# Slowing down of medium-energy electrons in solids

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The slowing down of energetic particles in solids has been studied for the special but important case that the energy loss process is independent of any other interaction characteristic. For a realistic variation of the scattering characteristics with energy this allows one to go over from the continuous energy variable to a discrete variable, being equal to the number of inelastic processes. Then, the energy dissipation can be described in terms of partial intensities of particles having experienced a given number of inelastic collisions and the partial energy distributions associated with them. The important advantage of this so-called partial intensity approach is that it can accurately account for the variation of the scattering characteristics with the energy in a simple way. The key point in this procedure is the distribution of the stochastic process governing multiple collisions in the multispeed case. The exact solution as well as an accurate and efficient analytical approximation is given for the probability for $n$-fold scattering as a function of the traveled path length. Application of the proposed method to inelastic backscattering of electrons from solids leads to excellent agreement with experimental results and simulation data and constitutes a significant improvement over methods that neglect the energy dependence of the interaction characteristics. The method should prove useful for slowing down problems of different types of particles like electrons, ions, etc., in solids. [S0163-1829(97)04321-X]

## I. INTRODUCTION

The transport of electrons, ions, neutrons, and other particles is of importance for various fields in physics. The phenomenon may be dealt with in the general framework of linear transport theory,[1–5] but unfortunately this does not generally provide an exact solution in a mathematically tractable form. Therefore it is very often necessary to either employ numerical solution approaches or invoke an appropriate approximation. Clearly, the meaningfulness of any approximation is strictly limited by the interaction characteristics and the boundary conditions of any particular case.

Several decades ago, Landau[6] studied the energy loss of fast particles in the case that the total energy loss is small compared to the initial energy. In this quasielastic energy regime, the constant cross section approximation applies. Landau's formalism is based on the Poisson stochastic process that governs multiple energy losses in the constant cross section (quasielastic) approximation. In many subsequent works, Landau's results have been applied to study the interaction of energetic ions, electrons, and other particles with solids. This author's approach is particularly useful in the quasielastic energy range below a peak in an electron spectrum, where the constant cross section approximation holds, since the path lengths of the electrons that contribute to the yield in this region are not much greater than the inelastic mean free path. The theory of noncoherent scattering in electron spectroscopy is therefore almost exclusively based on Landau's result.[7–12]

In the constant cross section approximation, for which the Landau theory was developed, an alternative and completely equivalent approach exists that describes the energy distribution as a superposition of groups of $n$-fold inelastically scattered particles. The energy distribution after $n$ losses is given by an $n$-fold self-convolution of the differential mean free path for inelastic scattering and therefore this method is often referred to as the convolution method.[13,10] If particle deflection can be neglected, i.e., if the rectilinear motion model represents a good approximation, this method becomes particularly simple since the relative contribution of $n$-fold scattered particles is then governed by the Poisson stochastic process.[13,14]

For many problems of particle transport both the constant cross section approximation and the straight line approximation are inadequate and alternative approaches have to be employed to properly account for the particle deflection and energy dissipation process. Finding an appropriate approach to deal with particle deflection is greatly facilitated by invoking the so-called generalized radiative field similarity principle,[15–17] which states that the deflection function in the transport equation may be replaced by, e.g., an isotropic transport cross section if the source angular distribution is sufficiently smooth. Thus for a (nearly) isotropic source angular distribution, the deflection process can be treated analytically in the transport approximation,[10,18] while for, e.g., a well-collimated beam of particles incident on a surface one is forced to resort to numerical procedures exactly accounting for the details of the deflection function.

Properly accounting for the energy dissipation beyond the quasielastic energy regime is not straightforward. If the energy dissipation takes place concurrently with rapid momentum relaxation, i.e., if the particle suffers intense deflections, the energy distribution is not determined by energy fluctuations, but mainly by the distribution of the traveled path lengths. Then one can use the continuous slowing down approximation.[6,19,20,5] Such an approach has been successfully employed to derive accurate yet simple solutions for some integral quantities like the backscattering coefficient[19,21] and has been widely used in numerical model calculations.[22,23]

 

A more accurate description is provided by multigroup transport theory (see, e.g., Ref. 2). Here the particle flux density is divided into groups of particles with a speed in a given interval and for each interval the scattering characteristics are assumed to be constant. This procedure requires extensive numerical calculations of high complexity, in particular if the details of the deflection process play a role in the considered problem, when the source angular distribution is highly anisotropic.

In the present paper, an approach is outlined that is free of the drawbacks of the constant cross section approximation while it is much simpler than the usual multigroup approaches and for certain cases it should be more convenient to use than the continuous slowing down approximation. In fact, it is only slightly more involved than the convolution method while it is significantly more accurate. In this approach, which will be referred to as the partial intensity approach, the flux density is divided into groups of particles that have suffered a certain number, say, $n$, inelastic collisions, just as in the convolution method. The advantage of the partial intensity method is that it can accurately account for the variation of the scattering characteristics with energy if the following two conditions are met: (1) The energy loss process is independent of all other phenomena relevant for the particle transfer, in particular the deflection process, and (2) the energy dependence of the scattering characteristics within a group can be neglected. The first condition implies that the energy of the flux density of the $n$th group is uncorrelated with the direction of the particle, while the second assumption allows us to go over from the continuous energy variable to the discrete variable, being equal to the number of inelastic processes a particle has suffered.

The structure of this paper is as follows: The first theoretical section, Sec. II A, shows how the two assumptions mentioned above lead to the formal solution of the kinetic equation for an arbitrary boundary problem in terms of partial intensities and the energy distributions associated with them. In the second theory section, Sec. II B, the stochastic process is studied for the case that the interaction characteristics vary with the number of inelastic processes. This is the key to generalizing the convolution method to energies beyond the quasielastic regime where the constant cross section approximation no longer holds. Ultimately, knowledge of the stochastic process gives rise to a very efficient procedure to calculate the partial intensities that is employed later on. The proposed approach is then tested by comparison of theory with experimental data on electron backscattering from solids and it is shown that the method generally performs better than the convolution method in the constant cross section approximation, while it is only slightly more involved.

