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# ENERGY LOSS IN MATTER BY FAST PARTICLES OF LOW CHARGE

BY

J. LINDHARD AND M. SCHARFF



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#### § 1. Introduction.

In the discussion of the energy loss by swift charged particles **A** passing through matter one may conveniently distinguish between two extreme cases. When a penetrating particle has a sufficiently high charge, or low velocity, it will carry electrons which to some extent will screen the field of the particle. The problem of its energy loss is then quite involved, even though it may be treated essentially on classical mechanics. But as soon as the velocity of the particle is high compared with the orbital velocity of an electron carried by it in the ground state, the energy loss can be computed rather accurately using a quantum mechanical perturbation treatment. In the present paper we shall be concerned with the latter simple case, and in particular with the slowing down of protons and α-particles. We shall try to build up a simple and consistent picture of the atomic processes displayed in stopping problems for atoms containing many electrons. But it may be useful to make first a few remarks regarding the different treatments and points of view on the subject.

We consider then the energy loss suffered by a heavy particle of velocity v and charge ze, passing through a substance of atomic number Z and with a density N atoms per unit volume. As long as v remains large compared with the velocity of the more strongly bound electrons in the substance, the average specific energy loss of the particle is with good approximation given by the general formula of Bethe (Bethe (1930), Bethe and Livingston (1937))

$$\frac{dE}{dR} = \frac{4\pi z^2 e^4}{mv^2} \cdot NZ \log \frac{2mv^2}{I},\tag{1}$$

where I is a constant characteristic of the substance, often denoted as the average excitation potential. This constant was by Ветне found to be determined by

$$L = \log \frac{2 m v^2}{I} = \frac{1}{Z} \sum_{i,k}^{7} f_{ik} \log \frac{2 m v^2}{\hbar \omega_{ik}},$$
 (1')

where  $f_{ik}$  is the atomic oscillator strength corresponding to the transition (i, k) with frequency  $\omega_{ik}$ ; a similar formula applies when chemical bindings come into play. The equation (1) holds for non-relativistic velocities. For very fast particles the relativistic term  $(-\log (1-v^2/c^2)-v^2/c^2)$  is to be added to the logarithm in (1). It is well known that half of this term arises from close collisions with the electrons in the substance, while the other half is due to distant collisions. It should be mentioned that the relativistic formula is not always quoted correctly in the literature, and also that some authors introduce a potential I defined in a manner different from that used here. A correction to the above relativistic term, depending on the density of the material, was introduced by Fermi (1940). We shall not be concerned with relativistic cases where this effect is important.

In connection with (1) it is worth recalling that for an analysis of stopping problems it can be of advantage to use a simple idealized picture of the energy transfer. The collision may then be described by a classical impact parameter, p, and a collision time,  $\tau = p/v$ . For a particle of high velocity the collisions can be divided into the close and the distant ones. In the violent close collisions the binding and mutual interaction of the electrons will not be important. In the more distant collisions the average energy transfer is equivalent to that in free classical impacts. It is now decisive for the magnitude of the total energy loss by the particle that the energy transfer becomes negligible for collision frequencies,  $1/\tau = v/p$ , less than the adiabatic frequency  $\omega$  characterizing the dynamical properties of the atomic system. The value of  $\omega$  is connected with I by the relation  $I = \hbar \omega$ . We shall repeatedly make use of the above simple concepts.

The energy loss suffered by electrons passing through matter cannot, for several reasons, be contained in formulae of type of (1). In the following we shall treat only heavier particles, but the estimates of the average excitation potentials, which involve only distant collisions for fast penetrating particles, can of course be used as well in the description of the stopping of electrons.

In Bethe's deduction I is determined by the transition fre-

quencies and corresponding oscillator strengths in the atomic system. A direct calculation of I on these lines is simple in the case of hydrogen, but becomes complicated for substances of high atomic number. A considerable simplification of the problem for heavier substances was achieved by Bloch (1933), when he applied the Thomas-Fermi model of the atom. Making some simplifying approximations in the Thomas-Fermi model when extending it to dynamical problems, Bloch found that the average excitation potential I = I(Z) in Bethe's formula (1) could be written as

$$I = I_0 \cdot Z, \tag{2}$$

 $I_0$  being a constant independent of Z. Bloch did not calculate the value of  $I_0$ ; in fact, it seemed difficult to compute this quantity from his model with sufficient accuracy, an empirical determination being preferable. A calculation of  $I_0$  was carried through later by Jensen (1937), but with a picture of the atom too simplified to allow quantitative comparisons.

The numerous data on the slowing-down of protons and  $\alpha$ -particles in substances of medium and high atomic number have shown that Bloch's relation (2) is rather well satisfied for sufficiently high velocities v. The values of  $I_0 = I/Z$  found empirically are about 10 eV, with only slight variations between the different elements.

When the velocity of the particle becomes comparable with those of the more strongly bound electrons in the substance—as is usually the case for natural α-rays—the energy loss is no longer well represented by the constant potential in equations (2) or (1). The potential will then vary with velocity because the logarithmic expressions in (1') apply only when the arguments are large. Bethe has here introduced corrections of the contributions from the electrons in the K-shell (Bethe and Livingston (1937)). In the following we shall be particularly interested in this velocity region, and also in the stopping for still lower velocities, where N. Böhr (1948) has accounted in a simple manner for the empirical  $v^3$ -relation for the range of α-rays.

It is the aim of this paper to bring out general relationships, embracing results like that of Bloch (2). We shall rely on simplified descriptions of atoms, where the main features of atomic

dynamics are most easily recognized. It may be that more than due emphasis is given to the comparison with a free electron gas and the description based on polarization. But we found this way of approach preferable; although less common it is perhaps the simplest one.

In order to get a first insight into the phenomena we shall, in § 2, try to arrive at a simple qualitative picture of the stopping in heavier substances, and in § 3 discuss in how far this picture can be said to agree with the data. These questions were treated briefly in a recent note (Lindhard and Scharff (1952)). A more detailed discussion of different kinds of approach employed in atomic dynamics is attempted in § 4. We shall endeavour to show the significance of the revolution frequencies of the electrons in these problems, and their connection with the adiabatic frequency. Moreover, the electronic interaction appearing in the polarization is found to be of decisive influence for the dynamics of heavier atoms. A formula with rather general applicability is derived for the energy loss in matter, and more quantitative results are then obtained in § 5. Among the questions there to be treated are the magnitude of the polarization effects and the reduction in energy loss for low velocities of the particle. Finally, the straggling phenomena are discussed briefly in § 6 on similar lines.

