Comparison of calculational methods for straggling in thin absorbers*

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The solution of the transport equation by the Laplace-transform method, introduced by Landau and modified by Vavilov and others, is compared with the solution of that equation by what we term a convolution method. An energy-loss cross section determined for solid aluminum has been used as input to the calculations. For very small values of absorber thickness, the straggling distribution is dominated by peaks at multiples of the Al plasmon frequency. At intermediate values, these peaks are a small structure superimposed on a broad energy-loss distribution which is not described well by any of the Laplace-transform theories. At larger values, the straggling distribution consists of one peak which is well described by the Vavilov theory.

I. INTRODUCTION

Statistical fluctuations in the energy losses experienced by fast charged particles penetrating thin targets have been of interest for many years. The major thrust of the study of the straggling process began with the work of Landau[1] in 1944. The subject has a practical application to particle identification by use of solid-state detectors[2] and low-pressure gas proportional counters.[3]

Landau introduced a straggling function $f(x, \Delta)$ which gives the probability that a particle having penetrated a thickness x of an absorber will have lost an energy Δ, small compared to its initial kinetic energy. (For convenience, symbols are summarized in Table A I.) This probability density function $f(x, \Delta)$ is given as the solution of the transport equation:

$$\frac{\partial f(x, \Delta)}{\partial x} = \int_0^\infty w(E) f(x, \Delta - E) dE - f(x, \Delta)\sigma_t , \tag{1}$$

where $w(E)$ is the differential collision cross section for single collisions with an energy loss E and $\sigma_t = \int_0^\infty w(E) dE$ is the total collision cross section. It may be noted that the macroscopic cross section referred to here has dimensions of inverse length. It is given by the microscopic cross section per atom or molecule times the number density of atoms or molecules in the absorber. In writing a transport equation, it is implicitly assumed that each collision suffered by the incident particle is independent of its previous history, i.e., that one has a stochastic process. For example, channeling processes are excluded from this discussion.

All the theoretical descriptions of the straggling process have been based on the transport equation. The methods of obtaining $f(x, \Delta)$ from Eq. (1) can be divided into two major categories—those based on Laplace-transform methods, including the original work of Landau[1] and Symon[4] and the modification of Blunck and Leisegang,[5] Vavilov,[6] Shulek *et al.*,[7] and Bichsel[8]; and that which we may term the convolution method, first described by Williams,[9] later by Herring and Merzbacher[10] and implemented by Kellerer.[11] We shall not consider a third approach based on a theory of moments employed by Tschalär.[12]

The Laplace-transform methods differ from each other chiefly in the form assumed for the single-collision spectrum $w(E)$ and in the corrections employed, if any, to account for the difference between the assumed $w(E)$ and a realistic spectrum. In one of the convolution methods, Eq. (1) is solved for an infinitesimal distance and the straggling function for all other distances determined by convoluting this function with itself many times. It may be assumed that given an exact $w(E)$, the convolution method gives the exact theoretical straggling function, limited only by the accuracy of the numerical integration. The reliability of the usual Laplace-transform solutions may be assessed by comparison with these results.

In this paper, therefore, as an example, we have determined a semiempirical realistic single-collision spectrum for solid aluminum, determined the straggling function by direct convolution, and compared it with various Laplace-transform solutions corresponding to different values of absorber thickness x. We pay particular attention to small values of x where the differences between the cross sections used in the transform theories and a realistic $w(E)$ produce the greatest effect. We may note that the cross sections used here describe energy losses only to the electrons of the absorber, since we will treat incident charged particles so fast that collisions with nuclei occur with negligible cross section.

In Sec. II we describe the different collision spectra which have been used in the transform theories as well as a realistic spectrum for Al. Next we briefly describe the Landau theory and its modifications. In Sec. IV the procedure for direct convolution and the results are presented, which

are compared with transform-method results in Sec. V.

In closing this introduction, it may be noted that the energy lost by the fast charged particle is only approximately related to the energy deposited in the absorber. In particular, energy may escape in the form of photons emitted in the deexcitation of inner-shell vacancies or of δ rays.[13,14] Therefore, although convolution methods produce what we have termed the correct theoretical straggling function, namely, the proper solution to Eq. (1) giving the energy loss of the incident particles, great care must be taken in comparing such results to experiment, particularly for the case of pulse-height spectra in proportional counters where, typically, the energy deposited in the material is observed. Further discussion of this important point is beyond the scope of this paper.

II. COLLISION SPECTRA

The differential collision cross section $w(E)dE$ describes the probability of the loss of an amount of energy between E and $E+dE$ in a single collision. It depends, of course, on the properties of the target material although the simple spectra assumed by Landau and Vavilov apply generally.

It will be useful to consider the moments M_n of the spectra:

$$M_n = \int_0^\infty E^n w(E)\, dE . \tag{2}$$

The total collision cross section σ_t defined previously is equal to M_0. The stopping power is equal to M_1, the "width" of a straggling distribution is related to M_2, and its asymmetry to M_3. The second moment is used in the modifications to the Landau theory, to be discussed below.

