## Study on the angular dependence of the average energy loss for ions in solids

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By using the transport theory, we have derived a general expression which connects  $\Delta E(\theta)$ , the average energy loss measured at different  $\theta$  emergence angles, and  $Q(\theta)$ , the average elastic and inelastic energy loss in a single scattering as a function of the scattering angle  $\theta$ , for ions transmitted through thin films. In this way, the effect of multiple scattering on the angular dependence of the energy loss is properly taken into account. By means of this procedure the average energy-loss function Q is retrieved from experimental  $\Delta E(\theta)$  data, as we show for the cases of 50–200-keV H<sup>+</sup> on C and Al foils.

## I. INTRODUCTION

The purpose of this paper is to present a theoretical study on the relation between the average energy lost by ions transmitted through a thin foil within a given scattering angle and that corresponding to a single collision. As is well known, the slowing down of ions in matter can be characterized primarily by the stopping power, or the average energy loss per unit path length |dE/dR|, given by

$$\left|\frac{dE}{dR}\right| = N \int d\sigma Q , \qquad (1)$$

where Q is the average energy loss per collision,  $d\sigma$  the differential scattering cross section, and N the atomic density of the target material.

One of the most widespread methods employed to measure the stopping power is the so-called transmission experiment, where a well-collimated ion beam passes through a thin solid film. The energy of those particles emerging within a given angle, usually in the forward direction, is then measured. The stopping power |dE/dR| is approximated by the quotient  $|\Delta E/\Delta R|$  of the observed energy loss and the foil thickness, respectively.

This procedure has, however, the following shortcoming: On one hand, a foil as thin as possible is desirable in order to keep the ion energy well defined. On the other hand, as the thickness of the foil decreases the observation angle cannot be reached through arbitrary scattering angles. Under this circumstance the measurements can no longer be identified with the stopping power as given by (1) since in the integration there is no restriction on the final direction of motion. The existence of angular as well as thickness dependences of the stopping power measured with very thin films<sup>1-5</sup> corroborates the above statement.

It is thus of interest to analyze in some detail the applicability of Eq. (1) for a given experimental condition. An appropriate procedure to deal with this problem is the transport equation. In the following section we obtain a general expression which states the relation between  $Q(\phi)$ , the average energy loss in a single collision characterized by the scattering angle  $\phi$ , and  $\Delta E(\theta)$ , the average energy loss of particles transmitted within an angle  $\theta$  in a beamfoil experiment. In Sec. III some consequences of our theoretical results are discussed. In Sec. IV a procedure is shown to obtain a  $Q(\phi)$  from  $\Delta E(\theta)$ . Finally, as an example, we apply the method to measurements of energy loss as a function of angle for 50–200-keV H<sup>+</sup> on Al and C foils. We focus our attention on light ions since they are more attractive than heavier ones because electronic processes dominate their slowing down and these are still subject to investigation.

### **II. CALCULATION**

Let us introduce the function  $F(z, \vec{\theta}, E)$  representing the distribution of particles after traveling a distance z in a solid within angular and energy intervals  $(\vec{\theta}, d\vec{\theta})$  and (E, dE), respectively, during bombardment with a well-collimated beam of particles of energy  $E_0$ . Here  $\vec{\theta}$  represents the component of the particle direction of motion perpendicular to the beam. F obeys the transport equation<sup>6,7</sup>

$$\frac{\partial F(z,\vec{\theta},E)}{\partial z} = N \int d\vec{p} [F(z,\vec{\theta}-\vec{\phi},E+Q)-F(z,\vec{\theta},E)] \quad (2)$$

with the boundary condition

 $F(z=0,\vec{\theta},E)=\delta(\vec{\theta})\delta(E-E_0)$ ,

where  $\vec{p}$  is the impact parameter,  $\vec{\phi}$  represents the change on the direction of motion of the particle due to the scattering, Q is the average energy loss corresponding to such an impact parameter, and N is the atomic density.