## § 2. Stopping by Heavier Substances in a Qualitative Description.

As well known the statistical description of Thomas and Fermi gives a surprisingly good account of the atomic structure and binding, in particular for electrons in the intermediate region of an atom. In the problem of the energy loss of a particle penetrating through atomic systems this method will seem especially well suited, because the atomic electrons all give comparable contributions to the stopping, so that the total effect is due mainly to the majority of the electrons with medium binding. Neither the individual characteristics of the atoms or molecules, determined by the outer electrons, nor the precise magnitude of the binding of the innermost electrons will be of importance in first approximation.

In the Thomas-Fermi model the electronic density distri-

butions for different atoms are similar, and the common unit of length is proportional to  $Z^{-1/3}$ . The charge density  $e\varrho$  is therefore proportional to  $Z^2$ , and the total binding energy of the atom behaves as  $Z^{7/3}$ . Since, thus, the binding per electron is proportional to  $Z^{4/3}$  the electrons may be said to have velocities proportional to  $Z^{2/3}$ .

In the present connection we are interested in the dynamics rather than the statics of the atom. When the dynamical treatment is based on the Thomas-Fermi model the motions can be described on classical mechanics, only with due regard to the exclusion principle in the initial state of the system. Suppose now that a small disturbance is set up in the atom. The development in time of this disturbance can be governed by only two kinds of frequencies. Of these, one is the frequency  $\omega_0$  $(4 \pi e^2 o/m)^{1/2}$ , determined by the densities of mass and charge and corresponding to the classical resonance frequency of an extended gas of charged particles. For heavy atoms the spectrum of classical resonance frequencies  $\omega_0$ , behaving as  $\rho^{1/2}$ , is thus contained in a single distribution with a scale proportional to Z. The second kind of frequencies can be pictured in the following manner. The disturbance will be propagated and at the same time damped with certain velocities, and since all velocities in the static model behave as  $Z^{2/3}$  the velocities of propagation and damping must show this dependence on the atomic number. Now. the linear dimensions of the system are proportional to  $Z^{-1/3}$ . and accordingly the frequencies of damping and revolution are proportional to Z. Thus, we have found that all frequencies entering in the dynamical description show the same dependence on the atomic number. This result was first obtained by Bloch (1933) on the basis of his simplified hydrodynamical model of atomic dynamics.

The general behaviour of the frequencies in the atom may also be accounted for by noting that the unit of time in the Thomas-Fermi model is proportional to  $Z^{-1}$ . It follows that in a perturbation treatment, i.e. in the approximation of linear field equations, where one can speak of a set of proper frequencies of the system, these frequencies must behave as Z.

As to the approximations involved in this picture of atomic dynamics, the use of a perturbation treatment was just a charac-

teristic of the stopping problem for fast particles of low charge. Moreover, the description by a classical approximation is appropriate here, partly because we are concerned with a calculation of the screening in distant collisions, where the classical treatment gives the same average result as the quantum mechanical calculations, and partly since we have described the atom by the semi-classical Thomas-Fermi model.

From the above results we can obtain a qualitative picture of the stopping of a heavy particle, with velocity v and charge ze. One may for instance argue as follows. The specific energy loss will always be of the same form as equation (1), i.e. equal to  $(4\pi z^2 e^4 ZN/mv^2)$  times a dimensionless function, L, independent of the charge ze of the particle. This function is in Bethe's formula an average over the atomic system of quantities of the characteristic logarithmic type. The logarithms depend on the maximum energy transfer  $2mv^2$ —or the corresponding frequency  $\omega_{\text{max}} = 2 m v^2 / \hbar$ — and on the transition frequencies in the atomic system. In the semi-classical description of Thomas and Fermi we must thus expect that the only frequencies which can enter in the function L are  $\omega_{\text{max}}$  and the atomic frequencies proportional to Z, even when L is no longer of the logarithmic type. The dimensionless function L will therefore depend on Zand v only through the ratio of the frequencies, proportional to  $Z/v^2$ , and we can write

$$\frac{1}{N} \cdot \frac{dE}{dR} \cdot \frac{mv^2}{4\pi z^2 e^4 Z} = L = L\left(\frac{v^2}{v_0^2 Z}\right),\tag{3}$$

where the so far unspecified function L(x) is determined by the distribution of the frequencies in the atom. In order to obtain a suitable dimensionless parameter in the function L in (3) we have introduced  $v_0 = e^2/\hbar$  as a measure for the velocities. The equation (3), as it stands, is applicable only for non-relativistic velocities, and normally the familiar term (log  $(1-v^2/c^2)+v^2/c^2$ ) must be added on the left hand side of (3) if v is comparable with c.

It is clear from the above deduction that, if the atomic frequencies entering in the description were, for instance, the binding frequencies of the electrons, proportional to  $Z^{4/3}$ , the function L

would instead depend on the ratio  $Z^{4/3}/v^2$ . But, as we have seen, the dynamic frequencies in the Thomas-Fermi atom are not of this kind. A quite different question is the limitations of the Thomas-Fermi description, mentioned in the beginning of this paragraph. In the first place, due to the individual variations in the binding of the outermost electrons in the atom, one will expect small fluctuations from one element to another, but on the average the formula should remain valid. We shall return to the problem of these fluctuations in § 5. In the second place, the binding of the innermost electrons is not well accounted for by the Thomas-Fermi model, and the corresponding frequencies do not behave as Z. While the most loosely bound electrons primarily give rise to fluctuations, the presence of the strongly bound electrons imply instead systematic deviations from the dependence of L on the single variable  $v^2/v_0^2Z$ . Still, since the individual contributions and the number of these electrons both are small, it is to be expected that they will not have an appreciable influence on the variation of the total stopping with Z and v.

A few simple results may be derived immediately from equation (3). If the velocity v of the particle is large compared with the electronic velocities in the atom, the dependence of the function L on v must be approximately as  $\log(v^2)$ , as in Bethe's formula (1). Equation (3) then leads to Bloch's formula (2), again with an undetermined value of the constant  $I_0$ . In § 5 will be given an approximate estimate of this constant. It may here be noted that the mentioned replacement of  $Z/v^2$  by  $Z^{4/3}/v^2$  in L would in this case give a formula deduced by Sommerfeld (1932), where Z in Bloch's formula (2) is replaced by  $Z^{4/3}$ , at variance with the measured stopping.