## II. THEORY

### A. Derivation of the basic relationships

The starting point of our considerations is a Boltzmann-type kinetic equation for the flux density $N(z,\vec{\Omega},E)$ of particles traveling at the depth $z$ in the direction $\vec{\Omega}$ with an energy $E$ due to sources at $z=z_0$ emitting in the direction $\vec{\Omega}=\vec{\Omega}_0$. For a case with plane symmetry, the transfer equation for the flux density reads[2]

$$\mu\frac{\partial N}{\partial z}=-\frac{N}{\lambda_t}+\frac{1}{4\pi\lambda_e}\int_{4\pi}I(\vec{\Omega},\vec{\Omega}',E)N(z,\vec{\Omega}',E)d\vec{\Omega}'+\int_0^\infty\frac{w(T,E+T)}{\lambda_i(E+T)}N(z,E+T,\vec{\Omega})dT+f_0(E)\delta(z-z_0). \tag{1}$$

Here $\mu=\cos\psi$ is the polar direction of the particle's motion relative to the outward surface normal, $\lambda_i$ and $\lambda_e$ are the inelastic and elastic mean free paths, and $\lambda_t=\lambda_e\lambda_i/(\lambda_e+\lambda_i)^{-1}$ is the total mean free path. $I(\vec{\Omega},\vec{\Omega}',E)$ is the scattering function, being equal to the normalized cross section multiplied with $4\pi$, and $w(T,E)$ is the normalized differential inverse inelastic mean free path (DIIMFP), i.e., the probability that a particle loses energy $T$ in an individual inelastic event. Finally, $f_0(E)$ is the normalized energy distribution at the source. Note that by virtue of the linearity of the kinetic equation, the solution of Eq. (1) allows us to find the solution for an arbitrary source function by superposition. The kinetic equation needs to be complemented with a boundary condition for a given problem. It is omitted here since the results in this section are valid for any physically meaningful boundary condition. It should also be noted that deflections associated with the momentum transfer in an inelastic event have been neglected in Eq. (1). This approximation is admissible when the transport mean free path for deflections in an inelastic collision significantly exceeds the corresponding mean free path during an elastic event. For medium-energy electrons this requirement is generally met.[24,25] If inner shell processes associated with large momentum transfers are significant, angular deflections in inelastic collisions must be taken into account.[26]

We divide the flux density into groups of $n$-fold inelastically scattered particles:

$$N(z,\vec{\Omega},E)=\sum_{n=0}^{\infty}N_n(z,\vec{\Omega},E). \tag{2}$$

In practical calculations, the upper limit in the summation is usually replaced by $n=n_{\max}$, where $n=n_{\max}$ corresponds to the number of collisions a particle experiences before its energy drops below a certain cutoff value. Inserting the expansion, Eq. (2), into the kinetic equation yields a system of coupled equations:

$$\mu\frac{\partial N_0}{\partial z}=-\frac{N_0}{\lambda_t}+\frac{1}{4\pi\lambda_e}\int_{4\pi}I(\vec{\Omega},\vec{\Omega}',E)N_0(z,\vec{\Omega}',E)d\vec{\Omega}'+f_0(E)\delta(z-z_0)\delta(\vec{\Omega}-\vec{\Omega}_0),$$

$$\mu\frac{\partial N_n}{\partial z}=-\frac{N_n}{\lambda_t}+\frac{1}{4\pi\lambda_e}\int_{4\pi}I(\vec{\Omega},\vec{\Omega}',E)N_n(z,\vec{\Omega}',E)d\vec{\Omega}'+\int_0^\infty\frac{w(T,E+T)}{\lambda_i(E+T)}N_{n-1}(z,E+T,\vec{\Omega})dT. \tag{3}$$

At this stage we make the two assumptions mentioned in the Introduction. First it is assumed that the energy loss process is independent of the other interaction processes. Since an inelastic collision is always accompanied by momentum

transfer, this is never strictly true. However, the characteristic length for deflection during an inelastic event in many cases significantly exceeds the corresponding quantity for elastic scattering. For example, for medium-energy electrons, the inelastic transport mean free path is at least one order of magnitude larger than the elastic one.[25] On the other hand, the energy transfer in an elastic collision is negligibly small in many cases, in particular if the mass of the incident particle is small compared to the mass of its collision partner. For electrons interacting with a solid the latter condition is therefore generally fulfilled. Under these circumstances it is reasonable to assume that the energy and angular variable of the $n$th group are uncorrelated. Note that this by no means implies that the energy and the angular variable of the flux density are uncorrelated. On the contrary, it is well known[10,27] that this is generally not the case. However, for the $n$th group, the energy should be uncorrelated with the other variables under these conditions. Therefore, we may write

$$N_n(z,\vec{\Omega},E)=F_n(E)P_n(z,\vec{\Omega}). \tag{4}$$

The second assumption concerns the energy dependence of the scattering characteristics. It will be assumed that the energy fluctuations within one group can be neglected:

$$\lambda(E)=\lambda(\overline{E_n})\equiv\lambda_n, \tag{5}$$

where $\lambda$ stands for any of the scattering characteristics (cross section or mean free path). The above approximations allow us to separate the energy variable in the kinetic equation (3). The separation constant follows from the normalization of the inverse inelastic mean free path. This immediately leads to the following equation for the partial energy distributions:

$$F_0(E)=f_0(E),$$

$$F_n(E)=\int_0^\infty F_{n-1}(E+T)w_{n-1}(T)dT. \tag{6}$$

Multiplying Eq. (6) by the energy and integrating gives the self-consistent solution for $\overline{E_n}$:

$$\overline{E_n}=E_0-\sum_{l=1}^{n}\langle T\rangle_l, \tag{7}$$

where

$$\langle T\rangle_l=\int_0^\infty w_{l-1}(T')T'dT' \tag{8}$$

is the mean loss in the $l$th collision. The equations for the partial escape distributions $P_n(z,\vec{\Omega})$ read

$$\mu\frac{\partial P_0}{\partial z}=-\frac{P_0}{\lambda_{t,0}}+\frac{1}{4\pi\lambda_{e,0}}\int_{4\pi}I_0(\vec{\Omega},\vec{\Omega}')P_0(z,\vec{\Omega}')d\vec{\Omega}'+\delta(z-z_0)\delta(\vec{\Omega}-\vec{\Omega}_0), \tag{9}$$

$$\mu\frac{\partial P_n}{\partial z}=-\frac{P_n}{\lambda_{t,n}}+\frac{P_{n-1}}{\lambda_{i,n-1}}+\frac{1}{4\pi\lambda_{e,n}}\int_{4\pi}I_n(\vec{\Omega},\vec{\Omega}')P_n(z,\vec{\Omega}')d\vec{\Omega}'.$$

Finally, the emerging spectrum $Y(\vec{\Omega},E)$ is found via the superposition

$$Y(\vec{\Omega},E)=\sum_{n=0}^{\infty}C_n(\vec{\Omega})F_n(E), \tag{10}$$

where the partial intensities $C_n$ are given by

$$C_n(\vec{\Omega})=\int_0^\infty\int_{4\pi}P_n(z=0,\vec{\Omega}|z_0,\vec{\Omega}_0)q(z_0,\vec{\Omega}_0)dz_0d\vec{\Omega}_0 \tag{11}$$

and $q$ is the source strength. Thus for a given boundary problem, Eqs. (6)–(11) completely specify the solution. To find this solution, Eq. (9) has to be solved for the boundary condition of the specific problem, where the scattering characteristics of the $n$th equation need to be evaluated at $E=\overline{E_n}$. The general solution approach for Eq. (9) is clearly beyond the scope of the present paper. A very simple, general, and efficient numerical approach to find the partial intensities for an arbitrary source distribution will be outlined further below. For the special but important case that elastic scattering can be neglected, the solution of Eq. (9) can be readily derived. This will be discussed in the next section.

In conclusion of this section it is noted that integration of the emerging spectrum, Eq. (10), gives the total intensity $I(\vec{\Omega})$. By virtue of the normalization of the partial energy distributions the total intensity is found to be equal to

$$I(\vec{\Omega})=\sum_{n=0}^{\infty}C_n(\vec{\Omega}). \tag{12}$$

This is the reason why the quantities $C_n$ are referred to as partial intensities. Finding the solution of the transport problem, Eq. (1), has thus been reduced to finding the appropriate partial intensities. Therefore the term partial intensity approach (PIA) is used for the proposed method. The partial energy distributions are given explicitly by Eq. (6) and thus the emerging spectrum can be calculated by Eq. (10) as soon as the partial intensities are known for the boundary problem of interest.

### B. Probability for $n$-fold scattering as a function of the path length

One of the main differences between the quasielastic energy regime and the energy regime where the constant cross section approximation no longer holds is the stochastic process governing multiple collisions. In the quasielastic case, the probability $W_n(s)$ for $n$ collisions as a function of the path length $s$ is given by the Poisson distribution $\mathcal{P}_n(s/\lambda)$:[26]

$$W_n^{\mathrm{QE}}(s)=\mathcal{P}_n(s/\lambda)\equiv\left(\frac{s}{\lambda}\right)^n\frac{e^{-s/\lambda}}{n!}. \tag{13}$$

In this section we will omit the subscript $i$ indicating inelastic scattering, for clarity. An important question in the present context is the behavior of the function $W_n(s)$ in the general case when the mean free path length depends on the energy. Just as in the quasielastic case, the equation describing $W_n(s)$ is found by letting $\lambda_e \to \infty$ in Eq. (9):

$$\frac{dW_n}{ds} = -\frac{W_n}{\lambda_n} + \frac{W_{n-1}}{\lambda_{n-1}}, \tag{14}$$

with the boundary conditions

$$W_n(s=0) = \begin{cases} 1, & n=0, \\ 0, & n>0. \end{cases} \tag{15}$$

Equations (14) and (15) can be solved by the method of variation of the constant. This leads to the recursion

$$W_n(s) = A_{nn}(s)\exp(-s/\lambda_n),$$

$$A_{nm}(s) = \frac{\lambda_m}{\lambda_m - \lambda_{n-1}}\left\{ A_{n-1,m} - A_{n-1,n-1} \times \exp\left( -s\left[ \frac{1}{\lambda_{n-1}} - \frac{1}{\lambda_m} \right] \right) \right\}, \tag{16}$$

$$A_{0,m}(s) = 1, \quad m \geq 0,$$

which completely specifies the solution. To study the physical properties of the distribution $W_n(s)$, it useful to consider its moments. These quantities are defined as

$$\langle s^k \rangle_n = \frac{1}{\mathcal{N}_n} \int_0^\infty W_n(s) s^k ds, \tag{17}$$

where the norm $\mathcal{N}_n$ of $W_n(s)$ is

$$\mathcal{N}_n = \int_0^\infty W_n(s) ds. \tag{18}$$

Multiplication of Eq. (14) by $s^k$ and integration immediately gives

$$\mathcal{N}_n = \lambda_n, \tag{19}$$

$$\langle s \rangle_n = \sum_{k=0}^{n} \lambda_k \equiv (n+1)\Lambda_n, \tag{20}$$

where $\Lambda_n$ is the average mean free path after $n$ collisions. Generally, one finds the following recursion for the moments:

$$\langle s^k \rangle_n = k\lambda_n \langle s^{k-1} \rangle_n + \langle s^k \rangle_{n-1}. \tag{21}$$

