necessary, since the cross sections for loss and capture may to a considerable extent depend on the excitation of the ion. Reckoning with suitably defined mean values for the loss and capture cross sections, depending on the actual degree of excitation of the ions, it is possible, however, to treat the problem in the same simple manner as above. The question of excited ion states may even have to be taken into account as regards balance between loss and capture for α-rays, but in this case the effect will in general be of minor importance due to the small electron binding in excited states, contrasting with the properties of ions carrying many electrons, where the excitation potentials may be several times smaller than the ionization potentials.

For fission ions escaping into vacuum from a solid surface, magnetic deflections permit measurements of the charge of the individual ions at a definite point of the path. In a gaseous medium, however, the continual change of ion charge, due to electron loss and capture, allows only to determine the average charge over a considerable part of the path. Still, by varying the gas pressure in the deflection chamber, Lassen was able to study in detail the gradual decline in average ion charge from the values in solids until balance in the gas is reached. The decline in charge is at first very rapid, showing a preponderance of electron capture over loss, but diminishes gradually and, in agreement with expectations, the average ion charge approaches a flat minimum through an approximately exponential slope (cf. LASSEN, 1950, fig. 2). The experiments on ion deflection in vacuum give not only values of the average charge higher than in gases, but exhibit characteristic charge fluctuations with approximate Gaussian distribution (cf. LASSEN, 1950, fig. 1). Notwithstanding the different conditions for the ions passing through solids, these fluctuations give, as we shall see, information about the dependence of the capture and loss cross sections on ion charge, supplementing the deductions which can be drawn from the gradual adjustment of the average charge of the ions emerging into gases.

§ 3. Approximate description of ion constitution.

A rigorous treatment of the collisions between highly charged ions and atoms presents us with complicated problems. An approximate account of the collision effects may, however, be obtained by means of a simplified description of atomic constitution (cf. I, § 3.5), in which the binding of the electrons is defined by the simple concepts of orbital extensions and velocities, using as a measure

$$a_0 = \frac{\hbar^2}{me^2} \quad \text{and} \quad v_0 = \frac{e^2}{\hbar}, \tag{3.1}$$

representing the "radius" and "velocity" of the electron in the ground state of the hydrogen atom. For an electron in an ion or atom we introduce in a similar way a radius a, characterizing the extension of the orbital region, and a velocity v, defined by

$$I = \frac{1}{2} mv^2, \tag{3.2}$$

where I is the binding energy. For an atom or ion with nuclear charge Z we thus write

$$a = a_0 \cdot \frac{\nu^2}{n}, \quad v = v_0 \cdot \frac{n}{\nu}, \tag{3.3}$$

where ν may be interpreted as the effective quantum number of the binding state, and $Z—n$ is the number of electrons with orbital radius smaller than a and, consequently, velocities larger than v.

For the ground state of an atom, ν will increase from a value close to unity for the most firmly bound electrons to a broad maximum and, finally, for the outermost atomic electrons, decline again to values of the order 1. For atoms containing many electrons, the maximum of ν will with close approximation be equal to $Z^{\frac{1}{3}}$, and from (3.3) we therefore get

$$dn = Z^{\frac{1}{3}} \cdot \frac{dv}{v_o} \tag{3.4}$$

as an approximate expression for the velocity distribution of the larger part of the electrons bound in the ground state of a heavy atom. The excitation of the atom demands the transfer of one or more electrons from the normally occupied states into unoccupied higher energy states. In the neutral atom such processes will for every electron require an energy exchange of the same order as the binding energy I, though in the case of inner electrons, part of this energy may be released in subsequent readjustment processes resulting in the excitation of other electrons and even in their ejection from the atom. In actual collision processes, a separation in well-defined stages is, however, limited and demands a closer comparison of the effective duration of the encounter and the times involved in the dynamics of the atomic processes.

The simplified description applies also approximately to the ground state of heavy ions of a total charge Z^*, corresponding to a considerable fraction of the nuclear charge. Still, since the maximum value of ν is not reached until $Z—n$ exceeds $Z/2$, it is essential for the applicability of formula (3.4) that Z^* is somewhat smaller than half the nuclear charge. As regards the excited states of highly charged ions, the situation is, moreover, in several respects different from that of neutral atoms, due to the presence of numerous unoccupied quantum states with comparatively strong binding. In fact, if by ν^* we denote the effective quantum number for the most loosely bound electrons in the ground state of the ion, with ionization potential I^*, the energy required for a larger part of the possible excitation processes will only be of the same order as I^*/ν^*.

For heavy ions we must in general reckon with a distribution of the excitation over several electrons. Not only will in actual collision processes often more than one electron be initially ex-

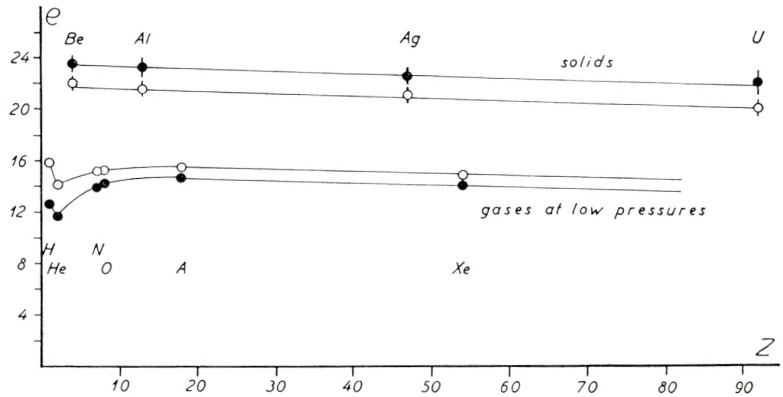

Fig. 1. (LASSEN, 1951 a, fig. 12). Average balance charge of fission ions with initial velocities, in solids and in gases at low pressures. The full circles refer to the heavy fragment group ($Z = 54$, $V = 4\,v_0$), and the open circles to the light group ($Z = 38$, $V = 6\,v_0$).