For low velocities of the particle, or values of v comparable with the velocities of the majority of the electrons in the atom, the more strongly bound electrons no longer contribute appreciably to the stopping, and the function L will not behave as in the Bloch formula. In the lower part of this region the specific energy loss is approximately proportional to 1/v, corresponding to Geiger's formula for the range of  $\alpha$ -rays. With this dependence on v it follows from (3) that the energy loss is proportional to  $Z^{1/2}$ , in fair agreement with the classical rule according to which the stopping behaves as  $A^{1/2}$ . We shall presently give a more quanti-

tative discussion of the region of low velocities on the basis of recent measurements.

The above-mentioned reduction in the contribution of the strongly bound electrons has been discussed by Bethe from a somewhat different point of view. For velocities comparable with the electron velocities in the K-shell Bethe estimates the decrease in stopping due to these electrons (Bethe and Livingston (1937), Brown (1950), Walske (1952)). This correction sets in at quite high velocities of the particle, and changes initially only rather slowly with velocity. It will therefore seem that a separate correction for the K-shell is a somewhat doubtful procedure, and it is of course not in line with the statistical treatment of the atom. Although further corrections for the L-shell and even higher states can be made, such an attack becomes highly complicated.

For extremely low velocities, as in the case of canal rays, the present description no longer applies. This is partly because there is a high probability that the particle will carry an electron when its velocity is of the order of that of an electron bound to it in the ground state, and partly because the stopping is then mainly due to the outermost electrons which do not follow the statistical model. The deviations set in for values of x somewhat lower than 1, depending on the substance and the charge of the penetrating particle. For values of x of the order  $z^2/Z$  the deviations are expected to be considerable.

Instead of the above formulation where L in (3) is a function only of  $v^2/Z$ , one might say that the specific energy loss itself is a function of  $v^2/Z$ , because dE/dR differs from L only by a factor proportional to  $Z/v^2$ . This formulation can be useful, but it is no longer valid when relativistic corrections set in. Moreover, if one wants to study the empirical justification of a picture of the kind suggested here, it is a better criterion to plot L, which function will show more clearly the presence of small deviations from the picture.

As to the range of the particle we may similarly write, for not too high energies,

$$R = \frac{M}{m} \cdot \frac{Z}{z^2} \cdot \frac{1}{8 \pi a_0^2 N} \cdot f\left(\frac{v^2}{v_0^2 Z}\right),\tag{4}$$

where R is the range, M the mass of the particle, and  $a_0$  the radius of the hydrogen atom. The function f(x) is connected with L(x) by

$$f(x) = \int_{0}^{ax} \frac{x' \, dx'}{L(x')}.$$
 (5)

For higher velocities the relativistic corrections of magnitude about  $v^2/c^2$  destroy the validity of (4); these corrections are primarily due to the kinetic energy being no longer  $\frac{1}{2}Mv^2$ . One might then attempt a more precise formulation in analogy to (3), but unfortunately this is not feasible on the same simple lines.

The formula (4) applies strictly for range differences only. The above-mentioned deviations from the description of stopping for extremely low velocities imply the presence of very small differences in range, depending on the substance and the particle. These differences we shall call differences in excess range, and their values will be found in § 3.

#### § 3. Comparison with Experiments.

Let us compare the formula (3) with experimental data available at present. In Fig. 1 we have plotted L(x) as a function of the variable  $x = (v/v_0)^2 \cdot Z^{-1}$ , using a number of absolute measurements of the stopping of protons of energy between 1—200 keV and 340 MeV, for metals ranging from uranium to the extreme case of beryllium. The values of L(x) are obtained by introducing the measured stopping on the left hand side of (3), and afterwards adding the relativistic correction if the velocity is high. As mentioned no correction should be made for the K-shell. The points on the figure are based on measurements of specific energy loss of protons, performed by the following authors: Bakker and Segrè (1951), Mather and Segrè (1951), 340 MeV. Sachs and Richardson (1951), 18 MeV. Warshaw (1949), 100–300 keV. Madsen (1953), 0.2–2 MeV. See, further, the note on page 14.

The points in Fig. 1 appear to give a rather well-defined curve. The fluctuations around the average are small, as was to be expected. In the present connection it is more significant that the different groups of elements, arranged according to atomic number, do not show a tendency to separate out into consecutive curves. The points for Be should of course not properly be included in this comparison based on the statistical model.

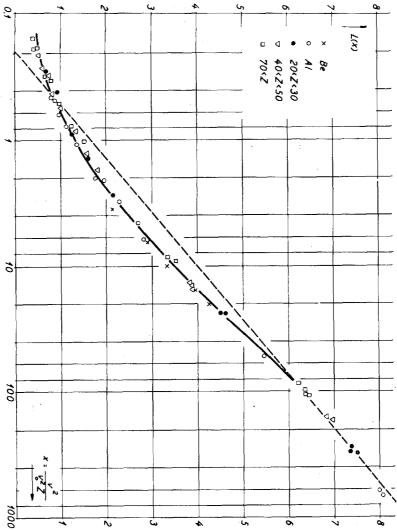


Fig. 1. Comparison between experiments on stopping power according to the statistical treatment. The points represent experimental values, for metals, of the quantity L given by equ. (3). The abscissa is the variable x, in a logarithmic scale. The dotted straight line gives the inclination in Bloch's asymptotic formula, and it corresponds to  $I_0=10\,\mathrm{eV}$ .