A. Free-electron spectra—Landau and Vavilov theories

Vavilov, assuming all the electrons in the absorber to be free and at rest, uses the Rutherford macroscopic cross section,

$$w'(E) = \frac{2\pi z^2 e^4}{m v^2} N_A \frac{Z}{A} \frac{(1-\beta^2 E/E_M)}{E^2}, \quad 0 \le E \le E_M$$

$$w'(E) = 0, \quad E > E_M \tag{3}$$

where m and e are the electron mass and charge, respectively, $v=\beta c$ and z are the velocity and charge of the incident particle, Z and A are the atomic number and weight of the target in grams (in the case of a molecular target, the sum over constituent atoms is used), and N_A is Avogadro's number. The maximum energy transferable in a single collision is $E_M = 2mc^2\beta^2/(1-\beta^2)$. The definition

$$k = \frac{2\pi z^2 e^4}{mc^2\beta^2} N_A \frac{Z}{A} = \frac{0.15354}{\beta^2} z^2 \frac{Z}{A} \ (\text{MeV cm}^2)$$

will be useful. Landau neglects the term $\beta^2 E/E_M$ but uses the same form otherwise. Quantities which refer to the cross section $w'(E)$ will be denoted by primes. Because of the divergence at $E=0$, the moments M_0' and M_1' are singular. The second moment is written $M_2' = kE_M$ in the nonrelativistic limit.

B. Free-electron spectrum with low-energy cutoff

Since there can be no energy losses for $E < E_m$, the energy of the lowest excitation of the target, it would be more realistic to apply the limits $E_m \le E \le E_M$ to the cross section in Eq. (3). The moments M_0 and M_1 can then be evaluated. Problems which occur in the application of these limits to the Vavilov theory are discussed in Appendix A.

C. Collision spectrum for aluminum

A realistic collision spectrum for the passage of 20-MeV protons through solid aluminum has been determined semiempirically. Aluminum is a propitious example because the dielectric theory used to describe its well-known plasmon loss may also be used for the characteristic energy losses in organic and biological[15,16] solids. The spectrum for Al is discussed separately for each shell.

K shell. Energy losses to K-shell ionizations have been calculated within the Born approximation as

$$w_K(E) = \frac{4\pi z^2 e^4 N_A}{m v^2 A} \int_{E^2/E_M}^\infty |F_K(Q)|^2 \frac{dQ}{Q^2}, \quad I_K \le E \le E_M \tag{4}$$

for I_K, the ionization potential of the K shell, and Q, the square of the momentum transfer. The inelastic-scattering form factors for the K shell, $|F_K(Q)|^2$, have been calculated by Walske[17] in a hydrogenic model. Discrete excitations out of the K shell can be neglected.[18]

L shell. An expression analogous to Eq. (4) for energy losses to L-shell ionizations has been used. For energies up to 1822.46 eV the calculation of inelastic form factors of Manson[19] using Hartree-Slater wave functions is used. Ionization from the $2s$ and $2p$ subshells was computed separately. Above that energy a hydrogenic calculation of the form factors by Walske[20] in which subshells were not distinguished was employed. The resulting cross sections were adjusted slightly to join smoothly. The cross section computed from the Hartree-Slater model is well approximated by the hydrogenic model at intermediate and high-energy losses.[19] Again, transitions to discrete states

have been neglected.[18]

M shell—plasmon excitation. According to the dielectric theory, the doubly differential cross section for the collective excitation of valence electrons in a solid, i.e., plasmon excitation, for the case of an incident proton, is given as[21]

$$\frac{d^2\sigma}{dE\,d\Omega}=\frac{1}{2\pi^2 n a_0}\frac{1}{\frac{1}{2}mv^2}\frac{1}{\theta^2+\theta_E^2}[-\mathrm{Im}(\epsilon(\omega))^{-1}], \quad (5)$$

where n is the density of electrons participating in the collective phenomena, Ω the solid angle into which the proton is deflected, a_0 the Bohr radius, and $\theta_E=E/pv$, with p being the momentum of the incident proton.[22] The frequency-dependent dielectric function is denoted by $\epsilon(\omega)$, where the energy loss $E=\hbar\omega$. The desired energy-loss cross section for the plasmon, $w_p(E)$, is given by an integration over angle using the small-angle approximation,

$$w_p(E)=\frac{1}{\pi n a_0}\frac{1}{\frac{1}{2}mv^2}\int_0^{\theta_c}\frac{\theta\,d\theta}{\theta^2+\theta_E^2}[-\mathrm{Im}(\epsilon(\omega))^{-1}], \quad (6)$$

where θ_c is the cutoff angle corresponding to momentum transfer k_c. For momentum transfers larger than k_c, the system no longer behaves collectively.[21]

Using the model of the free-electron gas, the dielectric function may be expressed in the Drude form, allowing for dispersion in the plasmon frequency,[23] as

$$-\mathrm{Im}(\epsilon(\omega))^{-1}=\frac{\omega\gamma\omega_p^2}{[\omega^2-\omega_p(\theta)^2]^2+\gamma^2\omega^2}, \quad (7)$$

where ω_p is the plasmon frequency; $\omega_p=(4\pi ne^2/m)^{1/2}$ and $\omega_p(\theta)$, to be taken from experiment, takes into account the dispersion in that frequency. The plasmon frequency $\hbar\omega_p=14.8$ eV, and width $\hbar\gamma=9$ eV were taken from the experiment of Swanson and Powell,[24] who used 20-keV incident electrons. The observed plasmon frequency corresponds to a valence-electron density of 2.64 valence electrons per atom. The experimental values $\theta_c=20$ mrad and

$$\omega_p(\theta)=\omega_p+mv^2(0.35\theta^2+260\theta^4)$$

are also taken from these authors' work.[15,25]

For large energy loss the cross section is expected[26] to become the classical cross section for energy transfer to a free electron, namely, $w_p(E)\propto E^{-2}$. The Drude formula, however, asymptotically is proportional to E^{-3}. Therefore, $w_p(E)$ was taken as

$$w_p(E)=\frac{2\pi z^2e^4N_A}{mv^2A}\left(\frac{3}{E^2}\right), \quad E>22.34\ \text{eV} \quad (8)$$

for energies greater than 22.34 eV, the point where Eqs. (6) and (8) match. All three valence electrons are assumed to participate in the free-electron-like collisions. This procedure is also desirable since $\omega_p(\theta)$ is certainly invalid for energies far from the plasmon peak.