cited, but redistributions of the excitation over the electrons can even in the case of less violent encounters take place in immediate succession of the collisions. If the total surplus energy of the ion exceeds I^*, the result will be electron ejection within an interval very short compared with the limitation of the lifetime due to radiation processes. Also for the estimation of the lifetime of the excited ions and their properties, it is essential to bear in mind that an excitation energy below I^* will ordinarily be distributed over several electrons.

For orientation as regards the values of Z^* of swift heavy ions, a survey of LASSEN's measurements of the charge of fission ions at the beginning of the path in solid materials and in gases at low pressures is given in Fig. 1. It is seen that, apart from some interesting anomalies in the lightest gases, the ion charge is nearly independent of the atomic number of the gas for both groups of fission ions. The same applies to the ion charge in solids, notwithstanding the remarkable difference from gases as regards absolute values, and the peculiar inversion of the relative charge values of the two groups of fission ions.

The explanation of such specific features will of course demand a closer examination of the collision processes between the ions and the atoms of the penetrated substances and, in particular, of the state of the ions at the beginning of the encounters. For a preliminary discussion it may, however, be reminded that cursory considerations of the competition between electron capture and loss by ions in their ground state lead to the conclusion that, in balance, the velocity v^* of the most loosely bound electrons in this state of the ion should be nearly equal to the ion velocity V. According to (3.4), this gives

$$Z^* = Z^{\frac{1}{3}} \cdot \frac{V}{v_0} \tag{3.5}$$

as a rough estimate of the ion charge in balance (cf. I, § 4.4).

This estimate actually coincides closely with Lassen's direct measurements of the average charge in gases at low pressures, for the heavier group of fission ions. In fact, for $V = 4\,v_0$ and $Z = 54$, we get from (3.5) the value $Z^* = 15$. For the light group of fission ions ($V = 6\,v_0$, $Z = 38$), however, we would from (3.5) get $Z^* = 20$, while the measured value for the average charge is about 16. Quite apart from the question of the basis for a comparison of absolute charge values, the apparent discrepancy in the relative values is easily explained by remembering that (3.4) is applicable only in cases where Z^* is somewhat smaller than $Z/2$. This condition is amply fulfilled for the heavier ion group, but not for the lighter fission ions, with the consequence that $Z^{\frac{1}{3}}$ in (3.5) must be replaced by a somewhat smaller value of ν.

Such a difference between the two ion groups is also clearly revealed by the stopping and ionization effects of fission ions penetrating through gases. In Fig. 2 are, as an illustration, reproduced Lassen's results as regards the energy loss along the path in argon for the two groups. As will be seen, the curves are composed of two parts, corresponding to ion velocities large and small compared with v_0, and in which the stopping effects are due mainly to electronic and nuclear collisions, respectively. For the heavier ion group, the energy loss decreases linearly in the first part of the range. But for the lighter group, anomalies are exhibited at the beginning of the path, and the linear descent appears only after the velocity and the ion charge have decreased considerably from their initial values. As mentioned in I (§ 5.3), it follows from simple theory of energy loss by charged particles[1] that, in heavier gases, a linear decrease in energy along the path

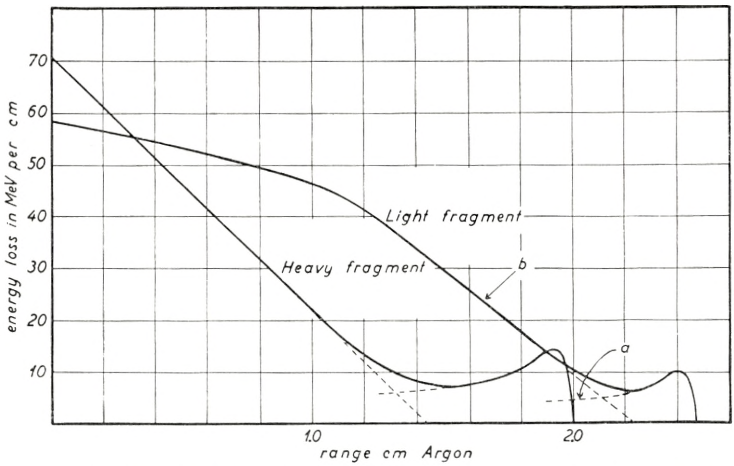

Fig. 2. (Lassen, 1949, fig. 23). Energy loss along the path of fission ions in argon. In the last part of the path, beyond the minimum, the energy loss by nuclear collisions is dominating. The magnitude of the separate contributions (a and b) from nuclear and electronic collisions is indicated by the dotted lines.

implies a proportionality between Z^* and V, corresponding to (3.5). While, for the heavier group of fission ions, this relation apparently applies for a large part of the range, $Z^{\frac{1}{3}}$ must for the lighter ion group evidently be replaced by a factor which