In order to cover a wide range of x-values we have used a logarithmic scale for the abscissa in Fig. 1. This has the advantage that points with the same value of  $I_0 = I/Z$  lie on a straight line. The common inclination of such lines is given

by the dotted line on the figure, which corresponds to  $I_0=10~\rm eV$ . For high values of x, i.e. high velocities, it is seen that Bloch's formula with constant  $I_0$  has approximate validity. The value of  $I_0$  thus obtained is about 10 eV, determined essentially by the measurements of Bakker and Segrè, and of Mather and Segrè. For decreasing values of x the curve dips gradually towards

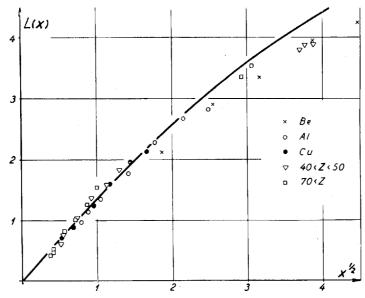


Fig. 2. Comparison between experiments according to the statistical treatment, for low values of  $x = v^2/Zv_0^2$ . As abscissa is used  $x^{1/2}$ . The curve represents formula (11), to be discussed in § 5.

higher values of I = I(v). When x is about 5 a maximum in I is reached, and this potential is here almost twice as large as for high velocities. Eventually, for x decreasing below 5, the value of I decreases again and passes through the original high velocity value when x is of the order 0.5. It should be mentioned here that for x-values between 1 and 5 the curve shown on the figure lies somewhat below the semi-empirical curve for air given by Bethe and Livingston (1937).

If one measures the specific energy loss in matter for a particle of a certain velocity it is possible to find the limiting ionization potential at high velocities by multiplying the measured values of *I* by the proper factor corresponding to the variation of the

potential along the curve in Fig. 1. It is difficult to estimate the accuracy of such a semi-empirical procedure. A correction of this kind has been suggested by Sachs and Richardson (1951), whose measurements cover a considerable part of the curve in Fig. 1. These authors, however, did not emphasize that for lower velocities the correction reaches a maximum, whereupon it decreases again.

The behaviour of the curve for low values of x is more apparent from Fig. 2, covering the interval  $0.2 \le x \le 20$ . As abscissa is used  $x^{1/2}$ , because a  $v^3$ -law for the range in this representation gives a straight line through the origin. In the first approximation the points on the figure may be said to correspond to this law, but the resulting curve is curved slightly downwards. This is particularly so for higher x-values. It should be noted that the figure includes quite high particle energies, e.g. 18 MeV protons in Ag, and that the  $v^3$ -law has been suggested only for considerably lower velocities.

The approximate result that the present parameter x even for low velocities collects the experimental points on a single curve was expected from the qualitative considerations in § 2, but its significance will be seen more directly in the discussion in § 5. It is here interesting that, if one uses instead of x the previously mentioned parameter  $v^2/Z^{4/3}$ , the measurements will separate out into a succession of curves for the different separate elements.

When this paper had been sent to press it came to our notice that KAHN in Chicago has investigated the specific energy loss by protons in Be, Al, mica, Cu, and Au, in the energy interval 0.5-1.3 MeV. His measurements for Be, mica, and Au agree well with Madsen (1953), whereas for Al and Cu Kahn finds values about 10 to 20 % higher than those of Madsen, the deviations being largest for Cu. The reason for the discrepancies is, as yet, not known. Further, measurements of the stopping in Cu have been made by Cooper in Ohio, whose results are about 5% lower than those of KAHN. The x-values involved in the new measurements are 0.6-2 for Cu and 1-4 for Al. Since, in the region around x = 1 on Figs. 1 and 2, the points for Au and Ag are somewhat higher than those for Al and Cu, a possible increase in the latter values should not impair the description by a single function depending only on x. However, for  $x \le 6$  the deviation from the straight Bloch line on Fig. 1 would be less, and the curve would lie closer to that for air. The maximum deviation from the Bloch line would not be changed

essentially, but instead occur at a somewhat higher value of x ( $x \sim 10$ ). The above-mentioned discrepancies may serve to emphasize the uncertainties prevailing in present determinations of energy loss, excepting, perhaps, energy loss in air and in photographic plates.

The measurements shown in Figs. 1 and 2 refer only to the specific energy loss in metals. As regards other substances-and in particular gases-the evidence is mostly relative measurements of ranges, or differences in range. The range observations even have the advantage of being more accurate than direct measurements of the specific energy loss. These two cannot immediately be compared, but instead a separate discussion of the ranges may be made. We shall therefore attempt to plot ranges as a function of the single variable x, in the manner prescribed by equation (4). As mentioned in § 2, this method of comparing ranges can only be applied in the non-relativistic region. There is another difference from the treatment of specific energy loss, because in the very last part of the range the stopping will no longer show the common behaviour assumed above. Even though the resulting range deviations are small they must be taken into account in an accurate representation of the data.

The procedure used in obtaining a range curve is now the following. The measured ranges in mg/cm2 are according to equation (4) multiplied by the factor  $(1/AZ) \cdot (z^2M_p/M)$ , where  $M_n$  is the mass of the proton, and M that of the particle. This will, apart from a constant factor, correspond to finding the quantity f on the right side of equation (4). As a first approximation the results are plotted as a function of the variable x. One finds roughly a common curve, but the sets of points for the separate elements will not precisely follow the trend of the common curve. There will be deviations which are significant only for the shortest range values. We then try to subtract a small individual amount-call it the excess range-from the ranges in order to obtain a curve on which all the measurements are collected. This amount will depend both on the substance and on the charge of the particle. The excess ranges are then so far determined apart from a common additive constant, the fixation of which is given below. The resulting points are shown in Fig. 3 in a double logarithmic scale, together with the values of the excess ranges. We have employed the extensive measurements by

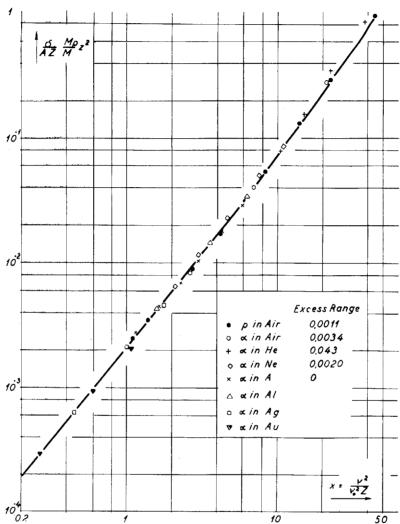


Fig. 3. Ranges plotted according to the statistical formula (4). The ordinate gives, in a logarithmic scale, ranges in  $mg/cm^2$  multiplied by  $(1/AZ) \cdot (z^2M_p/M)$ . The ranges used are experimental ranges minus the excess ranges (see text). The full-line curve was obtained by integration of the averaged curve in Fig. 1.