The above results allow us to numerically model the stochastic process beyond the quasielastic regime. However, a very effective and accurate analytic approximation for the distribution $W_n(s)$ is given by the expression

$$\mathcal{W}_n(s) = \frac{\lambda_n}{\Lambda_n} \mathcal{P}_n\left( \frac{s}{\Lambda_n} \right). \tag{22}$$

The advantage of this approximation is that all physically relevant properties of the stochastic process are immediately evident. Moreover, its accuracy is quite high, in particular for $n \lesssim n_{\max}$, so that it can be used in practical calculations. The moments of the approximate distribution $\mathcal{W}_n(s)$ are given by

$$\mathcal{N}_n^{\text{appr}} = \lambda_n, \tag{23}$$

$$\langle s^k \rangle_n^{\text{appr}} = \frac{(n+k)!}{n!} \Lambda_n^k. \tag{24}$$

Thus the position and the norm of the approximate and exact distribution are identical [cf. Eqs. (19) and (20)]. The ratio between the higher exact and approximate moments for values of $n$ not too close to $n_{\max}$ is bounded by the expression

$$1 \leq \frac{\langle s^k \rangle_n^{\text{appr}}}{\langle s^k \rangle_n} \lesssim \frac{1}{k} \sum_{l=0}^{k-1} \left( \frac{\Lambda_{n-1}}{\Lambda_n} \right)^l. \tag{25}$$

From Eqs. (23)–(25), it follows that the approximation (22) is quite good. For example, the deviation between the exact and approximate width $\sigma_n^2 = \langle s^2 \rangle_n - \langle s \rangle_n^2$ increases with $n$ and attains its largest value for $n = n_{\max}$. However, for a realistic energy dependence of the mean free paths,

$$\lambda(E) = \lambda(E_0)\left( \frac{E}{E_0} \right)^\alpha, \tag{26}$$

with $|\alpha| < 2$, the deviation in $\sigma_n^2$ does not exceed several percent, while it is negligible for $n \lesssim n_{\max}$. For small path lengths, when the energy dependence of the interaction characteristics can be neglected, the approximate and exact distribution $W_n(s)$ both reduce to the Poisson distribution, Eq. (13).

A comparison of the distribution $W_n(s)$ with the Poisson distribution is shown in Fig. (1). The parameters used in the calculation are $\lambda_0 = 25$ Å, $\langle T \rangle_n = n\langle T \rangle_0$, $n_{\max} = E_0/\langle T \rangle_0 = 30$, and $\alpha = 0.7$. For simplicity, the energy dependence expression (26) was adopted for the inelastic mean free path. The chosen parameters approximately correspond to 3 keV electrons traversing a Cu target (see Fig. 3 below). The exact [Eq. (16)] and approximate [Eq. (22)] function $W_n(s)$ are indistinguishably similar in this representation.

## III. APPLICATION TO INELASTIC ELECTRON REFLECTION FROM SOLIDS

The basic assumption of the proposed approach is that the energy loss and deflection process are independent. This allows us to separate the energy and direction in the kinetic equation for the group of $n$-fold inelastically scattered electrons. For medium-energy electrons this requirement is met to a very good approximation.[25] It should again be emphasized that the energy and angular variable in the total flux density are generally not uncorrelated at all. On the contrary, a correlation is found for isotropic emission of Auger electrons[10] and is particularly pronounced if the source angular distribution is sharply peaked as in the case of reflection experiments[17,27] when a well-collimated beam of particles is incident on a solid surface. Therefore, the latter case was selected to test the proposed approach.

The other basic point is the energy dependence of the interaction characteristics and the relation between the en-

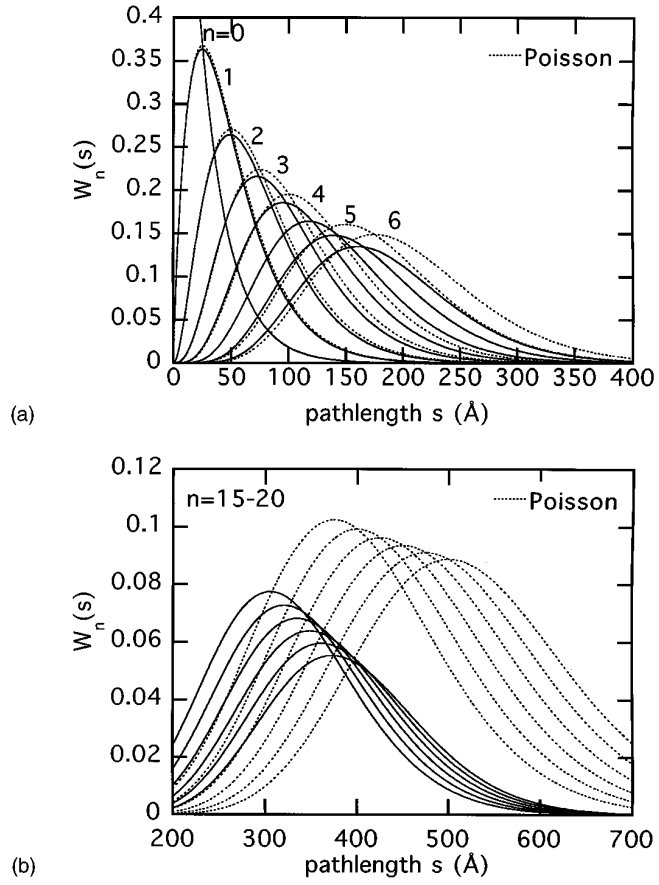


FIG. 1. Probability for $n$-fold scattering $W_n(s)$ as a function of the traveled path length. The solid lines correspond to the true slowing down case when the mean free paths for the interaction process vary with the energy. The exact [Eq. (16)] and approximate [Eq. (22)] distributions are indistinguishably similar in this representation. The dashed lines are the result in the quasielastic or constant cross section approximation, given by the Poisson distribution.