[1] This theory is especially developed in the case of particle charges and velocities for which quantum mechanical perturbation methods apply with high approximation. Recently, it has been shown (cf. Lindhard and Scharff, 1953) that it is possible, by means of a simple statistical treatment of atomic structures, on this basis to account for the stopping power over a wide region of atomic numbers and particle velocities. As discussed in I, special considerations are necessary in the case of highly charged particles where the conditions of perturbation theory are not fulfilled. The estimate given in I (§ 3.5) of the stopping power of a heavy atom needs, however, a certain correction. In fact, if in this estimate the dynamics of the electron binding is taken into account on the lines used by Lindhard and Scharff, the resulting stopping power will, like for α-rays in the same velocity region, not only be approximately proportional to the square of the ion charge and inversely proportional to ion velocity, but will also vary closely as the square root of the atomic number of the penetrated material.

in the beginning increases significantly with decreasing particle velocity.

As regards more quantitative estimates of the average ion charge from stopping and ionization effects of heavy ions, various complications have to be taken into account. Actually, earlier estimates of the ion charge from ionization in gases, based on the penetration theory of point charges, led to results almost as high as those obtained by direct charge measurements of the ions emerging from solids into vacuum. To explain this discrepancy, it is necessary to take the complex structure of the ion into account. In fact, in close collisions the atomic electrons will penetrate into the interior of the ion, where the effective nuclear charge is considerably higher than Z^*. The correction was in I (§ 4.4 and § 5.3) deemed to be insignificant because the collision diameter b, which in the stopping formula appears as an effective minimum impact parameter, is just equal to the diameter of the ion. However, the contribution of close collisions to the stopping effect is relatively large for fission ions, since the semi-adiabatic limit to the impact parameter in more distant encounters is only a few times larger than b. This circumstance makes an accurate evaluation of the stopping power difficult, but a simple calculation shows the correction due to ion structure to be of the order of magnitude required to explain the differences between the earlier estimates of the charge of fission ions in gases and the direct charge measurements.

§ 4. Mechanism of electron loss and capture.

In encounters between highly charged ions and neutral atoms, considerable changes in electron binding may take place particularly in the atom, where the more loosely bound atomic electrons at an early stage of the collision will be greatly influenced by the strong field around the ion. The transfer of energy accompanying the excitation and ionization of the atoms will, in fact, be the main source of energy loss of the ions. In the collisions, however, also processes can take place resulting in an excitation of the ion, or a change of ion charge due to electron capture and loss. A rigorous treatment of these processes presents a problem of great complication, but, due to the circumstance that the binding states in the ion involved in electron loss and capture are specified by high quantum numbers, simple mechanical considerations can be used in approximate treatments, and especially for the survey of the essential features of the mechanism of the different processes.

In the loss process, it is a question of a transfer of energy to ion electrons in the collision sufficient for electron escape. Due to the smallness of the forces acting between neighbouring electrons in the ion compared with the total ion field, we may, in considering such energy transfer, in first approximation examine separately the influence on the binding of individual electrons under the action of the forces to which they are exposed during encounters with atoms. In estimating these forces, however, we may only for light atoms compare the collision with separate impacts of the nucleus and the atomic electrons. For heavier atoms, where the orbital velocities of part of the electrons are larger than the particle velocity V, we must take into account that the charges of these electrons during the collision will effectively screen the charge ze of the nucleus, together with which they will act as an atomic core of a total charge number z^*, approximately equal to $z^* = z^{\frac{1}{3}} (V/v_0)$, corresponding to (3.5). Since the electrons more loosely bound to the atomic nucleus, due to their small charge and mass, are not able individually to transfer energy to the ion of the magnitude required, the main contribution to the loss process arises from the direct action of the bare nucleus in light atoms, and the atomic core in heavier atoms.

In order to estimate the loss cross section, we recall that the cross section for energy transfer greater than T in a collision between a free electron at rest and a heavy particle with charge z^*e and velocity V is given by the well-known formula (cf. I, § 3.1)

$$\sigma = 2\pi a_0^2 z^{*2} \cdot \left(\frac{v_0}{V}\right)^2 \cdot \left(\frac{mv_0^2}{T} - \frac{mv_0^2}{T_{\max}}\right), \tag{4.1}$$

where $T_{\max} = 2\,mV^2$ is the upper limit for energy transfer in such a collision.

Introducing for each ion electron $T = mv^2/2$, and summing by means of formula (3.4), we get from (4.1) as a first estimate of the loss cross section

$$\sigma_l = \pi a_0^2 \cdot z^{*2} \cdot Z^{\frac{1}{3}} \cdot \left(\frac{v_0}{v^*}\right)^3, \tag{4.2}$$

where z^* stands for the atomic number z, or core charge, for light and heavy gases, respectively, and where the binding of the most loosely bound electron in the ground state of the ion is characterized by the velocity v^*, close to V.

Such cursory consideration needs, however, essential corrections of different kind. In fact, the neglect of the effect of the electron binding during the encounter is not justifiable, as the orbital velocities are of the same order as V, and especially since the duration of the encounter is comparable with the orbital frequencies. Due to these circumstances, the estimate (4.2) of the cross section for direct removal of the ion electrons is somewhat too large, but in the estimation of the loss cross section it must be taken into consideration that, due to subsequent readjustment of the electron binding in the ion, electron escape will take place if only the total energy transfer to the ion in the encounter exceeds the binding energy I^* of the most loosely bound electron in the ground state. Still, the corrections due to these various effects, which are not strictly separable, may be expected largely to cancel, and it is in this respect interesting that the estimate of the loss cross section for fission ions in several gases, obtained by Bell (1953) by numerical computation, based on a somewhat different simplifying procedure, agrees approximately with the more comprehensive formula (4.2). We may therefore use this formula as a guide in the analysis of the experiments, and especially in the estimation of the variation of the loss cross section with ion charge.