From the cross section as the sum of the K-, L-, and M-shell contributions just described, the stopping power was computed to be within 3% of the experimental value for 20-MeV protons,[27] which gives some confidence in the appropriateness of the cross section.

III. LAPLACE-TRANSFORM METHODS

Using the theory of Laplace transforms, Landau[1] gave the solution of the transport equation as

$$f(x,\Delta)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}e^I\,dp, \quad (9)$$

where

$$I=p\Delta-x\int_0^\infty w'(E)(1-e^{-pE})\,dE \quad (10)$$

and $w'(E)$ is the cross section defined in Eq. (3).

The integral over E in Eq. (10), which may be performed analytically with some approximations,[28] is actually extended to infinity ignoring the constraint $w'(E)=0$, $E>E_M$, in contrast to the Vavilov case below. It is useful to define a reduced parameter[29]

$$\lambda=(\Delta-\bar{\Delta})/\xi+\lambda_1, \quad (11)$$

where $\xi=kx$, k having been defined in Sec. II,

$$\lambda_1=-(1-\gamma)-\beta^2-\ln\kappa$$

for γ the Euler constant 0.577 215 and $\kappa=\xi/E_M$. The mean energy loss $\bar{\Delta}$ for a cross section $w(E)$ is defined as

$$\bar{\Delta}=x\int w(E)E\,dE=xM_1,$$

where M_1 is the moment given in Eq. (2). Note that ξ and κ are also proportional to absorber thickness x. The desired distribution may then be written $f_L(x,\Delta)=(1/\xi)\phi(\lambda)$, where $\phi(\lambda)$ is a universal function (see Appendix A). The work of Landau just described and that of Vavilov and Blunck and Leisegang, to be described below, is concisely reviewed by Fano.[30]

The Vavilov[6] solution substitutes for Eq. (10) the equivalent expression

$$I'=p(\Delta-\bar{\Delta})-x\int_0^{E_M}w'(E)(1-e^{-pE}-pE)\,dE, \quad (12)$$

where the cross section defined in Eq. (3) is used

in the integral in Eq. (12). Theory of stopping power was used to calculate $\overline{\Delta}$. Again, the integral over E in Eq. (12) is analytical. In this case, however, the kinematic constraint is imposed explicitly resulting in a more complicated expression, detailed in Appendix A.

The distribution $f(x, \Delta)$ is not given by a universal function. It depends on x, or equivalently, on the parameter κ, which is the customary variable. In the limit $\kappa \to 0$, the Vavilov distribution goes over into the Landau distribution.

The Shulek *et al.*[7] modification of the Vavilov approach provides a means of correcting for the difference between a realistic cross section $w(E)$, which treats the electrons as bound in atoms, and that used by Vavilov, $w'(E)$. We may write an improved I as $I = I' + \delta I$, where

$$\delta I = -x \int_0^{E_M} [w(E) - w'(E)](1 - e^{-pE} - pE)\, dE . \tag{13}$$

Expanding in a Taylor series, and, since the functions differ most for small E, keeping the first nonvanishing term in $(1 - e^{-pE} - pE)$,

$$\delta I = -x \int_0^{E_M} [w(E) - w'(E)](\tfrac{1}{2} p^2 E^2)\, dE$$

$$= \tfrac{1}{2} x p^2 \delta_2 ,$$

where $\delta_2 = M_2 - M_2'$. Note that one need only know the second moment of the realistic distribution. The numerical integral over p to determine $f(x, \Delta)$ requires no more effort than for the unmodified Vavilov function (see Appendix A). One of us (H.B.) has noted that adding further moments to this correction procedure does not appear to be useful.[8]

A conceptually identical modification has been applied by Blunck and Leisegang[5] to the Landau function. Taking advantage of a theorem on Laplace transforms of convolutions, we write an improved distribution as

$$f_{BL}(x, \Delta, \delta_2) = \frac{1}{(2\pi x \delta_2)^{1/2}} \int_{-\infty}^{\infty} f_L(\Delta - y)\, e^{-y^2/2x\delta_2}\, dy . \tag{14}$$

Based on the universal Landau function, this distribution is easier to calculate than the Shulek *et al.* modification to the Vavilov function and often agrees quite well with it for small κ where the unmodified Vavilov solution agrees with the Landau function.

Finally, the approach of Ref. 8 is to use the realistic cross section $w(E)$ directly in place of $w'(E)$ in Eq. (12). The integral over E then must be performed numerically. The Laplace-transform method gives an exact solution of the transport equation, just as the convolution method, to be described next, provides an exact solution to the same equation. When the same cross section is used in both methods, the same result must be obtained. In practice, however, for a realistic $w(E)$, the transform approach requires two integrals to be performed numerically, first over E in Eq. (12) and second over p in Eq. (9), the latter of which is extremely difficult numerically, particularly for small values of κ (see Appendix A). It is therefore not a practical way to obtain the straggling distribution for small κ, while the convolution method presents no problems.

The Landau function itself, and as modified by Blunck and Leisegang, and the Vavilov function with the Shulek *et al.* modification, will be compared with the convolution solution using the realistic Al cross section.