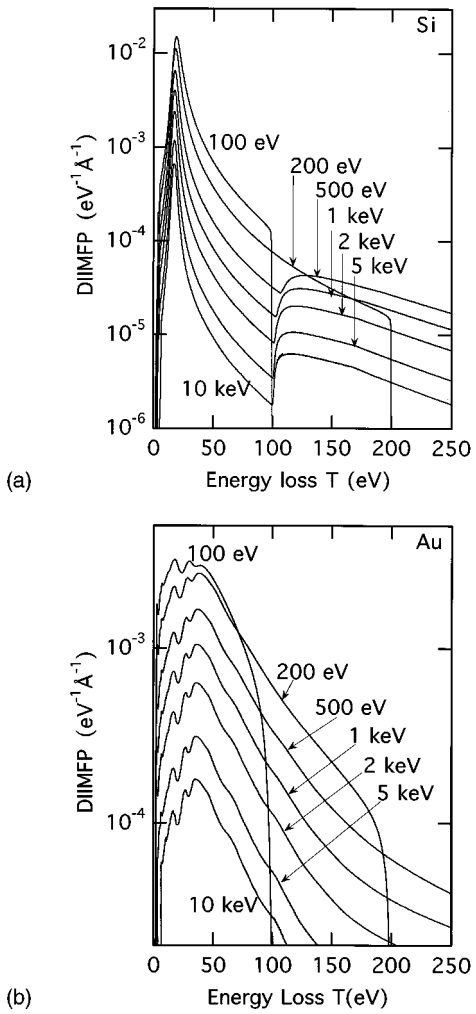


FIG. 2. Differential inverse inelastic mean free path (DIIMFP), i.e. , the energy loss probability, in individual collisions for electrons of different energies in solid matter: (a) Si and (b) Au.

ergy and the number of inelastic collisions. This will first be illustrated for the example of a medium-energy electron-solid interaction before we turn to the reflection problem. The basic quantity for the inelastic interaction is the differential inverse mean free path. The usual approach to establish this quantity is to extrapolate the optical limit of the dielectric function, corresponding to zero momentum transfer $\Delta\vec{q}=0$, onto the $(\Delta\vec{q},\Delta\omega)$ plane using an appropriate dispersion relation.[28,29,24,30] Following Penn[28] we have employed the single-pole approximation for the expansion function using a quadratic plasmon dispersion relation and applied it to the optical data compiled in Refs. 31 and 32. The resulting DIIMFP for two solids with distinctly different electronic properties, Si and Au, is shown in Fig. 2. While the loss function of Si is dominated by the strong and sharp plasmon excitation at $\sim$16 eV, the DIIMFP of Au is generally much broader, having its maximum at $\sim e^2/a_0$, corresponding to the binding energy of the outer electrons. The shape of the curves is seen to depend only rather weakly on the energy of the probing particle. Consequently, above a certain energy the mean energy loss is expected to be nearly independent of the number of inelastic collisions,

$$\langle T\rangle_n(E)=\int_0^\infty w_n(T,E)TdT\simeq\langle T\rangle_0, \tag{27}$$

since $w_n(T,E)$ is the normalized mean free path. In this case, Eq. (7) simplifies to

$$\overline{E_n}=E_0-n\langle T\rangle_0. \tag{28}$$

The actual dependence of the mean energy $\overline{E_n}$ after $n$ collisions is presented in Fig. 3 for several materials for $E_0=30$ keV. It is seen that Eq. (28) represents a good approximation over a broad range of inelastic processes corresponding to energies $\gtrsim$200 eV.

In order to calculate the partial intensities, values are needed for the elastic and inelastic mean free paths as well as the distribution of scattering angles for the discrete values $\overline{E_n}$. The IMFP was calculated from optical data[31,32] with the procedure developed by Penn,[28] while the elastic characteristics were evaluated from the Mott cross section for free atoms using a Thomas-Fermi-Dirac potential[33] and employing the partial wave expansion method, provided by the program of Yates.[34]

For a smooth source angular distribution an analytic solution for the partial intensities can be derived from Eq. (9) in the transport approximation in analogy to the quasielastic case.[10] According to the generalized radiative field similarity

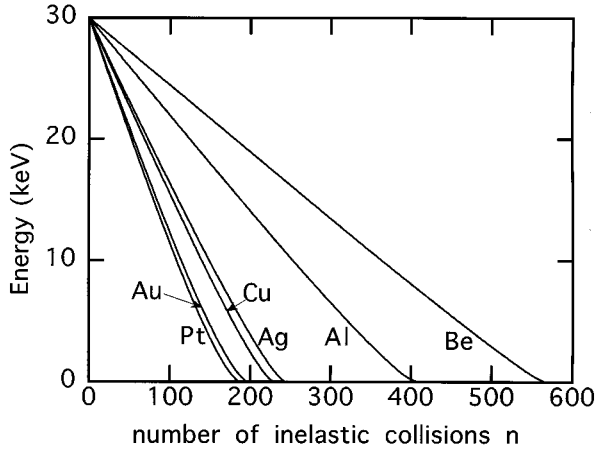


FIG. 3. Number of inelastic collisions an electron has experienced on the average after losing a certain amount of its energy for different elemental solids and an initial energy of 30 keV.