Besides electron loss, the encounters with the atoms will result in excitation of the ion. An estimate based on the simple formula (4.1) gives in fact a cross section for excitation by direct impact of the same order of magnitude as the loss cross section. Even if part of the excitation energy by subsequent readjustment will be spent in electron escape, we must therefore reckon that the collisions will result in excitation of the ion, amounting on the average to about $I^*/2$. In gases at low pressures, this excitation will be dissipated by radiation between collisions, but at higher pressures we have to take into account initial ion excitation in the encounters, with the result that the total loss cross section is increased. Thus, by a simple estimate based on (4.1), we obtain for an average residual excitation εI^* at the beginning of the encounter a relative increase ε in the loss cross section.

An estimate of the cross section for electron capture by the ion demands a somewhat more detailed consideration of the course of the encounter between the atom and the ion. In fact, the possibility of capture of an electron by the ion will largely depend on the circumstances under which it is released from the atom. Let us consider an atomic electron with orbital velocity v and radius a, as given by (3.3). During the approach of the highly charged ion, the electron will be exposed to a strong field of force, giving rise to an increasing polarization of the binding, which may subsequently lead to its rupture. In order to estimate when electron release takes place, we note that, at a distance R between the two systems given by

$$\frac{Z^* e^2}{R^2} = \frac{m v^2}{a}, \tag{4.3}$$

the force from the ion and the atomic binding force are approximately equal. Still, it has to be taken into account that the possibility for electron release is not only determined by a comparison between the forces, but that the completion of the process will require a time of the order a/v, and that therefore, especially in the case of the more loosely bound atomic electrons, the ion may have travelled a distance comparable with R before the electron is liberated from the atomic field.

After the release from the atom, the electron will be captured if its total energy relative to the ion has a negative value. In his estimate of capture cross sections, on similar lines as followed here, Bell (1953) assumes that an atomic electron is released at a distance R from the ion with velocities corresponding to the momentum distribution in its original binding state. It must, however, be taken into consideration that, under the combined action of the atom and ion fields, the electron velocity distribution will have changed considerably from that in the isolated atom, and that we must expect the velocity of the electron to be largely reduced during the gradual loosening of the atomic binding. At the completion of the release process, we may thus in first approximation assume that the velocity of the electron relative to the ion will not differ essentially from the ion velocity. On such assumptions, the condition for capture is that the process of electron release is effectively completed at a distance from the ion smaller than R', determined by

$$\frac{Z^* e^2}{R'} = \frac{1}{2} m V^2. \tag{4.4}$$

Assuming, in first approximation, that the release takes place at the distance R, we find that, if $R < R'$, capture occurs with a cross section πR^2, while for $R > R'$ there will be no capture. According to (4.3) and (4.4), it is seen that on this assumption only strongly bound atomic electrons can contribute to capture. Actually, in a heavy atom, the contribution will arise mainly from a comparatively narrow region of orbital velocities around $V/2$. Summing over the electrons in the atom, we obtain by means of formula (3.3) the approximate estimate

$$\sigma_c = \pi a_0^2 Z^{*2} \cdot z^{\frac{1}{3}} \cdot \left(\frac{v_0}{V}\right)^3 \tag{4.5}$$

for the total capture cross section for atoms in which a considerable part of the electrons have velocities comparable with V.

Notwithstanding the cursory character of the description of the capture process, the formula (4.5) may be expected not to be far in error, because the uncertainties introduced by the estimates of R and R' will be largely eliminated by the summation over the atomic electrons. This circumstance was also noted by Bell (1953) in his numerical computation of capture cross sections in several gases by fission ions of various charges and velocities. In spite of the different assumptions used by Bell as regards the kinetic energy of the released electrons, his results for heavier gases also agree approximately with formula (4.5). Moreover, it must be noted that the formulas (4.2) and (4.5) imply that, in a close encounter with a heavy atom, several electrons will be lost and captured by the ion, and that due to subsequent readjustment of ion excitation the various processes are not strictly separable in the resulting effects.

As regards capture in the lightest gases, we meet with an essentially different situation. In fact, for ions of high charge and velocity, the calculations leading to the capture cross section (4.5) would give no contribution for electrons bound in the lightest atoms, because for these the distance of release, R, would be larger than R', the limit where capture becomes possible. In order to explain the occurrence of capture, we must take into account that the release is a gradual process and, although R may represent the average release distance, escape will take place only with a probability per unit time comparable with (v/a), and thus over a considerable path. Accordingly, there is a small probability that a loosely bound electron will remain with the atom until the distance from the ion is so small that capture can take place.

The detailed analysis of the process presents, of course, a complicated problem, but by an estimate relying on simple mechanical concepts, and assuming that the probability of electron release from the atom within a distance from the ion smaller than R' is of the order $(R'/V)(v/a)$, we get

$$\sigma_c = \pi a_0^2 Z^{*3} \cdot \left(\frac{v_0}{V}\right)^7 \cdot \frac{n'^2}{\nu'^3} \tag{4.6}$$

as a cursory estimate of the capture cross section for a very loosely bound atomic electron, with a binding characterized by a screened nuclear charge $n'e$ and an effective quantum number ν'.

