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Abstract

Monte Carlo calculations are used to obtain the energy loss and spatial distribution of electrons penetrating matter. For this purpose, reliable cross section for the inelastic collisions must be known. As an approximation valid for large energy losses, the Coulomb cross section can be used. It can be modified in a simple way to account for the binding of electrons and for the exchange effect. In the Gryzinski model, collisions with moving electrons are assumed. In the quantum mechanical Bethe approximation, σ is closely related to the dipole oscillator strength (DOS), and its extension to finite momentum transfers, the generalized oscillator strength (GOS). Therefore, the influence of the state of a material on DOS is shown for the example of gaseous and solid silicon. Some details of the Bethe model are given for Si. The Bethe asymptotic approximation to the stopping power is derived, and the reason for the shell corrections is demonstrated. Collision cross sections calculated with three different models are compared. In general, models based on a detailed knowledge of the GOS should be used for applications.

Keywords: Monte Carlo, electron energy loss, differential collision cross section, silicon, Bethe theory, Gryzinski model, moments, stopping power, dipole oscillator strength, generalized oscillator strength.

Introduction

The energy loss of low energy electrons and positrons (kinetic energy E less than 10 keV) has been discussed in many recent papers, e.g. Nieminen (1988), Valkealahti and Nieminen (1983, 1984), Schou (1979), Liljequist (1983), Mills and Wilson (1982). Frequently, their paths are simulated with Monte Carlo calculations. Then a knowledge of the doubly differential inelastic collision cross section $\sigma(\epsilon, \theta)$ is of great importance (ϵ is the energy loss of the incident electrons in each collision, θ the angle of deflection). The cross section for elastic scattering of the electrons by the nuclei, $\sigma_e(\theta)$, is equally important, but will be discussed elsewhere. The word electrons shall mean "electrons and positrons", and the lower limit of the energy of the incident electrons is about 1 keV.

In general, electrons will suffer many collisions, losing in each only a small amount of energy, ϵ, and changing the direction of motion by a small amount. An initially narrow parallel beam will therefore spread into a broad pattern, eventually resembling a swarm of mosquitoes (Valkealahti and Nieminen, 1983, 1984). For most solids, the most probable energy loss in a single collision is between 15 and 25 eV (Ahn and Krivanek, 1983), the mean energy loss is about 50 eV. Specifically, for Si, for $E < 10keV$, $< \epsilon > \approx 8.9eV \cdot \ln(E/R)$, where R is the Rydberg energy (13.6 eV). Each electron will follow its own path, different from that of all others ("straggling").

For parallel beams of particles with energies of the order of MeV, traversing thin absorbers, the beam emerging from the absorber will have a small spread in energy and angle, and its average properties can be approximated quite well by averages calculated from the *moments* of the cross sections. Results for the moments therefore are given here. For low energy electrons, though, a reasonable description of the energy loss processes requires detailed transport calculations, in particular Monte Carlo calculations may be appropriate (e.g. Valkealahti and Nieminen, 1983, 1984). These calculations can be designed to reproduce the sequence of events experienced by the particles quite well.

Table of Symbols

B, B_i	binding energy of electrons, in atomic shell i
c	speed of light
dE/dx	stopping power
e	electric charge of electron
E	kinetic energy of incident electron
$f(\epsilon, 0)$	dipole oscillator strength
$f(\epsilon, K)$	generalized oscillator strength, Eq. (10)
$F(\epsilon, K)$	matrix element, Eq. (9)
g, h, j	parameters in Eq. (22)
I	mean excitation energy of absorber, Eq. (15)
k	constant of Coulomb cross section, Eq. (4)
K	momentum transfer in collision
m	rest mass of electron, mc^2=511 keV
M	rest mass of incident particle
M_ν	ν'th moment of collision cross section, Eq. (2)
${}_cM_1$	contribution to M_1 from close collisions, Eq. (14)
${}_dM_1$	contribution to M_1 from distant collisions, Eq. (16)
n	number of atoms in unit volume
n_i	number of electrons in atomic shell i
N	number of electrons in a beam, Eq. (1)
Q	$= K^2/2m$
Q_m	minimum energy transfer in collision, Eq. (12)
Q_M	maximum energy transfer in collision, Eq. (18)
Q_1	separates distant and close collisions, Eq. (12)
r_0	$= e^2/mc^2$, classical electron radius= $2.818{\cdot}10^{-13}$ cm
R	Rydberg energy= 13.6 eV
u	speed of electron in atomic orbit
v	speed of incident particle
x	absorber thickness
z	charge number of incident particle
Z	atomic number of absorber
β	=v/c
δ	kinetic energy of secondary electron
Δ	energy loss in an absorber of thickness x
ϵ	energy loss of incident particle
ϵ_1	energy of lowest excited state of atom
ϵ_M	maximum energy loss of incident particle
ϵ_p	plasmon energy
θ	angle of deflection
κ	complex dielectric function
λ	mean free path, below Eq. (2)
μ	average number of collisions in absorber
ν	index for moment $M_\nu, \nu = 0, 1, 2, \cdots$
$\rho(\epsilon)$	Coulomb cross section, Eq. (3)
σ	collision cross section
$\sigma(\epsilon, \theta)$	collision cross section differential in energy loss and angular deflection
$\sigma(\epsilon, Q)$	collision cross section differential in energy loss and momentum transfer, Eqs. (8) and (11)
$\sigma(\epsilon)$	collision cross section differential in energy loss
$\sigma_e(\theta)$	elastic collision cross section differential in angular deflection
$\sigma_L(\epsilon)$	asymptotic collision cross section for bound electrons, Eq. (5)
$\sigma_M(\epsilon)$	Møller cross section differential in energy loss, Eq. (6)