Mano (1934) of ranges of natural  $\alpha$ -rays in He, air, Ne and A. Further are used the accurate semi-empirical range curves for  $\alpha$ -particles and protons in air by Bethe (1950). Included on the

<sup>&</sup>lt;sup>1</sup> Recent measurements by Reynolds et al. (cf. Bull. Am. Phys. Soc. 27, No. 6 (1952)) with protons of low energies appear to deviate somewhat from the curve by Ветне. For higher energies, 2—8 MeV protons, Виксиам (1953) has obtained results in good agreement with Ветне.

figure are moreover the measurements by Rosenblum (1928) of range differences for  $\alpha$ -rays in the metals Al, Ag and Au. From these range differences one does not, of course, obtain the values of the excess ranges. On the figure is included He where the statistical considerations in § 2 do not apply and the average excitation potential is much higher than the value given by the Bloch formula. As seen from the figure, this gas cannot either be made to follow the common curve for higher values of the variable x. For air the charge value Z=7.22 can be used, since the expressions in question vary only with a low power of Z.

The ranges in Fig. 3 can be compared with the specific energy loss from Figs. 1 and 2. For this purpose we integrate according to equations (4) and (5) the averaged experimental specific energy loss given by the full-line curve in Fig. 1. However, the specific energy loss is not accurately known for x-values below  $\sim 0.2$ . We have here made the plausible choice of continuing the curve to the origin by a straight line on Fig. 2. The integration can then be performed, and the resulting curve is shown by the full-line curve in Fig. 3. There is good agreement with the range measurements.

Using the integrated curve we have fixed the additive constant in the excess ranges. This gives the not unreasonable result that the excess range for  $\alpha$ -rays is highest in He and vanishes in A. Moreover, the excess ranges are of the order of magnitude to be expected from the previously mentioned effects. For  $\alpha$ -rays they are higher than for protons, corresponding to the screening of the charge appearing for higher values of x.

The description in § 2 has thus been found to apply rather well for the average specific energy loss. Similarly, the ranges seem to show mutual agreement on this picture, and further to be consistent with the specific energy loss in metals. But one should not, of course, be deluded as to the accuracy obtained from the procedure used here in discussing the ranges and comparing them with the specific energy losses.

#### § 4. Collective and Independent Particle Descriptions.

In the following we shall consider the mechanisms involved in the present problems of atomic dynamics, and discuss the connection between the independent particle and collective de-

scriptions as they appear in statistical models of atoms. For a further justification of the present line of argument, the reader is referred to LINDHARD (1953).

A detailed description of an atom is afforded by the Hartree model, where the wave function of the total system is the product of one-particle wave functions. When the system is disturbed by an external field these wave functions will of course not develop independently in time. Still, since the one-particle wave functions are governed by the same one-particle Hamiltonian, they will automatically remain orthogonal. This has the advantage that the exclusion principle need not be taken into account in the dynamical treatment.

Now, in actual treatments of atomic dynamics, one usually has recourse to the so-called independent particle model. By the independent particle model we shall here understand a description of the system as particles moving independently of each other in a fixed atomic field. This differs from the Hartree description in that the dynamics is governed only by the action of the external forces, the internal forces being regarded as unchanged. It is apparent that in this simplified description too the exclusion principle may be disregarded.

Let us apply the general expression for the average energy loss given by Bethe, where the logarithmic term in equation (1) has the form

$$L = \frac{1}{Z} \sum_{i,k} f_{ik} \log \frac{2mv^2}{\hbar \omega_{ik}}.$$
 (1')

For the present we assume that the velocity of the particle is high compared with the electronic velocities in the atom, and ask for the corresponding limiting value of the average excitation potential I. If we were concerned with only one electron bound in a static potential it would be the spacing between neighbouring quantum states which determined I, equal to  $\hbar$  times the effective adiabatic frequency limiting the energy transfer. For high quantum numbers these quantities are then simply given by the frequency of revolution of the electron.

When the Hartree model or the independent particle model is introduced in (1') we may, according to the above, sum over

all final states k for each electron, without regard to the exclusion principle. We can then write, instead of (1'),

$$L = \frac{1}{Z} \sum_{i} \log \frac{2mv^2}{\hbar \omega_i}, \quad \log \omega_i = \sum_{k} f_{ik} \log \omega_{ik}, \tag{6}$$

where L contains only one simple summation over frequencies attributed to completely independent electrons. This formulation allows a direct application of correspondence arguments, as for one electron, and in the independent particle model the frequencies  $\omega_i$  will for high quantum numbers represent essentially the revolution frequencies for the electronic states in question. It is clear that, if one took the same electronic states in the independent particle model as in the Hartree model, the formula would lead to different results in the two instances, on account of the neglect of polarization effects in the former model.

The picture with independently moving electrons is thus characterized by certain frequencies of revolution in the classical limit, and it is based on an analogy with a single electron in a fixed potential. Now, we found in § 2 that the two kinds of frequencies one may imagine in the atom—the classical resonance frequency determined by the interaction, and the frequencies of revolution—behave in the same manner in the statistical description. It even appeared that a distinction between the two was rather artificial in the present dynamical problem. On account of this equivalence of the two kinds of frequencies it seems possible to picture the dynamical behaviour of the atom as being governed only by the interaction and inertia of the electron cloud, instead of by the frequencies of the independent particle model.

In order to appreciate the consequences of the interaction picture we shall first consider the effect of interaction in the simple case of an extended homogeneous electron gas. It has been shown by Kramers (1947) that the specific energy loss by a heavy charged particle in a gas of free electrons at rest is given by

$$\frac{dE}{dR} = \frac{4\pi z^2 e^4}{mv^2} \cdot \varrho \cdot \log \frac{2mv^2}{\hbar \omega_0},\tag{7}$$

where  $\varrho$  is the density of electrons, and  $\omega_0 = (4\pi e^2 \varrho/m)^{1/2}$  the classical resonance frequency of the medium.

Equation (7) shows that the frequency determining the adiabatic limit is just  $\omega_0$ . This result may be obtained directly when considering in more detail the competition between the polarization in the gas and the direct force from the penetrating particle (A. Bohr (1948)). In fact, for a collision with impact parameter p between an electron and the particle, the force on the electron will be approximately  $ze^2/p^2$ , and since the collision time is p/v the displacement in space of the electron during the impact must be  $(ze^2/mp^2)\cdot(p/v)^2=ze^2/mv^2$ . The electronic displacements give rise to a polarization force  $4\pi\varrho ze^4/mv^2$ , and accordingly the force from the particle will be compensated by the polarization for an impact parameter  $p=v/\omega_0$ , from which follows the formula of Kramers.