If the energy of transmitted particles does not differ significantly from that of the incident beam, we can then assume the scattering law  $\vec{\phi}(\vec{p})$  as well as the energy loss Q to be energy independent. Hence, Eq. (2) can be easily solved by using the Fourier-transform technique:

$$F(z,\vec{\theta},E) = \frac{1}{(2\pi)^3} \int d\vec{k} \, d\omega \exp[i\omega(E_0 - E) - i\vec{k}\cdot\vec{\theta} - zN\sigma(\vec{k},\omega)], \qquad (3)$$

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where

$$\sigma(\vec{\mathbf{k}},\omega) = \int d\vec{\mathbf{p}}(1 - e^{i\vec{\mathbf{k}}\cdot\vec{\phi} - i\omega\mathbf{Q}}) .$$
(4)

We want to point out that Eq. (3) has thus far been obtained by Meyer *et al.*,<sup>6</sup> but they solved it by introducing an unrealistic expression for Q as discussed in a recent article by Wedell *et al.*<sup>7</sup>

Let us now calculate the average energy of the particles transmitted through a *t*-thick foil at a given angle  $\theta$  disregarding path length enlargement effects (see the Appendix), i.e.,

$$\langle E(t,\vec{\theta}) \rangle = \frac{\int dE \, EF(t,\vec{\theta},E)}{\int dE \, F(t,\vec{\theta},E)} \,. \tag{5}$$

After some algebra we obtain from Eq. (3)

$$\langle E(t,\vec{\theta})\rangle = E_0 - \frac{tN \int d\vec{p} Q(p) F_{MS}(t,\vec{\theta}-\vec{\phi}(\vec{p}))}{F_{MS}(t,\vec{\theta})} , \qquad (6)$$

where

$$F_{MS}(t,\vec{\theta}) = \int F(t,\vec{\theta},E) dE$$

is the angular distribution function of particles transmitted irrespective of their energy; in other words, the multiple-scattering distribution.

If scattering angle is used instead of impact parameter, Eq. (6) changes to

$$\langle E(t,\vec{\theta})\rangle = E_0 - \frac{tN\int d^2\sigma(\vec{\phi})F_{MS}(t,\vec{\theta}-\vec{\phi})Q(\vec{\phi})}{F_{MS}(t,\vec{\theta})} , \qquad (7)$$

where Q is written now as a function of the scattering angle. Since Q and  $d^2\sigma(\vec{\phi})$  do not depend on the scattering angle direction, we may write

$$\langle E(t,\vec{\theta})\rangle = E_0 - \frac{tN \int d\sigma(\phi) G_{MS}(t,\theta,\phi) Q(\phi)}{F_{MS}(t,\vec{\theta})} , \quad (8)$$

where

$$d^{2}\sigma(\vec{\phi}) = d\sigma(\phi) \, d\alpha/(2\pi) ,$$
  

$$G_{MS}(t,\theta,\phi) = \frac{1}{2\pi} \int_{0}^{2\pi} d\alpha \, F_{MS}(t,\vec{\theta}-\vec{\phi}) ,$$

 $\alpha$  being the angle between vectors  $\vec{\theta}$  and  $\vec{\phi}$ .

# III. DISCUSSION OF THE ANALYTICAL RESULTS

It is illustrative to analyze Eq. (7) in some detail. We can see how the average energy loss in a single scattering is related to the observed energy loss  $[E_0 - E(t, \vec{\theta})]$  in a transmission experiment. For example, as the thickness decreases  $(t \rightarrow 0)$ 

$$F_{MS}(t,\vec{\theta}) \rightarrow \delta(\vec{\theta}) , |\theta| \simeq 0$$

and

$$F_{MS}(t,\vec{\theta}) \rightarrow tN\sigma(\vec{\theta}), |\theta| \neq 0$$

 $\sigma(\vec{\theta})$  being the differential scattering cross section. So Eq. (7) changes to

$$\langle E(t,\vec{\theta}) \rangle = E_0 - Q(\theta), t \to 0 \text{ and } |\theta| \neq 0.$$
 (9)

As expected, if the thickness tends to zero the measured energy loss corresponds to that of single scattering. This approximation was recently used by Iferov and Zhukova<sup>2</sup> to obtain Q from  $\Delta E(t,\theta)$  measurements; however, for their foil thicknesses the application of Eq. (9) leads to erroneous Q estimations.