Since many aspects of the problem will be discussed in other contributions to the conference, I shall discuss here only the inelastic cross sections. It is easy to derive the differential cross section for the collision of an electron with a free electron ("Coulomb cross section"). Various modifications of the Coulomb cross section have been made to include the effect of electron binding. Some are described here, and are compared with each other. Further examples may be found in Bichsel (1988). First, though, some general ideas will be examined.

2 Notes about inelastic collision cross sections

If a beam of N monoenergetic electrons with kinetic energy E (speed $v = \beta \cdot c$) passes through an absorber with atomic number Z, of thickness dx and with n atoms per unit volume, the number of electrons experiencing a collision with energy loss ϵ is

$$dN(\epsilon) = NnZ\sigma(\epsilon)d\epsilon dx, \qquad (1)$$

where $\sigma(\epsilon)$ is the collision cross section differential in ϵ; it depends on β. For the sake of simplicity, it is assumed that angular deflections can be disregarded. Eq. (1) shows how $\sigma(\epsilon)$ is obtained from experimental measurements. Here, I discuss theoretical approaches to the determination of $\sigma(\epsilon)$. First, some general observations are presented.

Since inelastic collision cross sections show a distinct structure near the ionization energies, it is useful to consider the cross sections separately for each electron shell. It is important to realize that large changes occur in the constitution of valence shell electrons if separate atoms (or molecules) are compared with the solid of the same composition (Fig. 1). In single atoms separated by large distances, the smallest energy losses are to discrete excited electronic states, and ionization begins at the binding energies B_i above which electrons are given a kinetic energy $\delta = \epsilon - B_i$. For ionization, energy losses are continuous (Fig. 1). If atoms are brought closer and closer together until they coalesce into a liquid or a solid, their valence electrons come under the influence of the cores of many atoms. A core is defined to consist of the nucleus and all the electrons inside of the valence shell. For carbon or oxygen, this would be only two electrons in the K-shell, for aluminum or silicon it would contain two electrons in the K-shell and eight electrons in the L-shell. For metals, the valence electrons will form a conduction band in which they are free. This means that it will take very little energy to move them, if for example an electric field is applied. On the other hand, if a charged particle moves through the solid, the transfer of very small energies by Coulomb interactions with individual electrons is not observed, instead, a large number of them is excited in each interaction of the particle. For metals this process is called a plasmon excitation, for insulators a collective excitation, Fig. 1. For most substances the most probable value of