For the discussion of residual ion excitation, we must take into consideration that the electrons will in general be captured in highly excited states. In fact, for heavy atoms to which formula (4.5) applies, the average excitation of an electron after capture by an ion in the ground state will be about $\frac{2}{3}$ I^* while, in the case considered in (4.6), the excitation will in general be still higher and closely approach I^*. As regards the examination in § 6 of the effect of residual excitation on the balance at higher pressures, we further note that, in contrast to the increase in the loss cross section due to residual excitation, discussed above, we must expect a decrease in capture cross section due to subsequent readjustment of the electron binding. Thus, an average residual excitation at the beginning of the encounter, amounting to εI^*, will give rise to a relative decrease in the estimate (4.5) of magnitude $\frac{3}{2}\,\varepsilon$ and even more in the estimate (4.6).

§ 5. Discussion of experimental evidence on capture and loss by fission ions in gases at low pressures.

In order to ascertain how far the approximate estimates of loss and capture cross sections, given in § 4, may be used as a guidance for discussion of the experimental evidence, it may be recalled that, while the estimate (4.2), with proper definition of z^*, applies to electron loss in both light and heavy gases, we have as regards the capture problem in the two cases to do with essentially different mechanisms, leading to the estimates (4.5) and (4.6), respectively. In the comparison with the experimental evidence, we shall therefore treat the two cases separately.

In the case of the heavy gases, in which the binding of a major part of the atomic electrons is characterized by orbital velocities comparable with or exceeding the ion velocity V, the formulas (4.2) and (4.5) give simple variations of the capture and loss cross section with ion charge, and in opposite directions. In fact, the capture estimate (4.5) is proportional to Z^{*2}, while the loss cross section (4.2) is inversely proportional to v^{*3} and therefore varies approximately as Z^{*-3}. In particular, we note that the two expressions in all heavier gases become equal for a value of the velocity of the most loosely bound ion electrons closely given by $v^* = V$, in agreement with the cursory estimate of the balance charge used in the discussion in § 3.

From (4.2) and (4.5) we get, with the notation of § 2,

$$\Omega = \pi a_0^2 Z^{\frac{1}{2}} \cdot z^{\frac{1}{2}} \cdot \frac{v_0}{V} \tag{5.1}$$

for the equal loss and capture cross section in balance. As regards the estimates of the mean free path between collisions involving electron capture and loss, and determining for the dependence of balance charge on gas pressure (cf. § 6), it must, however, be taken into consideration that just in heavier gases several electrons will in general be exchanged in the encounters, and that we must therefore reckon with a somewhat larger value for the mean free path than would correspond to (5.1).

At low pressures, the experimental results, given in Fig. 1, show that the balance charge in heavier gases is nearly independent of atomic number, as also corresponds to the theoretical

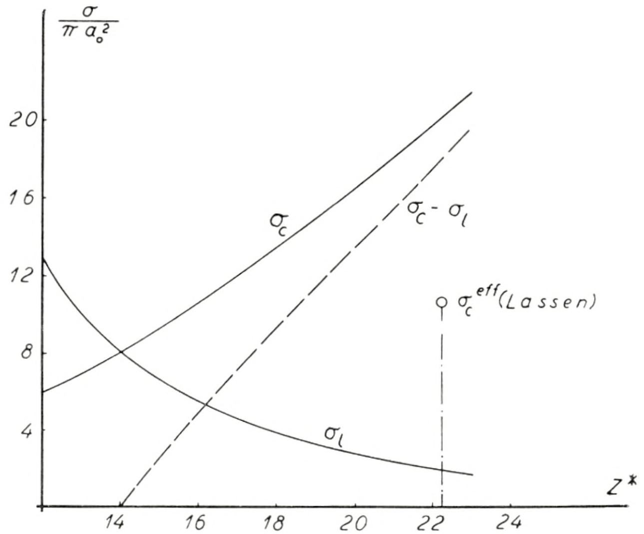

Fig. 3. Capture and loss cross sections for the heavier group of fission ions with initial velocities in argon at low pressures, as functions of ion charge Z*. Comparison with the average effective capture cross section estimated by LASSEN.

expectations. However, it is to be kept in mind that, for such comparison, we are in the first place only dealing with the ratio between the loss and capture cross sections, and that the rapid and opposite variation of the cross sections implies that the balance charge is not very sensitive to this ratio. It is therefore important that an approximate test of the numerical values of the expressions (4.2) and (4.5) can be obtained from LASSEN's studies of the transitional effects observed for ions emerging from solids into gases.