In this deduction the electrons were supposed to be at rest before the collision. But it can be shown easily that, even for a degenerate gas, the formula (7) remains valid for a penetrating particle of velocity high compared with the velocities of the electrons in the gas. Indeed, we may, as mentioned, neglect the exclusion principle in the calculation of the perturbed motion of the electrons, and it is then seen that the displacements of the individual electrons during the collision will be just as above. We thus find the same adiabatic limit as before, and, since the average energy transfer for a given impact parameter is to the first order independent of the electron velocities, we arrive again at formula (7).

In calculations of the effects of polarization, where one is concerned primarily with large impact parameters, the behaviour of the electrons can be described on classical mechanics when their quantum numbers are sufficiently high. Already from this circumstance it could be foreseen that for a free electron gas the adiabatic limit is determined by the classical resonance frequency, which is the only frequency defined in a classical description of the system.

For lower velocities of the particle the energy loss differs from that given by the equation (7). In the limit for very slow particles, Fermi and Teller (1947) have shown that the energy loss in a degenerate gas is approximately proportional to v. We shall here use a similar estimate by Lindhard (1953), where the logarithmic term per electron, L, is found to be approximately

$$L \approx \frac{1}{20} \left( \frac{2 \, mv^2}{\hbar \, \omega_0} \right)^{3/2} \tag{7'}$$

instead of the value  $L = \log (2 mv^2/\hbar \omega_0)$  in (7). The two formulae are to be joined for an argument somewhat larger than ten.

In this discussion of polarization effects we have used the conventional picture of a free electron gas. It may be that this picture does not represent closely an actual extended system of electrons. Still, it does seem to give a sufficient indication of the behaviour to be expected in atomic systems, when combined properly with the corresponding ideas regarding the orbital motions and revolution frequencies of the electrons.

We have seen that for the present purpose the extended electron gas may be described essentially as a compressible classical liquid of a given density of mass and charge. It is clear that, if the density of the system—and consequently also the classical resonance frequency  $\omega_0$ —varies only slowly in space, we can compute the total stopping of the particle by averaging (7) over space. But if we try to extend this liquid picture to the case of an atom we meet with the apparent difficulty that here the density varies quite rapidly in space. Still, let us tentatively apply the procedure of averaging (7) over the atom. This leads to the following expression for the logarithmic term in the stopping formula

$$L = \frac{1}{Z} \int d^3 r \cdot \varrho (r) \log \frac{2 m v^2}{\hbar \omega_0}, \tag{8}$$

where  $\omega_0^2 = 4 \pi e^2 \varrho(r)/m$  varies in space proportionally to the electron density  $\varrho(r)$ . It is now seen from the structure of formula (8) that it may be expected approximately to account for the stopping by heavier atoms. Indeed, in formula (7) a summation is supposed already to have been made over the distribution in momentum space. When we now average over ordinary space this will correspond to an integration over phase space. More precisely, we can consider (8) as an outcome of the Bethe formula (1') with generalized oscillator strengths, when the statistical model is applied in a literal manner and only the electronic interaction is included in the dynamics.

In order to illustrate the connection between the two pictures

used above we shall compare them for the case of an extremely simple statistical model of atoms. Let the orbital velocity and radius of the n'th electron in an atom be given by

$$a_{n} = a_{0} \frac{v^{2}}{n},$$

$$n = 1, 2, \dots, Z.$$

$$v_{n} = v_{0} \frac{n}{v},$$

$$(9)$$

where the effective quantum number  $\nu$  is supposed to be the same for all the electrons, and equal to a constant,  $\gamma$ , times  $Z^{1/3}$ . In this description the atoms have the same similarity as in the Thomas-Fermi model, and if  $\gamma$  is slightly less than 1, formula (9) even gives approximately the same density distribution as the Thomas-Fermi model for the major part of the atom. For the n'th electron we now find that the revolution frequency  $\omega_n = v_n/a_n = (v_0/a_0) \cdot (n^2/\nu^3)$  is exactly equal to the classical resonance frequency  $\omega_0$  for the density given by (9) at the distance  $a_n$  from the nucleus. The frequencies  $\omega_i$  and  $\omega_0(r)$  entering in (6) and (8), respectively, are thus the same and the two formulae give equal results.

Thus far, we have treated separately the revolution frequencies of the independent particle model and the classical resonance frequencies. As mentioned, it will seem difficult to distinguish between the two, and it is possible to describe the dynamics of the atom using only one of the two concepts. Nevertheless, for any particular model of the atom the magnitude of the separate contributions of the two frequencies is prescribed. When trying to find the total effect on the stopping we note that the force constants involved will be proportional to the squares of the frequencies. The total effective frequency squared is then the sum of the two squares. Since the two kinds of contributions behave in the same manner we can write

$$L = rac{1}{Z} \int d^3r \cdot arrho \left( r 
ight) \log rac{2 \, m v^2}{\chi \, \hbar \, \omega_0}, \qquad \qquad (8')$$

where  $\chi$  is a constant. The value of  $\chi$  we estimate from the simplified model (9) where  $\omega_n$  and  $\omega_0$  are equal in magnitude. On the basis of this result we assume in the following, for simplicity,

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the value  $\chi = 2^{1/2}$  for heavier substances. For the very lightest substances, where the polarization in the atom is of minor importance, the quantity  $\chi$  will be expected to approach the value 1.

The formula (8'), as it stands, should give an account of non-relativistic polarization phenomena in dense substances, and from the deduction of (7) it is clear that the formula will give a fair representation also of the stopping contribution from free electrons. An interesting question is here the effect of damping by resistance, due to random collisions of the freely moving electrons with the lattice. If the collision frequency were comparable with the resonance frequency an essential change would result in the stopping formula. However, in all known cases the collision frequencies are small, and the effects of resistance can be neglected. This circumstance was not recognized by Halpern and Hall (1948) in their treatment of polarization effects in carbon. The damping introduced by these authors leads to an anomalously high effective value of I in graphite ( $\sim 190 \text{ eV}$ ). Their result is at variance with recent measurements (BAKKER and Segrè (1951), I = 76 eV).