IV. CONVOLUTION METHOD

Since the individual collisions experienced by the incident charged particle may be assumed to be statistically independent, the straggling distribution $f(x, \Delta)$ which is the solution to Eq. (1) has the property

$$f(x_1 + x_2, \Delta) = \int_0^{\Delta} f(x_1, \Delta - F) f(x_2, F)\, dF . \tag{15}$$

Equation (15) may be understood as follows: The distribution of particles which lost an energy F in absorber thickness x_2 convoluted with the distribution which lost energy $\Delta - F$ in x_1 gives the distribution which lost energy Δ in $x_1 + x_2$. It may be demonstrated mathematically using the equivalent formulation for $f(x, \Delta)$ given in Eq. (19) below and taking the Laplace transform of Eq. (15).

For a differential distance dx, we may write

$$f(x + dx, \Delta) = f(x, \Delta) + df(x, \Delta), \tag{16}$$

where $df(x, \Delta)$ is found by multiplying Eq. (1) by dx. Applying Eq. (16) at $x = 0$ with the initial condition $f(0, \Delta) = \delta(\Delta)$ the expression

$$f(dx, \Delta) = \delta(\Delta)(1 - \sigma_t\, dx) + dx\, w(\Delta) \tag{17}$$

is obtained. Equation (17) has a clear physical interpretation: the δ function at $\Delta = 0$ has been diminished by the probability of experiencing a collision $\sigma_t\, dx$, which is distributed among the energy losses according to the cross section for a single collision, $w(\Delta)$. It may be noted that the same initial condition was assumed by Landau in the Laplace-transform solution.

Using this expression and repeatedly applying Eq. (15), the distribution $f(x, \Delta)$ for any desired x may be determined.[11] In practice, the convolution in Eq. (15) is used for equal segments, dou-

bling the distance each time. The distance x is related to the mean number of collisions experienced by the incident particle, m_c, by

$$m_c = x\sigma_t, \tag{18}$$

or equivalently, $m_c\langle E\rangle/M_1 = x$, where M_1, as noted previously is the stopping power. The average energy $\langle E\rangle$ lost in a single collision is given by $\langle E\rangle \equiv M_1/\sigma_t$. The mean energy loss may be written $\overline{\Delta} = m_c\langle E\rangle$. Physically, then, an appropriate dx corresponds to a value of m_c small enough that the probability of sustaining more than one collision is negligible.

It may be noted parenthetically that the prescription just described for determining $f(x, \Delta)$ is equivalent to the equation[10,11,31]

$$f(x, \Delta) = \sum_{n=0}^{\infty} \frac{m_c^n e^{-m_c}}{n!} w(\Delta)^{*n}, \tag{19}$$

where $w(\Delta)$ is the cross section for a single collision which we have been using and $w(\Delta)^{*n}$ denotes the n-fold convolution of $w(\Delta)$, i.e.,

$$w(\Delta)^{*0} = \delta(\Delta),$$

$$w(\Delta)^{*1} = w(\Delta),$$

$$w(\Delta)^{*n} = \int_0^{\Delta} w(E)\, w^{*(n-1)}(\Delta - E)\, dE.$$

Equation (19) has a clear physical interpretation. Each term of the sum in Eq. (19) is the probability of undergoing n collisions, as given by a Poisson distribution, multiplied by the distribution of energy losses in n collisions.

Calculations were performed according to the convolution method using a computer program modified from one kindly provided by A. Kellerer. A value of the differential dx corresponding to $m_c = 1/1024$ was used and the realistic cross section for 20-MeV protons on solid aluminum described in Sec. II was employed. Since we are interested in energy losses over a very broad range, for the integrals in Eq. (15), grid points were equally spaced in $\ln E$.

Results will be discussed in terms of mean collision number, related to absorber thickness by Eq. (18). The mass stopping power for 20-MeV protons on Al is 19.69 MeV cm^2/g; in consistent units x, absorber thickness, is expressed in g/cm^2. The average energy $\langle E\rangle$ is 77.03 eV for the spectrum used here. Since, as mentioned previously, the stopping power determined from the theoretical cross section differed by 3% from the experimental value,[27] the cross section $w(E)$ was multiplied by the ratio of the experimental to theoretical stopping powers.

All of the following observations on the convolution results will be found to agree with expectation. For small mean collision numbers the major contribution to the straggling distribution is due to the plasmon peak in the cross section. If one studies the straggling function as m_c increases through small values, the probability of zero energy loss is seen to decrease while probability builds up at values corresponding to multiples of the plasmon frequency. This is demonstrated in Fig. 1 for $m_c = 2$, 4, 8, where peaks at multiples of $\hbar\omega_p = 14.8$ eV dominate the distribution. A peak due to the onset of L-shell ionization at 80.9 eV cannot be distinguished from that at $6\hbar\omega_p = 88.8$ eV.

For the limiting case of a cross section $w(E)$ represented by a δ function located at the plasmon energy, one would expect a straggling function with peaks only at multiples of the plasmon frequency, the areas of which are given by a Poisson distribution. This is demonstrated in Appendix B for both the convolution and transform methods. For $m_c = 4$ (Fig. 1) the areas of the first five peaks conformed within 10% to a Poisson distribution with a mean collision number of approximately 3.25. A number smaller than four is anticipated since some of the collisions have involved inner-