For the heavy group of fission ions with initial velocities in argon at low pressures, the theoretical estimates σ_l and σ_c as functions of Z^* are represented by the two curves in Fig. 3. The intersection point of the curves, corresponding to the balance charge, agrees closely with the experimental value. The dotted curve in the figure represents the difference between the capture and loss cross section for ion charge higher than the balance value. Further is on the figure indicated LASSEN's estimate of the effective capture cross sections for the average ion charge of the heavy fission fragments emitted from a solid surface. As

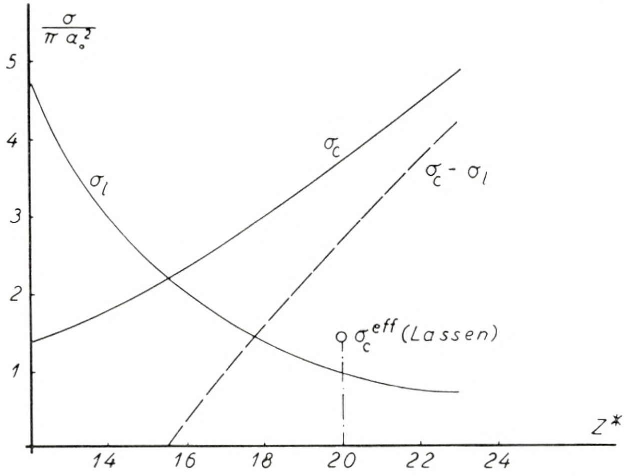

Fig. 4. Capture and loss cross sections for the lighter group of fission ions with initial velocities in argon at low pressures.

mentioned in § 2, this estimate was deduced from the rate of decline in charge for ions emerging into the gas chamber, and it was, for simplicity, assumed that in these effects electron loss could be neglected and the capture cross section considered as constant over the charge interval in question. Considering that, in the transitional effects, we have to do with an averaging over the difference between the capture and loss cross sections within an interval between the charge of the emerging ions and the balance charge in the gas, it is seen that LASSEN's estimate is in quite satisfactory agreement with the cross section curves in Fig. 3.

In a similar way, the curves in Fig. 4 represent theoretical estimates for σ_l, σ_c, and $\sigma_c - \sigma_l$ for the light ion group in argon. Still, in conformity with the considerations in § 3, we have in formula (4.2) introduced, instead of $Z^{\frac{1}{3}}$, a somewhat smaller value of the effective quantum number, so as to obtain coincidence between the intersection point of the curves and the measured balance charge. It will be seen that LASSEN's estimate for the effective capture cross section of light fission ions emerging from a solid surface into the gas is also consistent with the theoretical expectations when considering the run of the curves within the charge region of the transitional effects.

As shown in § 2, the rates of variation with ion charge of the cross sections for loss and capture are determining for the charge fluctuations of the ions along their path. The formulas (4.2) and (4.5) lead to the value $1/(\alpha_l - \alpha_c) = Z^*/5$ in heavier gases at low pressures. While the charge fluctuations in a gas escape direct measurements, it is interesting that this estimate of the average square fluctuation corresponds approximately to the observation of the charge fluctuations of fission ions emerging from solids (LASSEN 1950, 1951 a).

As regards the competition between loss and capture in lighter gases, it is seen that, while the capture cross section for fission fragments in air should be approximately given by the formula (4.5), the charge of the atomic core entering in the loss cross section (4.2) will be somewhat smaller than the value $z^{\frac{1}{3}} (V/v_0)$ holding for heavy substances. Thus, the balance charge may be expected to be slightly lower in air than in argon, as was also found by LASSEN (cf. Fig. 1). The anomalies in average ion charge in the lightest gases like helium and hydrogen are, however, of particular interest. Especially the comparatively high value of the ion charge in hydrogen points to a decrease in the capture cross section even more rapid than the decrease in loss cross section, which for the lightest elements is proportional to z^2.

Although the estimate (4.6) may not give accurate numerical results, the relative variations with atomic number, ionic charge and velocity are expected not to be far in error. Such dependence is brought out by comparing (4.6) with the loss cross section (4.2) for the light and heavy fission groups with initial velocities in H_2 and He. The measurements show here that for both groups of fission ions the average charge in He is about 10 % lower than in H_2. This circumstance is readily explained from (4.2) and (4.6), since σ_l varies as z^2, while σ_c is nearly proportional to z^3 and, accordingly, the charge must remain slightly smaller in He.

For a more quantitative comparison with (4.2) and (4.6), we calculate σ_c, σ_l, and the balance charge for the two groups of fission ions in H_2 and *He*. In order to compute σ_l as a function of Z^*, one must know the value of the effective quantum number, ν^*, for the most loosely bound ion electrons. For the heavy group of fission ions, we may put ν equal to $Z^{\frac{1}{3}}$, while for the light group we can take the somewhat lower value given by the measured balance charge in argon at low pressures, assuming that the velocity of the most loosely bound ion electrons is $v^* = V$. The theoretical estimates of the balance charges are given in Table 1, together with the measured balance charges, and it is seen that the agreement is quite close. In the table is also given the effective capture cross section calculated by Lassen from the transitional effects for ions emerging into gases from solids. The comparison with the difference of the theoretical estimates of σ_c and σ_l for the charge value of the emerging ions shows agreement, at any rate as regards order of magnitude.

Also the measurements of the balance charge of fission ions with lower velocities (Lassen, 1951 a) bring out a difference between heavy and light gases, which seems to be in approximate agreement with the theoretical estimates. Thus, the observation that for argon Z^* is closely proportional to V for the heavier ion group, while for the lighter group Z^* varies more slowly with ion velocity, is in conformity with the assumption that in both cases the velocity v^* of the most loosely bound ion electrons is closely equal to V. In the lightest gases, however, an approximate pro-

Table 1.

Balance charge and effective capture cross section (in units of πa_0^2) of fission ions with initial velocities in H_2 and *He*. Comparison between measurements on the ions emerging from uranium (Lassen, 1951 a, 1954) and theoretical estimates based on (4.2) and (4.6).