On the other hand, when t is sufficiently large,  $F_{MS}$  is a smooth function of the angle  $\theta$ ; therefore,  $F_{MS}(t, \vec{\phi} - \vec{\theta})$  approximately cancels with  $F_{MS}(t, \vec{\theta})$  in Eq. (7). Thus, we have

$$\langle E(t,\vec{\theta}) \rangle = E_0 - tN \int d^2 \sigma(\vec{\phi}) Q(\phi) .$$
 (10)

Observe that Eq. (10) is the formula commonly used in the stopping-power literature. We are now in a position to show in which cases Eq. (10) is applicable for a given experimental condition.

By the way, it is worthwhile to point out that Eq. (7) or (8) contains the answer to one of the most frequent questions made by stopping-power experimentalists: How frequently does an individual scattering angle having modulus between  $\phi$  and  $\phi + d\phi$  take place during the passage of a particle in a *t*-thick film, when the observation angle  $\vec{\theta}$  is fixed? Note that if no condition on the observation angle is given, or the particle detector has a wide angular acceptance, the answer is just

 $tN\,d\sigma(\phi)$ .

Therefore, by direct comparison with Eq. (8) we may conclude that the restriction imposed by the observation angle is taken into account by the function<sup>8</sup>

$$\frac{G_{MS}(t,\theta,\phi)}{F_{MS}(t,\vec{\theta})} . \tag{11}$$

To calculate this function, we introduce the Gaussian approximation for the multiple-scattering distribution

$$F_{MS}(t,\vec{\theta}) = C(t) \exp\left[-\frac{\theta^2}{\Omega^2(t)}\right], \qquad (12)$$

where  $\Omega^2(t)$  is a measure of the angular spread of the distribution and C(t) a normalization factor. The Gaussian approximation for  $F_{MS}$  is only valid in the limit of infinite thickness; nevertheless, we will use it here only to obtain a simple analytical expression for  $G_{MS}$ :

$$G_{MS}(t,\theta,\phi) = C(t) \exp\left[-(\theta^2 + \phi^2)/\Omega^2(t)\right] I_0 \left[\frac{2\theta\phi}{\Omega^2(t)}\right],$$
(13)

where  $I_0(x)$  is the modified Bessel function of the zeroth order.<sup>9</sup>

Afterwards, when calculating the average single collision energy loss, we will perform a numerical calculation of  $G_{MS}$  using real multiple-scattering distributions.

By replacing (12) and (13) into (11), we have

$$\frac{G_{MS}(t,\theta,\phi)}{F_{MS}(t,\vec{\theta})} = e^{-\phi^2/\Omega^2(t)} I_0 \left[ \frac{2\theta\phi}{\Omega^2(t)} \right] . \tag{14}$$

Figure 1 shows the quotient given by Eq. (14) as a function of individual scattering angle  $\phi$  and for different observation angles  $\theta$ , both in units of  $\Omega$ . We observe in Fig. 1 that when the scattering angle tends to zero the function (14) tends to unity independently of  $\theta$ ; in other words, scattering angles  $\phi$  smaller than  $\Omega$  occur with nearly the same frequency irrespective of the observation angle. On the other hand, scattering angles  $\phi$  greater than  $\theta + \Omega$ should be considered as very unlikely. We find also that in the case of  $\theta > \Omega$ , Eq. (14) reaches its maximum at  $\phi$ values around  $\theta$ . In all cases the value of (14) at the maximum increases monotonically with  $\theta$ .

#### IV. OBTAINING $Q(\phi)$ FROM $\Delta E(\theta)$

The connection that we have just established between the average energy loss per collision  $Q(\phi)$  and the energy lost after traversing the foil  $\Delta E(\theta)$  allows us to obtain one of these functions in terms of the other. It is clear that this connection works well when  $Q(\phi)$  is known, since the  $\Delta E(\theta)$  is obtained explicitly by use of expression (8). Unfortunately, there are no calculations available for  $Q(\phi)$ for the systems and velocities of interest. To retrieve  $Q(\phi)$  from a knowledge of  $\Delta E(\theta)$ , for example, from experimental measurements, is also possible, at least in principle, but we should mention that  $\Delta E(\theta)$  is greatly dependent on individual collisions with small scattering angles as can be seen in Fig. 1 and, therefore, the bulk of the  $\theta$ dependence of  $\Delta E(\theta)$  comes from  $Q(\phi)$  for small  $\phi$  and the action of the multiple scattering. Under these cautious considerations, we will now study the way in which we can obtain the average energy lost in a single-scattering process  $Q(\phi)$  from the measured mean energy loss as a function of the emergence angle  $\Delta E(\theta)$ .