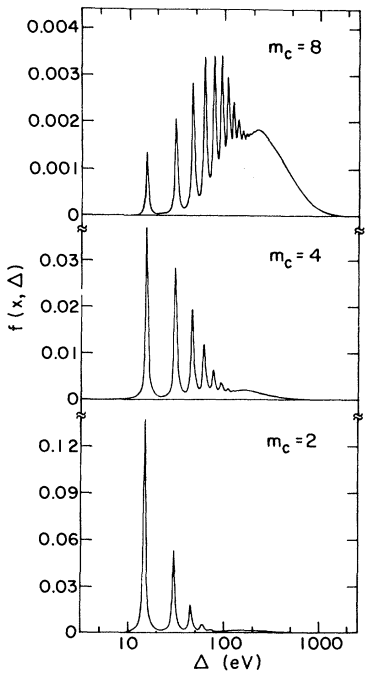

FIG. 1. Straggling distribution $f(x, \Delta)$ for 20-MeV protons incident on Al from the convolution calculation is plotted vs energy loss Δ on a logarithmic scale for $m_c = 2$, 4, and 8. For normal density Al, 2.7 g/cm^3, these cases correspond to x expressed in distance of 0.029, 0.058, and 0.116 μ, respectively. The probability of zero energy loss for $m_c = 2$ is 0.1352. For $m_c = 4$ and 8, zero loss probabilities are 0.0183 and 0.0003, respectively.

shell electrons instead of valence electrons.

As one goes from $m_c = 2$ to $m_c = 8$ in Fig. 1, a broad peak at higher energy losses also may be observed to grow and move to higher energies, which is expected since the mean energy loss is directly proportional to mean collision number. The magnitudes of the peaks due to the plasmon loss decrease as the straggling distribution gets broader since it is normalized.

For the case of $m_c = 16$, Fig. 2, the peaks at multiples of $\hbar\omega_p$ produce only slight structure on the over-all broad peak. For much higher values of mean collision number the straggling distribution consists simply of a broad peak, the location and exact shape of which depend on m_c. In other words, for these cases no detailed structure resulting from the plasmon peak at low energies may be discerned so it is reasonable to compare the convolution results with the Landau and Vavilov solutions; this will be done in Sec. V.

V. COMPARISON AND DISCUSSION

In Figs. 2–4, the straggling distribution $f(x, \Delta)$ is compared with the Landau function $f_L(x, \Delta)$, with the modified Landau function of Eq. (14), $f_{BL}(x, \Delta, \delta_2)$, and with the modified Vavilov function $f_V(x, \Delta, \delta_2)$. The functions have been plotted against the reduced variable λ defined in Eq. (11). Because the Al cross section was computed in a nonrelativistic model, the Vavilov function was evaluated with $\beta^2 = 0$ in the second term in Eq. (3).

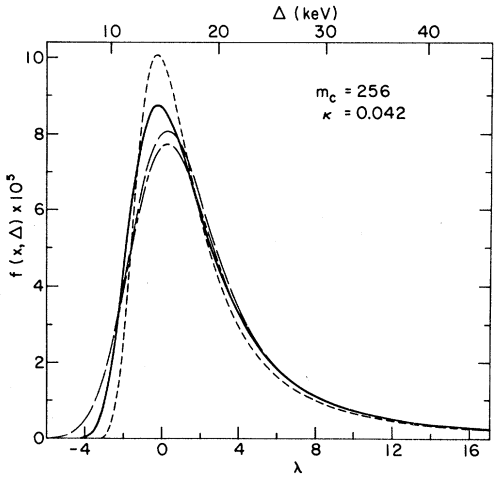

FIG. 3. Straggling distribution vs λ for intermediate absorber thickness: $m_c = 256$, $x = 3.71\ \mu$, $\kappa = 0.042$. $f(x, \Delta)$, solid line; $f_L(x, \Delta)$, short-dashed line; $f_V(x, \Delta, \delta_2)$, long-dashed line; and $f_{BL}(x, \Delta, \delta_2)$, long-and-short dashed line. The latter two are indistinguishable at low λ.

No comparison is provided with the Laplace-transform approach of Bichsel[8] which, in principle, would reproduce the convolution result although, in practice, as stated in Sec. III, numerical difficulties limit its application.

The comparison plotted in Fig. 2 is typical for what we have termed small values of absorber thickness x. In this case, which corresponds to a

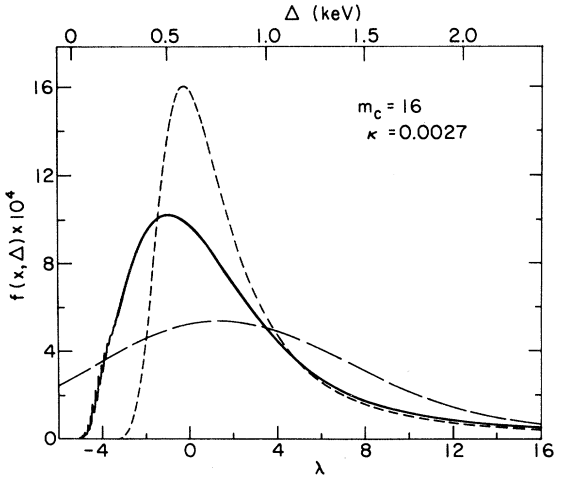

FIG. 2. Straggling distribution vs reduced-energy-loss parameter λ for $m_c = 16$, or equivalently, $x = 0.232\ \mu$, $\kappa = 0.002$: convolution result $f(x, \Delta)$, solid line; Landau function $f_L(x, \Delta)$, short-dashed line. The modified functions $f_V(x, \Delta, \delta_2)$ and $f_{BL}(x, \Delta, \delta_2)$ are indistinguishable on this scale and are represented by long-dashed line. The energy-loss scale in keV is also given.