	H_2		*He*	
	heavy	light	heavy	light
Z^*_{bal} (exp.)	12.7	15.8	11.6	14.1
Z^*_{bal} (th.)	12.2	15.7	10.9	14
σ_c^{eff} (exp.)	0.9	0.025	3.2	0.3
$\sigma_c - \sigma_l$ (th.)	0.9	0.02	(7.5)	0.2

portionality between Z^* and V was found both for the heavy and light ion group, corresponding to a different connection between v^* and V, as is also borne out by a comparison between (4.2) and (4.6).

§ 6. Dependence of average ion charge on density of material.

Even if collisions with atoms will generally leave the ion in an excited state, we may in gases at low pressures assume that such excitation will be dissipated by radiation between successive collisions, and that the average charge of the ions simply depends on the cross sections for capture and loss by the ion in the ground state. In gases at higher pressures, or in solid materials, we must, however, take into account that the ions to a greater or smaller extent will remain in an excited state, and for an estimation of the average ion charge the influence of the residual excitation on the balance between loss and capture must be considered.

As already mentioned, the excitation of the ion produced directly by the collisions with the atoms will in general be shared among the ion electrons and, if larger than the minimum ionization potential $I^* = mv^{*2}/2$, give rise to subsequent electron ejection. While in solids such adjustment may not be completed between successive encounters with the atoms, we may in gases, even at comparatively high pressures, assume that the ions at the beginning of each collision have a more or less distributed excitation, never exceeding I^*. In heavier gases, the average excitation after a collision will be about $I^*/2$, but in the lightest gases like hydrogen, and especially for swiftly moving ions, capture will result in a very loose electron binding, and the average excitation may be somewhat higher.

In order to estimate how large a part of the excitation of the ion will remain between collisions in the gas, we shall assume that its dissipation by radiation is characterized by a mean lifetime, τ, corresponding to an ion path τV. Reckoning with a mean free path λ of the ion between collisions, we get therefore, in a well-known manner, that the fraction of the ions which on the average have retained the excitation between collisions will be given by $\tau V/(\tau V + \lambda)$. Assuming that the collisions only involve capture or loss of a single electron, we may, with the notation of § 2, write $\lambda = 1/(2\,\Omega\,\varrho)$, and we get thus for the ratio $\bar{\varepsilon}$ between the average residual excitation and I^*

$$\bar{\varepsilon} = \frac{\tau V \Omega \varrho}{2\,\tau V \Omega \varrho + 1}. \tag{6.1}$$

In the case of heavy gases, where the relatively large probability for loss and capture of several electrons in close collisions may necessitate the use in (6.1) of a value somewhat smaller than Ω given by (5.1). For the lightest gases, however, due to the considerable probability of collisions giving rise to excitation without electron loss or capture, the value Ω in (6.1) should be replaced by a somewhat larger cross section.

In order to estimate the influence of the residual excitation on the balance between loss and capture, we shall, in direct generalization of (2.3), write

$$\left.\begin{aligned} \sigma_c &= \Omega\cdot(1 - \beta_c\,\varepsilon + \alpha_c\cdot(\tau - \omega)), \\ \sigma_l &= \Omega\cdot(1 + \beta_l\,\varepsilon + \alpha_l\cdot(\tau - \omega)), \end{aligned}\right\} \tag{6.2}$$

where Ω and ω as well as the constants α_c and α_l refer to the ground state, while $\beta_c\varepsilon$ and $\beta_l\varepsilon$ are the relative variations in the cross sections for excitation εI^*.

In the absence of excitation, the balance charge of the ion is $Z - \omega$, and the equations (6.1) and (6.2) thus imply a shift in balance charge, of magnitude

$$\Delta Z^* = \frac{\beta_l + \beta_c}{\alpha_l - \alpha_c}\cdot\bar{\varepsilon} = \frac{\beta_l + \beta_c}{\alpha_l - \alpha_c}\cdot\frac{\tau V \Omega \varrho}{2\tau V \Omega \varrho + 1}. \tag{6.3}$$

For low densities the shift ΔZ^* is proportional to ϱ, while for high densities it reaches a maximum value, $(\beta_l + \beta_c)/2\,(\alpha_l - \alpha_c)$. Introducing for $\alpha_l - \alpha_c$ values corresponding to (4.2), (4.5) and (4.6), and for $\beta_l + \beta_c$ the estimates in § 4, we get for the maximum value of $\Delta\,Z^*$ about $Z^*/5$ in heavy, and slightly more in light materials. This result is in good agreement with the experiments by Lassen (1951 b), where the average charge with increasing pressure seems to reach a constant value about 3 units higher than at the lowest pressures.

While the constant charge value at higher pressures is independent of the emission of radiation, the initial increase in ion charge at low pressures is a direct consequence of the competition between collisions with gas atoms and dissipation of excitation by radiation. The above simple description of the radiative

Table 2.

Measured values of p_1, for the two groups of fission ions in various gases (Lassen 1951 a, b) and the corresponding lifetimes. The uncertainty in p_1 may be a factor $\sim$ 2.

	Heavy group			Light group		
	H_2	He	A	H_2	He	A
p_1 mm	11	12	4	30	15	5
$\tau \cdot 10^{11}$ sec.	2.7	1.2	0.2	4	3.5	0.4

decay by an effective lifetime τ is in agreement with the observed approximative linear increase in ion charge with gas pressure. In Table 2, the values of p_1 represent the pressure for which the average ion charge has increased by one unit, estimated from the slope of Lassen's curves in various gases. The table also gives the corresponding values for the radiative lifetime τ, deduced from (6.3).