Our aim is to solve Eq. (8) which is an integral equation. For a set of experimental values we have



FIG. 1. Plot of the function (14) evaluated with a Gaussian approximation for the multiple-scattering distribution. It gives the weight of single-scattering angles  $\phi$  when the observation angle is  $\theta$ . Angles are measured in units of  $\Omega$ , the angular spread of the multiple-scattering distribution.

$$\Delta E(\theta_j) = E_0 - \langle E(t,\theta_j) \rangle$$

$$= \frac{tN \int d\sigma(\phi) G_{MS}(t,\theta_j,\phi) Q(\phi)}{F_{MS}(t,\vec{\theta}_j)}$$
(15)

with j = 1, ..., n, n being the number of experimental points. Here we are taking t as the foil thickness which implies disregarding path length enlargement effects (see the Appendix).

A solution of Eq. (15) can be found by assuming a functional form for Q, and adjusting their parameter values to minimize, for instance, the sum of the squared errors

$$\sum_{j} \epsilon_{j}^{2} , \qquad (16)$$

where

$$\epsilon_{j} = \Delta E(\theta_{j}) - \frac{tN \int d\sigma(\phi) G_{MS}(t,\theta_{j},\phi) Q(\phi)}{F_{MS}(t,\vec{\theta}_{j})} .$$
(17)

The function Q can be split into two components:

 $Q = Q_e + Q_n$  ,

where  $Q_n$  is the nuclear or elastic energy loss and  $Q_e$  is the inelastic energy loss. For small scattering angles  $Q_n$  can be approximated by

$$Q_n \simeq rac{M_1}{M_2} \phi^2 E$$
 ,

where  $M_1$  and  $M_2$  are the projectile and target masses, respectively.

In search of analytical expressions for  $Q_e$ , we find that there are basically two kinds of theoretical models which supply impact-parameter-dependent  $Q_e$  functions. They are those based on a first-order Born approximation<sup>10,11</sup> valid for high ion velocities, and those who start from Firsov's model<sup>12,13</sup> which are specially designed for low ion velocities and atomic numbers of the colliding partners similar to each other and both substantially greater than unity. Even though these theories are valid in rather unconnected ranges of energies and of colliding species, they both have the same behavior as far as the impact-parameter dependence is concerned; that is,  $Q_e$  decreases monotonically with the impact parameter p and reaches its maximum value at p = 0. Bearing in mind this just-mentioned characteristic behavior of  $Q_e$  predicted by both groups of models we propose the following ad hoc expression:

$$Q_e(\phi) = Q_{e,m} \frac{\phi^{\nu}}{\phi^{\nu} + \phi_0^{\nu}},$$
 (18)

where  $\phi$  is the scattering angle and  $Q_{e,m}$ ,  $\phi_0$ , and  $\nu$  are variational parameters. By introducing (18) in the numerical scheme given by (16) and (17) we process the experimental data and obtain the  $Q_e$  function.

As pointed out at the beginning of this section, the inversion procedure does not yield, in general, a unique  $Q(\phi)$ -function, especially in the range of large  $\phi$  values; furthermore, as foil thickness increases, the results become less confident. We must take them as Q functions which

reproduce the measured energy loss as a function of angle within uncertainties of the order of few percents. Discussion of the results is presented in Sec. V.