#### § 5. Theoretical Estimates of Stopping Power.

On the basis of equation (8') we can compute the excitation potential per electron,  $I_0$ , in Bloch's formula (2). The results for various atomic models are listed in Table 1. For the constant  $\chi$  we have chosen the value  $2^{1/2}$  introduced above. One finds approximate agreement with the empirical value of the Bloch constant, about 10 eV. The result of the Lenz-Jensen description appears to be a fairly good average of the Hartree model. The value for the Thomas-Fermi model is a little lower; it is characteristic that, while the average binding in the Thomas-Fermi model is closely equal to that in the Lenz-Jensen descrip-

Table 1. Values of the Bloch constant  $I_0 = I/Z$  (in eV), calculated from (8') with  $\chi = 2^{1/2}$ .

	Thomas-Fermi	Lenz-Jensen	Hartree	
			A	atomic Hg
$I_0$	8.9	10.7	11.0	9.6

tion, averages of the kind (8') are better represented by the latter model. Regarding the value of  $I_0$  for mercury we note that in the metallic state it will be slightly higher than for free atoms.

The agreement in Table 1 with the empirical values may be regarded as fortuitous, but it gives a useful guidance in the further treatment. The above results apply in the case of particles of velocity so high that the potential I remains velocity-independent. We shall now consider some of the aspects of the rather involved case presented for lower velocities, and in particular treat the simpler question of stopping for the lowest velocities covered by the statistical model, or  $0.1 \lesssim x \lesssim 5$ . As before, it can be useful to apply the less familiar picture of polarization when estimating the energy loss.

Formulae such as (6) show that I is no longer independent of velocity when the arguments  $2 m v^2 / \hbar \omega_i$  in the logarithmic terms are not large compared with unity. The resulting gradual change in I with velocity may just as well be calculated from the electron gas description corresponding to (7). In this picture the energy loss will be much reduced if  $\omega_{\rm max}=2mv^2/\hbar$  is of the order  $\omega_0$ , as seen from (7'). The reduction in the contribution to the energy loss from the individual electrons, or from the different regions in the atom, does not set in at all abruptly, but the summation over electrons with widely different revolution frequencies makes it natural to proceed at first as if the change were abrupt. We then assume that the low frequencies in the atom contribute in the usual manner to the energy loss, while the high frequencies give no contribution, the division between high and low frequencies being given by  $\omega_{\rm max}/C$ , where C is a constant. The effect of this cut-off procedure can be seen most directly from the simplified atomic model (9). Indeed, the more refined statistical models of the atom lead in this case essentially to the same result. Using (9) we now sum in (8') over the frequencies less than  $\omega_{\rm max}/C$  and find

$$L = \frac{(2\gamma)^{3/2} v}{v_0 Z^{1/2}} \cdot \frac{1 + \log C^{1/2}}{C^{1/2}},\tag{10}$$

where the effective quantum number v is written as  $\gamma \cdot Z^{1/3}$ . The specific energy loss thus becomes proportional to  $Z^{1/2}/v$ , corresponding to the simple law for  $\alpha$ -rays mentioned earlier. This

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derivation is similar to that used by N. Bohr (1948), equ. (3.5.7), but the resulting formula is somewhat different.

As to the value of the constant of proportionality in (10), we see that it varies only slowly with the cut-off C. The parameter  $\gamma$  is now so adjusted as to give for high velocities the correct value of  $I_0 = 10$  eV; this requires  $\gamma = 0.71$ . We can then compare with the approximate value of L determined from the experiments at low velocities, which lead to  $L = 1.35 \cdot x^{1/2}$  (see Fig. 2). The value required for C from this is C = 5.6. Such a high value for the cut-off in the energy loss is in line with the discussion by Bethe for hydrogen, or K-shells in general, where the reduction is considerable when v is of the order of the electron velocity, i.e. for a value of the argument in the logarithm much larger than unity.

A similar result is obtained when formula (7') is applied. In the atomic model (9) the integration is performed over the electrons for which respectively (7) and (7') are applicable. This gives

$$L = 1.36 \cdot x^{1/2} - 0.016 \cdot x^{3/2},\tag{11}$$

and it so happens that formula (11) for lower values of x is in close agreement with the experiments shown in Fig. 2. The full-line curve on the figure represents (11). If one allows for an uncertainty by a factor 2 in (7') a latitude will result of about  $10^{-0}/_{0}$  in the formula (11). The formula joins smoothly to the one which applies for high velocities at a value of x equal to 19.

The variation of (11) with x is qualitatively of the kind found on Fig. 1. For increasing x the effective potential defined by (11) =  $\log (2 mv^2/I)$  increases from low values to a maximum of about 16–17 eV, occurring at  $x \sim 5$ , and then decreases to 10 eV. The gradual cut-off (7') is in this respect superior to the abrupt one leading to (10), because it is effective for considerably higher values of x. But even though the initial part of the curve (11) is not wrong the maximum in I is too narrow, and for x between about 6 and 30 the curve should lie somewhat lower.

One might suppose that the shortcomings of formula (11) for higher values of x are due to the defects of the simplified atomic model (9), where the firmest electron bindings are not properly accounted for. One could then attempt to use, e.g., the Lenz-

Jensen model. This leads to a slight improvement for higher velocities, but only part of the experimental deviation from the Bloch formula can be accounted for. For low velocities the Lenz-Jensen description gives even worse agreement with the experiments, which indicates that the good agreement of (11) is somewhat accidental.

We shall not attempt further improvements of the present treatment, which would appear to demand not merely a more detailed picture of the electronic states, but rather a description of the adiabatic effects and the polarization considerably more elaborate than in the present discussion.

Let us return to the question of the calculation of the average excitation potential I in the limit of high velocity of the particle. In the first part of this paragraph we found values of I from various atomic models. As soon as a more detailed atomic description as the Hartree model is used there will appear minor individual deviations from the relation of Bloch (2). This is indicated in Table 1, for argon and atomic mercury. Similar differences occur when the bindings in molecules or solids are taken into account. One can attempt to evaluate such differences on the basis of equation (8'). However, estimates of this kind are not expected to be very accurate, since they involve the most loosely bound electrons in atoms.

In order to see how much the formula (8') can be in error for the most loosely bound electrons we use it in the case of the lightest elements, where it should be least applicable. We introduce the actual density distributions in the light atoms and find then from (8') a corresponding potential I. For these substances it will be natural to put  $\chi=1$ , because in the dynamics each electron moves in a nearly static atomic field, the separate effects of polarization being small. The results are shown in Table 2, where for molecular hydrogen we have simply put Z=1.2. For helium we have used hydrogen wave functions, with Z=1.69. The valence electrons in metallic lithium are assumed to be distributed with constant density in space. These rough descriptions of the lightest elements will be accurate enough for our purpose.