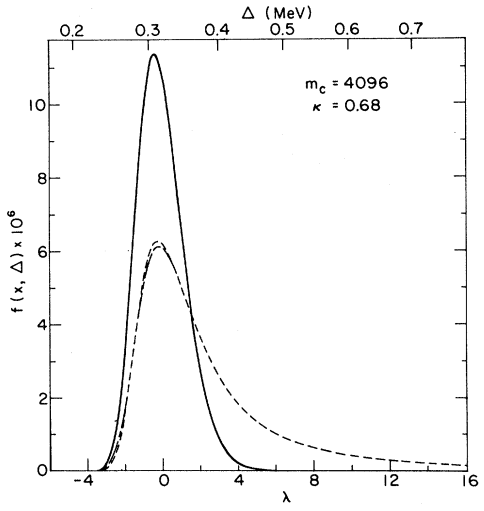

FIG. 4. Straggling distribution vs λ for larger thickness: $m_c = 4096$, $x = 59.4\ \mu$, $\kappa = 0.68$. In this plot, the modified Vavilov function $f_V(x, \Delta, \delta_2)$ is indistinguishable from the convolution result $f(x, \Delta)$, solid line; $f_L(x, \Delta)$, short-dashed line; $f_{BL}(x, \Delta, \delta_2)$, long-and-short dashed line.

mean collision number of 16 or to $\kappa = 0.0027$, $f_V(x, \Delta, \delta_2)$ and $f_{BL}(x, \Delta, \delta_2)$ cannot be distinguished from each other on the scale of the graph. This is expected since their respective unmodified functions agree[6] for $\kappa < 0.01$. The peak of the Landau function itself is narrower and higher than that of the convolution result, $f(x, \Delta)$. The introduction of the second-moment modification appears to overcompensate for this discrepancy, producing a function which is lower and broader than $f(x, \Delta)$.

In fact, it can be seen in Fig. 2 that $f_V(x, \Delta, \delta_2)$ has a significant probability for values of λ corresponding to negative energy loss. This corresponds to applying the solution of Eq. (1) for a nonphysical situation. For m_c smaller than the value plotted here, a similar difficulty occurs for the Landau function as well. In their derivations, the functions are assumed to have physical meaning only for positive energy loss. One possible interpretation is to attribute all probability described by, e.g., $f_L(x, \Delta)$ for $\Delta < 0$ to $\Delta = 0$. This interpretation is not needed in practice, however, since, when the situation occurs, neither $f_V(x, \Delta, \delta_2)$ nor $f_L(x, \Delta)$ provides a good description of the convolution result.

An intermediate case is plotted in Fig. 3 for which $m_c = 256$ and $\kappa = 0.042$. Here, $f_V(x, \Delta, \delta_2)$ and $f_{BL}(x, \Delta, \delta_2)$ agree to within 3% at their maxima and approximate the shape of $f(x, \Delta)$ better than in the previous case. Finally, for large x, $m_c = 4096$, and $\kappa = 0.68$ (Fig. 4), $f_V(x, \Delta, \delta_2)$ agrees very well with the convolution result. In this case, the modified and unmodified Landau functions are very similar to each other and are lower than $f(x, \Delta)$ and are shifted to higher energy losses. The modified and unmodified Vavilov functions (not shown) also agree to within a few percent. Note that we are still considering energy losses much smaller than the incident energy.

The observations on the Laplace-transform solutions may be understood as follows. The discrepancy between cross sections $w(E)$ and $w'(E)$ for which the second-moment modification compensates, occurs at small E. For the large energy losses, with which we are concerned in Fig. 4 ($\bar{\Delta} > 0.1$ MeV), the straggling distribution is not very sensitive to the details of the cross section at small E. Thus, the larger the absorber thickness or mean collision number, the smaller is the effect of the second-moment modification. One may also note that the high-energy tail on the Landau function in Fig. 4 may be attributed to the lack of kinematic constraint on $w'(E)$, as Landau employed the cross section.

In summary, it has been observed from the convolution results that for very small x, the straggling distribution is only sensitive to the cross section at low energies, i.e., for the case of solid Al studied here, only to the plasmon peak. For somewhat larger x, corresponding to $m_c = 16$, although structure in the straggling function at multiples of the plasmon frequency is not an important feature, the convolution result is not approximated well by the Vavilov function, even as modified. As one goes to even larger values of x or κ, the modified Vavilov function comes into better agreement with the result of the convolution procedure. In other words, for large x, the straggling distribution becomes less sensitive to the details of the single-collision cross section. For κ greater than about 0.2, $f_V(x, \Delta, \delta_2)$ is found to be in excellent agreement with the convolution result.

The situation in silicon, which is of interest due to its use in solid-state detectors, can be expected to be quite similar. This work may be viewed as a study of the consequences of calculating straggling functions using cross sections which depart from free-electron cross sections. The results may be applied quite generally independent of the particular physical processes which cause that departure. Whatever the origin of the characteristic energy losses in biological solids,[15,16] they do show a peak in the energy-loss cross section at low energies, similar to the Al example treated here. The details of the straggling distributions will depend of course on the details of the cross sections. We have not attempted in this paper to survey the effects of different features in the cross section on the agreement between the Vavilov theory and convolution results; such a study would be useful in the future.

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APPENDIX A

Landau evaluates the integral in Eq. (10), expressed in present notation, as

$$x \int_0^\infty w(E)(1 - e^{-pE})\,dE = \xi p[1 - \gamma - \ln p + \bar{\Delta}/\xi - \ln(\xi/\kappa) - \beta^2]. \tag{A1}$$

Substituting (A1) into Eq. (9) with the change of variables $u = \xi p$ gives

$$f_L(x, \Delta) = (1/\xi)\phi(\lambda),$$

$$\phi(\lambda)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} e^{u\ln u+\lambda u}\,du,$$

which must be evaluated numerically in the limit $c\to 0$ to give the Landau function.