### V. COMPARISON WITH EXPERIMENT

In Fig. 2 we display some experimental results from measurements performed in our laboratory for H<sup>+</sup> bombarding Al and C foils.<sup>14</sup> Details of the equipment can be found elsewhere.<sup>14,15</sup> The figure shows the difference between the energy loss  $\Delta E(\theta)$  measured at a given angle  $\theta$  and that of zero angle  $\Delta E(0)$ . The angular range covers approximately three half-widths of the angular spectra which, by the way, are in good agreement with theoretical multiple-scattering distributions.<sup>16</sup> In order to illustrate the importance of this effect let us mention that the average zero angle energy losses are as follows: 2230, 2020, and 1720 eV for 50-, 100-, and 200-keV protons on Al, respectively, and 5280 eV for 200-keV protons on C.

In Fig. 3 we show the  $Q_e$  functions calculated from the experiments by using our previous theoretical results. In these calculations, we have used the multiple-scattering distribution function corresponding to a Thomas-Fermi interatomic potential.<sup>16</sup> In addition to that, we used the differential scattering cross section for the same potential in the analytical approximation contained in Ref. 17. This gives self-consistency to our theory since, in the derivation of the multiple-scattering distributions, the same scattering cross sections were used. On the other hand, these expressions are based on classical scattering treatment; therefore, they are not valid for sufficiently high velocities or very low scattering angles.

Our first observation from Figs. 2 and 3 is that at large  $\theta$ , the obtained single-scattering energy-loss function  $Q_e$  lies well below the corresponding  $\Delta E(\theta) - \Delta E(0)$  at the same angle. This is an important feature since it tells us that we cannot identify the difference  $\Delta E(\theta) - \Delta E(0)$  with  $Q_e(\phi)$  for  $\phi = \theta$  in a straightforward manner as was done elsewhere.<sup>2</sup> This peculiar result is due to the fact that there are, roughly speaking, two kinds of contributing processes to the difference  $\Delta E(\theta) - \Delta E(0)$ : a scattering



FIG. 2. Difference between the energy loss measured (Ref. 14) at a given angle  $\theta$  and that at angle zero for protons traversing 170-Å aluminum and 410-Å carbon foils at various energies.  $\Box$ : 50-keV H<sup>+</sup> on Al.  $\Delta$ : 100-keV H<sup>+</sup> on Al.  $\odot$ : 200-keV H<sup>+</sup> on C. For the case of 100-keV H<sup>+</sup> on Al, also, the energy loss integrated over all angles was determined. Its difference with the zero angle energy loss is indicated by the arrow on the left.



FIG. 3. Calculated inelastic energy losses for single-scattering events of angle  $\phi$ . Symbol convention: same as Fig. 2.

of angle  $\phi \simeq \theta$  and a series of rather small-angle scatterings. It so happens that, for a certain intermediate foil thickness, the latter kind of scatterings sum up to a considerable energy loss, causing the  $\Delta E(\theta) - \Delta E(0)$  curve to lie above the calculated  $Q_e$  values.

We can also see in Figs. 2 and 3 that when going from the energy-loss difference  $\Delta E(\theta) - \Delta E(0)$  to  $Q_e(\theta)$ , the effect of the multiple scattering as well as of the scattering cross section strongly depends on the projectile energy. For instance, the energy-loss differences belonging to H<sup>+</sup> on Al at 100 and 200 keV lie a factor of 4 above those of H<sup>+</sup> on Al at 50 keV for large angles; however, the corresponding  $Q_e$  values differ in a factor of the order of 2. It should be mentioned that, for our cases, nuclear energy loss  $Q_n$  as well as path length enlargement can be neglected.

#### VI. CONCLUDING REMARKS

We offer a way to analyze the observation angle dependence of the energy loss  $\Delta E(\theta)$  through a single-scattering energy-loss function  $Q(\phi)$ , where  $\phi$  is the scattering angle. By means of the transport theory we obtained the equation (8) which connects  $\Delta E(\theta)$  with  $Q(\phi)$ . We have studied the cases of  $H^+$  bombarding Al and C foils in the 50-200-keV energy range and obtained from them suitable  $Q_e$  functions. It is found that  $Q_e$  functions are not related to  $\Delta E(\theta)$  in a straightforward manner, but by an integral equation. This equation (8) also provides a way to analyze, for a given experimental condition, which kind of scattering angles have suffered those particles we are observing. This should be taken into account in highaccuracy stopping-power measurements by thin foil transmission experiments. For instance, roughly speaking, those ions emerging from a thin foil in the forward direction have not undergone scattering angles greater than that of the width of the angular distribution, a result which is quite obvious; however, it claims for a careful analysis of the effect of this type of scattering event on the average energy loss. It follows from our study that besides nuclear energy loss, the inelastic energy loss might depend on foil thickness too. Finally, it is clear that a theoretical calculation of  $Q_e$  would be valuable in order to allow the extension of current stopping-power investigation towards its angular dependency.