principle, the exact details of the deflection function need to be accounted for if the source angular distribution is highly anisotropic. This is exactly the case when a well-collimated beam of particles is reflected by a solid surface. Then analytic treatment can become rather complex[17] and it was therefore decided to directly evaluate the partial intensities employing a Monte Carlo scheme [see Eq. (33) below]. This can be done efficiently by means of the method of statistical weights: Only elastic scattering is explicitly modeled, and upon termination, a trajectory is weighted with the function $W_n(s)$. Of course, the change of the elastic characteristics with energy has to be accounted for. This can be done at each point of the trajectory by estimating the number of inelastic collisions, $n_i$, as a function of the traveled path length $\Delta s$:

$$n_i := \mathcal{I}\left( n_i + \frac{\Delta s}{\lambda_i} \right), \tag{29}$$

where the symbol $\mathcal{I}$ stands for the nearest integer. Then the energy dependence of the elastic characteristics can be incorporated by evaluating them at $E_{n_i}$. This algorithm is similar to the one by Jablonski[35] for the quasielastic case. The important difference consists in the use of the stochastic process when the scattering characteristics change with the energy. The approximate function $\mathcal{W}_n(s)$ has been used in the calculations since it is accurate enough and simple to evaluate. The most important advantage of this method over conventional Monte Carlo models[36,23] is that each trajectory contributes to all energies in the spectrum and not just to one channel. Therefore, convergence is attained very fast.

Once the partial intensities have been established in this way, the backscattering coefficient follows from

$$\eta(\vec{\Omega}) = \sum_{n=0}^{\infty} C_n(\vec{\Omega}). \tag{30}$$

The differential backscattering coefficient is obtained by calculating the partial energy distributions, Eq. (6), and weighting them with the partial intensities:

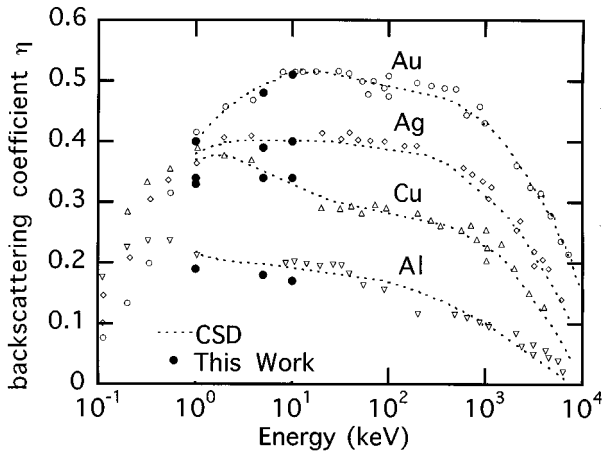


FIG. 4. Integral inelastic backscattering coefficient $\eta$ for Au, Ag, Cu, and Al. The open symbols represent experimental results, (Refs. 37–57), and the dashed lines were calculated using Tilinin's analytical expression Eq. (32) in the continuous slowing down approximation (Refs. 19 and 20). The solid symbols are the results of the partial intensity approach, proposed in the present work.

$$\frac{d\eta}{dE}(E,\vec{\Omega}) = \sum_{n=0}^{\infty} C_n(\vec{\Omega}) F_n(E). \tag{31}$$

In both cases, the actual experimental geometry has to be accounted for in the modeling of the partial intensities.

The result of this procedure is shown in Figs. 4 and 5 for 1–10 keV electrons normally incident on an Au, Ag, Cu, and Al targets for the total (Fig. 4) and differential (Fig. 5) backscattering coefficients. For the total backscattering coefficient, a large amount of experimental data is available in the literature,[37–57] which are represented by the open symbols in Fig. 4. The dashed line corresponds to Tilinin's expression,[19] which was corrected at lower energies for the contribution of

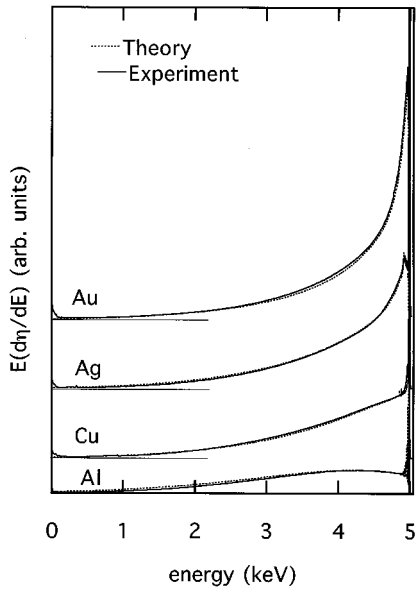


FIG. 5. Differential backscattering coefficient of 5 keV electrons normally incident on Au, Ag, Cu, and Al, measured with a CMA (solid lines) (Refs. 58–61). For comparison, the results of the presented approach are shown as dashed lines.

secondaries,[20] and Bethe's theory was used to determine the necessary inelastic interaction characteristics. Tilinin's expression was derived in the continuous slowing down approximation and reads[19,20]

$$\eta \simeq \frac{\sqrt{1+\sigma}-1}{\sqrt{1+\sigma}+1.9}, \tag{32}$$

where $\sigma = R/\lambda_{tr}$ is the scattering parameter in the continuous slowing down approximation[20,19] and $R$ is the electron linear range. In spite of its simplicity, it is seen that this formula perfectly describes the energy and atomic number dependence of the total backscattering coefficient. The results of the present work, represented by the solid symbols, also agree well with the experimental data. Note that in the present calculations generation of secondaries was completely disregarded.

Significant deviations between the presented theory and experiment can be observed, in particular for Al. A possible explanation for this fact is the use of the quadratic dispersion relation to calculate the IMFP. Ding and co-workers[58] also note in their work that this may lead to significant deviations. Furthermore, Penn's formalism to calculate the IMFP at energies above $\gtrsim 10\,000$ eV has not been substantiated by a direct comparison of experimental IMFP values with theory and may therefore also introduce a certain error in the reflection probability. It should be stressed that the observed deviations are definitely not due to the proposed approach for the particle transfer. This follows from a comparison of the presented data with results of a conventional Monte Carlo simulation that was also performed. These data mutually agree to within less than 1%. Hence it is likely that the input parameters of our calculation are responsible for the deviations.