As a simple estimate of the radiative lifetime τ of an excited electron state, we may write

$$\tau \simeq \tau_o \cdot \frac{\nu^5}{Z^{*4}}, \qquad \tau_o = 0.9 \cdot 10^{-10} \text{ sec}, \tag{6.4}$$

where Z^* is the charge of the ion, and ν an effective quantum number somewhat higher than, but comparable with the quantum numbers of the most loosely bound electrons in the ground state of the ion. The radiative lifetimes to be expected from (6.4) are of the same order of magnitude as those derived from (6.3) and given in Table 2. Moreover, the larger values for τ in hydrogen and helium, compared with argon, may perhaps be explained by the smaller ion charge and the higher excitation states of ion electrons to be expected in the lighter gases. Still, such closer comparison contains much uncertainty, especially in the estimate of Ω, which quantity, as already mentioned, may have to be considerably increased in the lighter gases, in a way which may at any rate partially account for the larger estimate of τ in hydrogen and helium compared with argon.

While in gases at comparatively low pressures, the time between collisions can, as we have seen, be of the order of the radiative lifetimes of excited electron states on the ion, the passage of ions through solids implies an extremely rapid succession of collisions and, as in gases at high pressures, the dissipation of ion excitation by radiation can be neglected. However, even in solids, the collision frequency, V/λ, will remain smaller than the revolution frequency, $\omega = v/a$, for the orbital motion of the ion electrons. In fact, since the orbital velocities of the ion electrons are comparable with V, the two mentioned frequencies will, for heavier atoms, approximately have the same ratio as the ion radius to the spacing of atoms in the solid, and have an even smaller ratio for lighter atoms. As regards the initial stages, the mechanism of the individual capture and loss processes should thus not differ essentially for gases and solids, and the marked difference in balance charge in the two cases therefore points directly to the importance of subsequent readjustment of the distribution of ion excitation.

Just as regards such readjustment, the rapid succession of the collision processes in solids will restrict the possibility of sharing excitation between electrons on the ion. In fact, the time, τ_{dis}, necessary for distribution over several electrons of an excitation initially confined to one, will be long compared to the revolution frequency, and we may reckon that the time between collisions in solids is shorter than τ_{dis}. The competition between collisions and distribution of energy between ion electrons may thus allow the excitation of the ion to exceed the minimum energy for ionization, I^*. The description of the ion state in balance becomes particularly simple if it may be assumed that there is not sufficient time for redistribution of ion excitations. In this case, an electron captured in an excited state will be lost from the same state, so that for each single electron state there is a direct competition between capture and loss.

Due to the very rapid increase of the cross section for electron loss with decreasing binding energy, the balance between capture and loss will therefore be essentially shifted by suppression of readjustment of ion excitation, in spite of the circumstance that such readjustment in itself may lead to electron release from the ion. In fact, in collisions with atoms, ion electrons can be removed from states with binding velocity nearly as large as $2V$, and we may reckon that even in solids more strongly bound electron states on the ion are occupied, while in higher states only a few electrons will remain, due to the competition between capture and loss. In a rough estimate on such lines we find that the ion charge will be about $(3/2) \cdot (V/v_0)\, \nu$, where ν is the quantum number of ion electrons with orbital velocity between V and $2V$.

In Figure 1 was shown the measured balance charge of fission ions in various solids and in gases at low pressures. It is seen that, for the heavier group of fission ions, the charge in solids approximately corresponds to the above estimate, since the effective quantum number is $\nu \approx Z^{\frac{1}{3}}$. For the light group, however, we found that already for the charge values in gases, ν was somewhat lower than $Z^{\frac{1}{3}}$, and for the high stripping in solids ν must have decreased even further. This circumstance accounts for the result that the charge of the light group in solids becomes slightly lower than that of the heavy group, opposite to what is the case in gases. Figure 1 also shows a small but marked decrease of ion charge with increasing atomic number of the solid substance penetrated. This effect points to a gradual minor change in the balance between capture and loss, probably connected with the greater average binding of electrons captured in heavier substances and thus reducing the probability for subsequent loss.

Although it thus appears that many of the characteristic features of the difference between average ion charge in dense and dilute materials may be explained by simple mechanical arguments, it must be stressed that we are dealing with a highly complicated problem, the detailed treatment of which needs further experimental and theoretical investigation. In a closer comparison with the empirical results, it must thus be taken into consideration that the high excitation of the ions in dense materials may result in a subsequent emission of electrons from the ions immediately after their escape into vacuum and thus, to a certain extent, increase the measured charge values. It may also be remarked that, in a comparison between the stopping power for ions in dense and dilute materials, attention must be paid primarily to the considerable difference in the two cases of the charge of ions with given velocity.

Such problems must also be taken into account in comparisons between the phenomena accompanying the penetration of swift heavy ions through gaseous media and the remarkable observation of tracks of highly charged cosmic ions in photographic emulsions (cf., e. g., Bradt and Peters, 1950). The rich material with which we in such observations are concerned extends, however, over a far wider energy region than the experiments with fission ions. Estimates of cross sections for electron capture and loss, determining for the balance charge of such rapid cosmic ions and its variation on their path through the photographic emulsion, therefore obviously demand considerations beyond the scope of the discussion in this paper.

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