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### APPENDIX

As mentioned in Sec. V, the stopping power may depend on the emergence angle since the path length increases with this angle due to a simple geometrical effect. The trajectory of a projectile through the foil is determined by multiple scatterings, and its path length z fluctuates around a mean value which depends on the emergence angle.

Going back to Eq. (5) let us recall that the variable z which appears in our calculation represents the distance traveled by the ion along its path. Therefore, Eq. (5) really establishes the average energy of particles at a fixed path length. In order to compare (5) with experimental data, which are obtained at a fixed foil thickness, we may replace t by  $\langle z \rangle$  in this expression. Here  $\langle z \rangle$  means the average path length as a function of the emergence angle  $\theta$ and foil thickness t. Thus, we have

$$\langle E(t,\vec{\theta}) \rangle \simeq \frac{\int dE \, EF(z,\vec{\theta},E)}{\int dE \, F(z,\vec{\theta},E)} \,.$$
 (A1)

Let us consider

$$z = t + \Delta$$
, (A2)

where  $\Delta$  represents the path length excess due to multiple scattering and geometry. By expanding (A1) in a power series of  $\Delta$ , and keeping up to the first-order term, we obtain

$$\langle E(t,\vec{\theta}) \rangle \cong \frac{\int dE \, EF(t,\vec{\theta},E)}{\int dE \, F(t,\vec{\theta},E)} + \Delta \left| \frac{\partial}{\partial z} \left[ \frac{\int dE \, EF(z,\vec{\theta},E)}{\int dE \, F(z,\vec{\theta},E)} \right] \right|_{z=t} .$$
 (A3)

In order to evaluate the derivative in the last term we may

approximate

$$\frac{\int dE \ EF(z,\vec{\theta},E)}{\int dE \ F(z,\vec{\theta},E)} \sim E_0 - NSz , \qquad (A4)$$

where NS is the stopping power; thus, we have

$$\langle E(t,\vec{\theta}) \rangle \sim \frac{\int dE \, EF(t,\vec{\theta},E)}{\int dE \, F(t,\vec{\theta},E)} - NS\Delta \;.$$
 (A5)

Equation (A5) implies that Eq. (5) can be used to compare with experimental data setting z = t if a correction by excess of path length  $\Delta$  is made. Let us, therefore, evaluate this quantity  $\Delta$  in what follows. It can be shown<sup>18</sup> that the average path length is given in terms of the foil thickness t and the emergence angle  $\theta$  by the expression

$$\langle z \rangle = t + \frac{1}{2} \frac{\int d\vec{\alpha} \, \alpha^2 \int_0^t ds \, F_{MS}(s,\vec{\alpha}) F_{MS}(t-s,\vec{\theta}-\vec{\alpha})}{F_{MS}(t,\vec{\theta})} \,. \tag{A6}$$

We can solve Eq. (A1) analytically by introducing the Gaussian approximation:

$$F_{MS}(t,\vec{\theta}) = C(t)e^{-\theta^2/\Omega^2(t)}$$
,

where

$$C(t) = \frac{\lambda}{\pi t}$$

and

$$\Omega^2(t) = t/\lambda$$
.

Here

$$\lambda = N \int d\vec{\theta} \, \vec{\theta}^2 \sigma(\vec{\theta})$$

where N is the atomic density and  $\sigma$  is the differential elastic scattering cross section. Moreover, we assume that this integral does not diverge. So we obtain

$$\Delta = \frac{t\Omega^2(t)}{12} \left[ 1 + \frac{2\theta^2}{\Omega^2(t)} \right]$$
(A7)

which coincides with a previous result of Yang.<sup>19</sup>

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