The advantage of the presented theory lies in the fact that it also describes the energy dependence of the phenomenon fairly well, as can be seen in Fig. 5. Here, theory is compared with the experimental data by Goto and co-workers.[58–61] These data are special in that they are not distorted by the analyzer electron optics or electronics and therefore represent the true energy distribution, allowing a meaningful comparison to be made. Such data have only recently become available.[59,62,63]

The theoretical results were normalized to experiment at the energy just below the elastic peak. Although the general agreement is quite good, theory is in error at lower energies by a factor sometimes exceeding 2. In this respect, the Monte Carlo results of Ding and co-workers[58] agree better with the data.

In conclusion of this section, it is noted again that for the present case of the reflection problem, the solution of Eq. (9) has to be found for the realistic Mott cross section for elastic scattering in order to obtain quantitative agreement with the experimental data. This follows directly from the radiative field principle and was confirmed by additional calculations performed for an isotropic cross section. In this case, the differential backscattering coefficient deviates significantly from experiment. Therefore the numerical approach that was adopted here seems to be the most efficient way to establish the partial intensities for the backscattering problem.

## IV. DISCUSSION

Two main assumptions have been made in the present work [see Eqs. (4) and (5)], allowing us to describe the transfer of fast charged particles in matter and accounting for the energy dependence of the interaction characteristics. The first assumption states that the energy loss and deflection process should be independent. In the theory section, Sec. II, it was outlined that such an assumption is justifiable when the transport mean free path for inelastic scattering greatly exceeds the elastic one and generally this assumption holds for electrons interacting with solids over a broad energy range. This conclusion is supported by the good agreement between theory and experiment in Figs. 4 and 5. It is important to emphasize again that the energy and the angular variable in the total flux density are correlated.[10,27] For example, if the detection or incidence angle is changed in the reflection experiment, the energy distribution changes markedly.[27,64] The reason is that the path length distribution changes in this case and consequently the relative contribution of the $n$th group, i.e., the partial intensities, also changes, giving rise to a different emerging energy spectrum.

Next, the assumption that the energy in the $n$th group may be replaced by the mean energy after $n$ collisions is due to be discussed. Clearly, for sufficiently small values of $n$ where the constant cross section approximation applies due to the weak variation of the scattering parameters with the energy, this assumption poses no problem. For in that case, the width of the partial energy distributions is small compared to the energy range over which a noticeable change in the scattering characteristics occurs.

To fully appreciate the situation for larger energy losses, it is useful to regard the path length distribution function. This quantity is defined as the distribution of path lengths that the particles travel for a given boundary problem. On recalling that the partial intensities are the contributions to the spectrum of particles that have experienced $n$ losses one can establish a relationship between these quantities and the path length distribution. The partial intensities can simply be expressed as the product of the path length distribution $Q(s,\vec{\Omega})$, and the probability $W_n(s)$ for $n$ losses for a given path length, and integrated over all path lengths:

$$C_n(\vec{\Omega}) = \int_0^\infty Q(s,\vec{\Omega}) W_n(s) ds. \tag{33}$$

Now it is clear that for large values of $n$, the energy spread as a cause of statistical fluctuations in the partial energy distributions is small compared to the uncertainty in the energy introduced by the broad path length distribution. This allows us to neglect the energy fluctuations within a group for a large number of collisions.

This is illustrated in Fig. 6, which compares the experimental reflection coefficient with the theoretical result and also shows the partial intensities as a function of the mean energy after $n$ collisions. The latter quantities are represented by open circles and were normalized to theory at 3 keV. This comparison clearly shows the transition between the quasi-elastic regime, where the energy fluctuations governed by the DIIMFP determine the overall energy distribution, and the

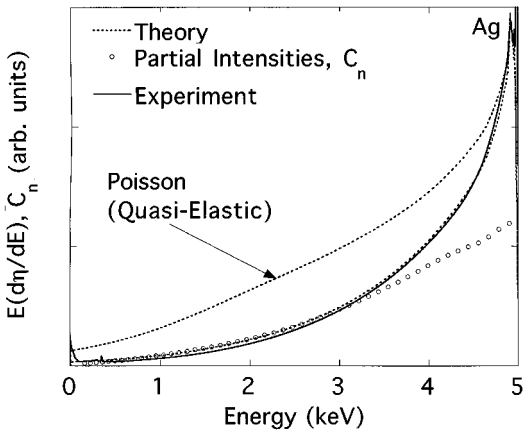


FIG. 6. Comparison of the experimental backscattering coefficient (solid line) for 5 keV electrons reflected from a solid silver target with theory (dashed line). Dashed line labeled ''Poisson'': result of the convolution method in the constant cross section approximation. Open circles: partial intensities (normalized to theory at 3 keV).

regime where the path length distribution (or partial intensities) plays the dominant role: Below $\sim 3$ keV, the energy dependence of the partial intensities quite perfectly coincides with the spectral shape of the backscattering coefficient. For the latter regime, the assumption of continuous energy loss is clearly reasonable. The major advantage of the partial intensity approach is that it encompasses both energy regimes merely by the use of the proper stochastic process, Eq. (22). Thus its level of complexity is almost the same as the convolution method in the constant cross section approximation that relies on the Poisson stochastic process while its accuracy is generally much higher. This can also be seen in Fig. 6 where the result of the convolution method is shown as the dashed line labeled ''Poisson.''