The integral occurring in the Vavilov theory, Eq. (12), may be evaluated to give

$$I'=p(\Delta-\bar{\Delta})-p\xi(1+\beta^2)+\kappa(1-e^{-pE_M})+(\kappa\beta^2+\xi p)[\gamma+\ln(pE_M)-\mathrm{Ei}(-pE_M)], \tag{A2}$$

where Ei is the exponential integral function and the relation $x=\kappa E_M/k$ has been used. Substituting in Eq. (9) and performing the integral along the imaginary axis with the change of variables $y=pE_M$ gives

$$f'(x,\Delta)=\frac{1}{\pi E_M}e^{\kappa(1+\beta^2\gamma)}\int_0^\infty e^{\kappa f_1}\cos(y\lambda_1+\kappa f_2)\,dy,$$

where

$$f_1=\beta^2[\ln y-\mathrm{Ci}(y)]-\cos y-y\,\mathrm{Si}(y),$$
$$f_2=y[\ln y-\mathrm{Ci}(y)]+\sin y+\beta^2\,\mathrm{Si}(y), \tag{A3}$$
$$\lambda_1=\kappa\lambda+\kappa\ln\kappa.$$

The symbols Si and Ci stand for the integral sine and integral cosine functions, respectively.

The Shulek modification adds the term $-\frac{1}{2}xp^2\delta_2$ to the exponential in the integrand which may now be written, expressing x in terms of κ as,

$$f_V(x,\Delta,\delta_2)=\frac{1}{\pi E_M}e^{\kappa(1+\beta^2\gamma)}\int_0^\infty \exp\left[\kappa\left(f_1-\frac{\delta_2 y^2}{2kE_M}\right)\right]\times\cos(y\lambda_1+\kappa f_2)\,dy. \tag{A4}$$

For the numerical-integration approach of Bichsel, it is useful to express the integral in Eq. (12) in terms of the pure imaginary variable $q=ip$,

$$I_2(q)=\int_0^{E_M} w(E)(1-e^{-iqE}-iqE)\,dE. \tag{A5}$$

Dividing this integral into real and imaginary parts and using the symmetry of the integrals, we obtain

$$f(x,\Delta)=(1/\pi)\int_0^\infty \exp\{-(\kappa E_M/k)\,\mathrm{Re}[I_2(q)]\}\times\cos\{y(\Delta-\bar{\Delta})-(\kappa E_M/k)\,\mathrm{Im}[I_2(q)]\}\,dq. \tag{A6}$$

It may be shown that when the cross section used by Vavilov, $w'(E)$, is used in Eq. (A5), Eq. (A6) is the same as in the Vavilov case (A3).

Numerical difficulties in the evaluation of Eq. (A6) arose for $\kappa<0.003$ for the Al spectrum used here. In general, $\mathrm{Re}[I_2(q)]$ is an oscillatory function with the oscillations becoming damped as q increases; $\mathrm{Re}[I_2(0)]=0$ and $\mathrm{Re}[I_2(q)]>0$ for $q>0$. The imaginary part $\mathrm{Im}[I_2(q)]$ monotonically becomes more negative with increasing q and $\mathrm{Im}[I_2(0)]=0$. For sufficiently large x or κ which appears in the exponential, all the contribution to the integral comes near $q=0$ where $\mathrm{Re}[I_2(q)]$ is small. However, for small values of κ, the integral does not converge as one integrates over q. Also see Ref. 8.

It may be instructive to examine the situation when the free-electron cross section, Eq. (3), to which the low-energy cutoff has been applied is used in Eq. (A5). The same numerical problems again arise using the integration limits E_m to E_M in Eq. (A5); we may write

$$\mathrm{Re}[I_2'(q)]=k\left(\frac{\cos(qE_M)}{E_M}-\frac{\cos(qE_m)}{E_m}-\frac{1}{E_M}+\frac{1}{E_m}+q\,\mathrm{Si}(qE_M)-q\,\mathrm{Si}(qE_m)\right). \tag{A7}$$

For large q, using

$$\mathrm{Si}(q)\sim\tfrac{1}{2}\pi-(\cos q)/q+\cdots,$$

we find $\mathrm{Re}[I_2'(q)]\approx k/E_m$, which is a constant. Substituting in Eq. (A6) again, we find that for small values of x or κ the integral does not converge as one integrates over q.

APPENDIX B

The convolution method may be applied for a model cross section $w(E)=\sigma_t\delta(E-E_0)$. Using the initial condition stated in Sec. IV, $f(0,\Delta)=\delta(\Delta)$, Eq. (17) becomes

$$f(dx,\Delta)=\delta(\Delta)(1-\sigma_t\,dx)+dx\,\sigma_t\delta(E-E_0).$$

Applying the convolution procedure [Eq. (15)] p times, the expression

$$f(n\,dx,\Delta)=\sum_{m=0}^{n}\binom{n}{m}(\sigma_t\,dx)^m\times(1-\sigma_t\,dx)^{n-m}\delta(\Delta-mE_0), \tag{A8}$$

where $n=2p$ is obtained. In the limit $m\ll n$, $\sigma_t dx\ll 1$, the binomial distribution in Eq. (8) becomes a Poisson distribution and $f(n\,dx,\Delta)$ may be written

$$f(n\,dx,\Delta)=\sum_{m=0}^{n}\frac{\tau^m e^{-\tau}}{m!}\delta(\Delta-mE_0), \tag{A9}$$

where $\tau=n\sigma_t\,dx$ may be interpreted as the mean collision number. Note that σ_t is expressed in cm^2/g and dx in $\mathrm{g/cm}^2$. Equation (A8) says the

TABLE A I. Glossary. In general, the symbols from the earlier papers have been used here. Therefore, some symbols are used for different quantities. (Symbols occur in or near the equation given.)