The approximate correctness of the results in Table 2 and Table 1 seems to indicate that one can estimate the changes  $\Delta I$ 

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Table 2. Comparison between the measured I (in eV), and the value given by (8') with  $\chi$  = 1. The measurements are by Mano (1934), and Bakker and Segrè (1951).

	Formula (8')	Bethe's formula	Exp.
$rac{1}{2} extbf{H}_2$	16	17.6 <sup>2)</sup>	15—16
He	37	43	35
solid Li	36	45 <sup>1)</sup>	3437

<sup>1)</sup> Estimate by A. Bohr (1948).

in I due to chemical bindings, or due to deviations from the statistical model for different atoms, by introducing the density changes in formula (8'). The calculated values of  $\Delta I$  can hardly be more in error than corresponding to the latitude in the values of  $\chi$ . The changes  $\Delta I$ , thus obtained, can never become very large, so that only minor deviations from the Bragg rule will occur for a particle of high velocity. We note that this conclusion disagrees with some of the measurements concerning liquid water and water vapour, where considerable deviations from the Bragg rule have been reported (cf. Platzman (1952)).

#### § 6. Straggling in Energy Loss and Range.

The straggling in energy loss, or straggling in range, can be calculated in a direct manner when the cross sections for the individual possible energy transfers are known (N. Bohr (1948)). We shall for the present consider only the case where the particle has penetrated a layer of thickness sufficient to ensure that the distribution in energy loss is approximately Gaussian. The average square fluctuation in energy loss then determines the distribution completely, and it is given by the familiar formula

$$\Omega^{2} = \langle (dE - \langle dE \rangle)^{2} \rangle = NdR \sum_{i} T_{i}^{2} \sigma_{i}, \qquad (12)$$

where  $\sigma_i$  is the cross section for energy loss  $T_i$ . The contributions to the straggling are thus weighted towards the close collisions, and the effect becomes independent of the screening

<sup>2)</sup> PLATZMAN (1952) quotes the value 19 eV.

in the distant collisions if the velocity v is large. For a fast particle the cross section for energy transfer T in close collisions is

$$d\sigma = \frac{2\pi z^2 e^4}{mv^2} \cdot \frac{dT}{T^2} \left( 1 - \beta^2 \frac{T}{T_{\text{max}}} \right), \tag{13}$$

where relativistic effects are included, and  $\beta = v/c$ . We assume that the particle is so heavy that the maximum energy transfer to an electron is  $T_{\text{max}} = 2 \ mv^2/(1-\beta^2)$ . The straggling in energy loss is accordingly

$$\Omega^{2} = 4 \pi z^{2} e^{4} N \cdot dR \cdot Z \frac{1 - \frac{\beta^{2}}{2}}{1 - \beta^{2}}, \tag{14}$$

which leads to an average square fluctuation in range given by

$$(\Delta R)^{2} = 4 \pi z^{2} e^{4} N Z \int_{0}^{\bullet E} \left( \frac{dE'}{dR'} \right)^{-3} \frac{1 - \frac{\beta^{2}}{2}}{1 - \beta^{2}} dE', \qquad (15)$$

where  $\beta = \beta(E')$ .

For lower velocities the problem is more involved and, as for the average energy loss, one will expect that the more strongly bound electrons give reduced contributions. The discussion here is in line with that in the preceding paragraphs, but of a more qualitative kind. Of course, the same holds for the measurements where the straggling cannot be determined as accurately as the range or specific energy loss.

While the absolute value of the straggling per unit path is a constant for high, but non-relativistic, velocities it will for lower velocities decrease towards zero. Since this is true for each separate frequency in the atom, one may introduce a cut-off for a suitable value of  $2 mv^2/\hbar \omega_0$ , and in this way estimate the straggling in collisions with atoms. The important question is then the place at which the reduction sets in. This can be estimated by taking, for instance, the result for the case of a free gas. It is here convenient to quote the value of the straggling relative to the energy loss, which quantity is a measure for the magnitudes of the energy transfers in the collisions. For a free Fermi gas of high density one finds roughly, in the limit of low velocities (Lindhard (1953)),

$$\frac{\Omega^2}{dE} = v \left(5 \, m\hbar \, \omega_0\right)^{1/2},\tag{16}$$

corresponding to an effective cut-off at about  $2 mv^2/\hbar \omega_0 \cong 3$ .

In collisions with atoms sufficiently heavy for the statistical model to apply, the relative straggling is according to (16) of the form

$$\frac{\Omega^2}{EdE} = \frac{m}{M} \cdot u(x),\tag{17}$$

where M is the mass of the particle. When integrating over the atom, using the formulae (14) and (16) in the atomic model (9), it is found that for low values of x the function u tends to a constant value, given approximately by

$$u(x) \cong 1, \quad x \lesssim 3. \tag{17'}$$

This result for the relative straggling coincides with that found by N. Bohr (1948). For higher velocities the straggling approaches smoothly the value given by (14), and one may write

$$u(x) = \frac{2}{L(x)},\tag{17"}$$

where the formula holds when relativistic corrections can be neglected.

The straggling in range for lower velocities can be found directly from (17'). We obtain here

$$\frac{(\Delta R)^2}{R^2} = \frac{3}{4} \frac{m}{M}, \qquad x \lesssim 5. \tag{18}$$

For higher values of x one can derive from (15) the approximate formula (cf. N. Bohr (1948), equ. (5.2.7))

$$\frac{(\Delta R)^2}{R^2} = \frac{2}{L(x)} \frac{m}{M} \left( 1 - \frac{2v^2}{5c^2} \right),\tag{19}$$

where terms in v/c are included up to second order. The simple formula (19) is not much in error for proton energies even as high as those used in the measurements of Mather and Segrè (1951). It should be mentioned that these authors employed an

essentially non-relativistic formula for the straggling in range, different from (15) and (19).

The measurements of straggling in energy loss are mainly in the region of high values of x. As regards a comparison with experiments for cases where also smaller values of x enter, and equation (17') should apply, the reader is referred to a recent publication by C. B. Madsen (1953).

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Institute for Theoretical Physics, University of Copenhagen, Denmark.

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