In passing we have introduced a third approximation that deserves to be discussed, although it is not essential. It is the approximate expression $\mathcal{W}_n(s)$ for the stochastic process of multiple scattering. In view of its simplicity, its accuracy is remarkably high. It is interesting to note that it would even coincide with the exact solution if the right-hand side of Eq. (25) were equal to unity. We rewrite this condition in the form

$$\frac{\Lambda_n^k-\Lambda_{n-1}^k}{\Lambda_n-\Lambda_{n-1}}=k\Lambda_n^{k-1}. \tag{34}$$

For a continuous variable $\Lambda_n$ (or $n$) this is identical to the differentiation operation. Thus, it turns out that replacement of $W_n(s)$ with the approximate distribution $\mathcal{W}_n(s)$ [Eq. (22)] constitutes an approximation similar to making the transition from the continuous variable to the discrete variable describing the energy of a particle.

Finally, it seems advisable to place the presented method in context with other approaches that account for the change of the interaction characteristics with energy. We will first confine the discussion to the case when angular deflections can be neglected. The connection of the present basic assumptions with those usually made is best illustrated by considering Landau's approach[6] in the continuous slowing down approximation (CSDA). In this approach the energy loss spectrum is written as

$$G(s,E)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dk\exp\left\{ik(E_0-E)-\int_0^s\frac{ds'}{\lambda_i}[1-\tilde{w}(\overline{E(s')},k)]\right\}, \tag{35}$$

where $\tilde{w}$ is the Fourier transform of the differential mean free path. The particles' energy as a function of the path length $\overline{E(s)}$ is obtained in the CSDA by expressing the energy loss as the product of the stopping power $\langle T\rangle/\lambda_i$ and the path length $s$. When the path length is small enough, the energy dependence of the quantities in the second term of the exponent may be neglected and this term reduces to $s(1-\tilde{w})/\lambda_i$. Expanding the exponent in powers of $\tilde{w}$, we immediately arrive at the energy loss function expressed in terms of the $n$-fold convolution of the inelastic cross section, the so-called partial loss distributions $L_n$ [cf. Eq. (6)]:

$$G(s,E)=\sum_{n=0}^{\infty}L_n(E)W_n(s). \tag{36}$$

This is the standard result for the energy loss function in the convolution method approach. Since it was assumed that the interaction characteristics do not depend on the energy, the probability for $n$-fold scattering, $W_n(s)$ in expression (36), is given by the Poisson distribution. The partial intensity approach merely consists of replacing the Poisson distribution by the general probability for $n$-fold scattering, $W_n(s)$. This becomes practical on account of the effective approximation $\mathcal{W}_n(s)$ for the stochastic process. In this sense, Eq. (22) is the central result of the present work.

The above clearly demonstrates the conceptual simplicity of the partial intensity approach: For a given group of $n$-fold scattered particles, the probability that a particle that has traveled a path length $s$ belongs to this group is given by $W_n(s)$. Multiplying this with the energy (loss) distribution associated with the $n$th group, $L_n(E)$, then gives the energy loss function for this group as a function of the traveled path length. Summing up the contribution of all groups, one obtains the total energy loss function $G(s,E)$, Eq. (36). Generally, in the partial intensity approach, many expressions for relevant quantities like the loss function (36), the spectrum (10), etc., are essentially trivial once the concept of dividing the flux density into groups of $n$-fold scattered particles is clearly understood.

Both approaches, Eqs. (35) and (36), are similar in the way they account for the energy dependence of the interaction characteristics: These should be evaluated at the energy $\overline{E_n}$ corresponding to the independent variable keeping track of the particles' energy:

$$\overline{E(s)}\simeq E_0-\frac{s}{\lambda}\langle T\rangle \quad \text{(CSDA)}, \tag{37}$$

$$\overline{E_n}\simeq E_0-n\langle T\rangle \quad \text{(PIA)}. \tag{38}$$

According to Poisson statistics, the mean number of collisions after a given path length $s$ is just $s/\lambda$. This illustrates that both approaches are equivalent in this sense, the essential difference, of course, being the use of a continuous as opposed to a discrete variable keeping track of the energy. The freedom we have in choosing the independent variable is related to the fact that the energy variable does not explicitly enter the transport equation, as already pointed out by Landau.[6] The validity of both approaches is restricted by the same underlying assumption that energy fluctuations are small after traveling a certain path length (or experiencing a given number of collisions) [see Eq. (5)].

Now let us consider the situation when angular deflections are significant. This will lead to enhancement of the traveled path lengths compared to the straight line case. In other words, the distribution of path lengths $Q(s,\vec{\Omega})$ will be broadened owing to angular scattering. Using Landau's approach, the spectrum is obtained by multiplying the energy loss function with the path length distribution and integrating over the path length:

$$Y(E,\vec{\Omega})=\int_0^\infty Q(s,\vec{\Omega})G(s,E)ds. \tag{39}$$

It is now seen that in the conventional CSDA it is also assumed that energy losses and deflections are independent. This becomes even clearer by inserting Eq. (36) into Eq. (39) and using the definition for the partial intensities, Eq. (33). The result exactly corresponds to the formula for the spectrum in the partial intensity approach, Eq. (10). Thus the assumption underlying Eq. (4)—that energy losses and deflections are independent—is also very common and the associated approaches have a broad field of application. One important example where this assumption is invalid is when inner shell excitations are significant, as in angle-resolved electron energy loss spectroscopy. Even in that case, however, a yield equation of the type of Eq. (10) can be used, but the partial energy distributions will be geometry dependent. See Ref. 26 for further details.

Summarizing, it may be stated that the results of the present work allow us to extend the convolution method to the case of energy-dependent interaction characteristics in a very simple way by using Eq. (22) for the stochastic process for multiple collisions. The basic assumptions as well as the range of validity are the same as for Landau's approach in the continuous slowing down approximation.

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