Symbol	Equation or section	Meaning
E	(1)–(4), (8), (10), (12), (13), (A1), (A5), (A8)	Energy loss of incident particle in a single collision
$\langle E \rangle$	(18)	Average energy lost in a single collision (here, 77.03 eV)
E_m	II B	The smallest permitted value of E
E_M	(3), (4), (13), (A2), (A4)–(A7)	The largest possible value of E (42.2 keV)
E_0	Appendix B	Discrete energy loss if $W(E)$ is represented by a δ function
$f(x, \Delta)$	(1), (9), (15)–(17), (19), (A3), (A6), Appendix B	Straggling function in general
$f'(x, \Delta)$	(A3)	Vavilov's straggling function
$f_L(x, \Delta)$	(11), (14), V, (A1)	Landau's straggling function
$f_{BL}(x, \Delta, \delta_2)$	(14), V	Blunck-Leisegang modification of Landau's straggling function
$f_V(x, \Delta, \delta_2)$	V, (A4)	Modified Vavilov function
$\lvert F_K(Q)\rvert^2$	(4)	Inelastic scattering form factor for K shell (Walske)
f_1, f_2	(A3), (A4)	Function occurring in Vavilov integral for $f(x, \Delta)$
F	(15)	Total energy loss of a particle
I	(9), (10)	Integral used in Landau theory
I'	(12), (A2)	Integral used in Vavilov theory
δI	(13)	Integral used to obtain modified Vavilov function
k	II, (11), Appendix A	Factor in $w(E)$
k_c	(6)	Maximum momentum transfer permissible in plasmon excitation
M_n	(2), (11), (13), (18)	The nth moment of a probability density function
M_1		Stopping power (here assumed to be 19.69 MeV cm^2/g)
M_2'	(3), (13)	Second moment of $w'(E)$, Eq. (3)
m_c	(18), (19), IV, Figs. 1–4	Mean number of collisions experienced by particle
$w(E)$	(1), (2), (11), (13), (A1), (A5), Appendix B	Differential collision cross section for a single collision
$w'(E)$	(3), (10), (12), (13), V	Special form (Rutherford) of $w(E)$
$w_K(E)$	(4)	Contribution to $w(E)$ due to collisions with K shell of atom
$w_p(E)$	(6), (8)	Contribution to $w(E)$ due to energy losses to plasmon excitations
$w(\Delta)$	(17), (19)	Same as $w(E)$ but the total energy loss Δ is achieved in a single collision
$w(\Delta)^{*n}$	(19)	The n-fold convolution of $w(\Delta)$
x	(1), (10), (11), (12)–(14), (16), (18), V, (A1), Appendix B	Absorber thickness (g/cm^2)
x_1, x_2	(15)	Partial absorber thickness, $x_1 + x_2 = x$
Δ	(1), (10)–(12), (A2), (A6)	Total energy loss of a particle in x
$\bar{\Delta}$	(11), (12), (18), V, (A2), (A6)	Mean value of Δ
δ_2	(13), (14), (A4)	$M_2 - M_2' = 0.074\, M_2'$
$\epsilon(\omega)$	(5)–(7)	Frequency-dependent dielectric function of absorber
θ	(5), (6)	Angle of deflection of incident particle
θ_E	(5), (6)	E/pv
θ_c	(6)	Cutoff angle corresponding to k_c
κ	(11), (14), V, (A3), (A4), (A6)	ξ/E_M
λ	(11), (A1), (A3), V	Landau parameter
λ_1	(A3)	Vavilov parameter
$\bar{\lambda}$	(11)	Defined below Eq. (11)

TABLE AI. (*Continued*)

Symbol	Equation or section	Meaning
ξ	(11), (A2)	kx
σ_t	(1), (17), (18), (A8), Appendix B	Macroscopic total collision cross section (here 2.56×10^5 cm^2/g, corresponding to a mean free path of 1055 Å)
$\frac{d^2\sigma}{dE\,d\Omega}$	(5)	Doubly differential cross section for the collective excitation of valence electrons in a solid
τ	Appendix B	Mean collision number
$\phi(\lambda)$	(11), (A1)	Universal Landau function
ω	(5), (7)	Frequency of collective excitation, energy loss $=\hbar\omega$
ω_p	(7)	Plasmon frequency
Ω	(5)	Solid angle for deflection of incident charged particle

straggling distribution is nonzero only for energy losses which are multiples of the single-collision loss E_0 and the areas of these peaks are given by a Poisson distribution. Equation (A9), as expected, is identical with Eq. (19).

The same result is obtained from the Laplace-transform method. With the δ-function model cross section, Eq. (10) becomes

$$I = p\Delta - x\sigma_t(1 - e^{-pE_0}).$$

Substituting in Eq. (9) with the change of variables $p = iy$, the expression

$$f(x, \Delta) = \frac{e^{-\sigma_t x}}{2\pi} \int_{-\infty}^{\infty} e^{iy\Delta} \exp(\sigma_t x e^{-iyE_0})\, dy$$

is obtained. Expanding the second exponential in the integrand leads to

$$f(x, \Delta) = \sum_{m=0}^{\infty} \frac{(\sigma_t x)^m e^{-\sigma_t x}}{m!} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iy(\Delta - mE_0)} dy$$

$$= \sum_{m=0}^{\infty} \frac{\tau^m e^{-\tau}}{m!} \delta(\Delta - mE_0),$$

where $x = n\,dx$. The transform result is the same as Eq. (A9). The extension of the sum to infinity may be ignored since the Poisson distribution becomes vanishingly small for large m.

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