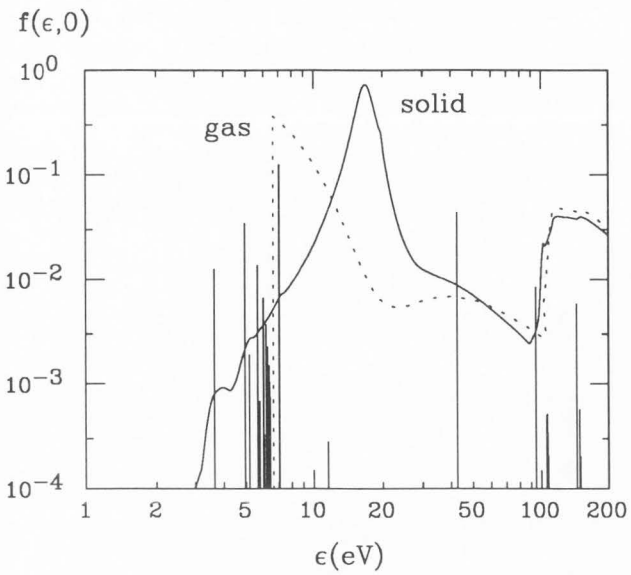


Fig. 1. Comparison of the dipole oscillator strength spectra for atomic (vertical and dotted lines) and solid silicon (solid line). The ordinate is DOS, $f(\epsilon, 0)$, in eV^{-1}, the abscissa the energy loss ϵ in eV. Data for the atom were calculated by Dehmer et al. (1975), based on numerical calculation with the Herman-Skilman potential and ground state wavefunction, and single electron excited state wavefunctions. Data for the solid are from Bichsel (1988) and were obtained from various experimental and theoretical sources. The broad peak at $\approx 17eV$ represents the collective excitations. While the uncertainties of certain features and at some energy losses may exceed 10%, the general structures remain well represented. Particularly notable is the disappearance of the discrete atomic excitations in the solid, and the large shift in the peak of the continuum excitations for the valence shell (from $\approx 7eV$ to $17eV$). On the other hand, at the L-shell edge ($\approx 100eV$), the shifts in the energies and in the shape of the function are relatively small. The I-values, defined in Eq. (15), are 131 eV for the atom, 174 eV for the solid.

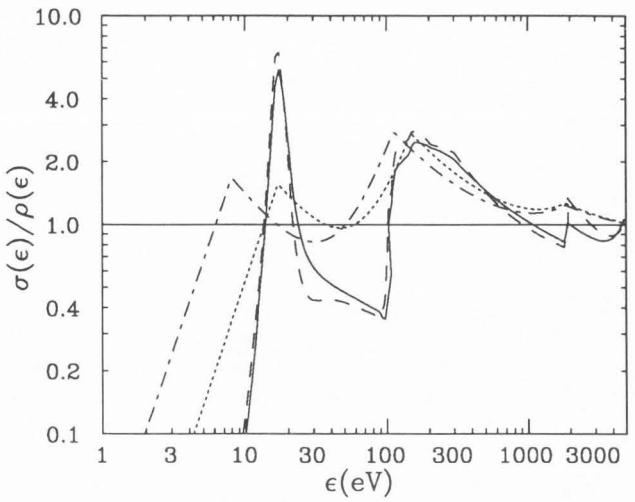


Fig. 2. Comparison of the single collision cross sections $\sigma(\epsilon)$ for incident electrons with an energy $E = 10keV$, calculated with different theories. In order to show the structure of the functions clearly, the ordinate is $\sigma(\epsilon)/\rho(\epsilon)$, where $\rho(\epsilon)$ is the Coulomb cross section, Eq. (3), the abscissa is the energy loss ϵ. The Coulomb cross section (for $Z = 14$ electrons) is represented by the horizontal line at 1.0. The functions given by Eqs. (5) and (6) are not shown. The solid line was obtained with the Bethe theory, Eq. (11), using the data described by Bichsel (1988). It includes the Møller-Mott cross section (Uehling, 1954), resulting in the increase at $\epsilon > 3000eV$. The cross section calculated with the DOS approximation of Eq. (23) is given by the dashed line. The dotted line was calculated from the binary encounter theory, Eq. (7), including K-, L- and M-shell excitations with the parameters $B_K = 1840eV(n = 2), B_L = 150eV(n = 8)$, and $B_M = 17eV(n = 4)$. The dashed-dotted line was calculated with $B_K = 1840eV(n = 2), B_L = 112eV(n = 8)$, and $B_M = 8eV(n = 4)$, used by Valkealahti and Nieminen (1983). Clearly, the assumption of an ionization energy for the M-shell at 8 eV differs substantially from the observed plasmon energy. The Gryzinski expression with $B_L = 150eV$ approximates the behaviour of the L-shell not too badly, but is quite bad for the M-shell. It may be noted that, for the solid line, 80% of the total collision cross section accumulates up to 33 eV, while for the dashed-dotted line this already occurs at 19 eV. This explains the large values of M_0 for G-8 and G-L in Table I.

the plasmon excitation energy, ϵ_p, is much larger than the energy of the lowest excited state of the atom, ϵ_1. For example, for Be, $\epsilon_1 = 3.6eV$ (calculated by Dehmer et al, 1975), while the plasmon energy is $\epsilon_p = 19eV$ (shown in Ahn and Krivanek, 1983). Similarly, for silicon, Fig. 1, $\epsilon_1 = 3.6eV$ (Dehmer et al., 1975), $\epsilon_p = 16.7$ eV (Bichsel, 1988).

For molecules, experimental measurements of electron energy losses (Killat, 1973), provide information about the difference between gas and solid and the structure of $\sigma(\epsilon)$. For example, for Benzene C_6H_6, the vapor showed distinct structures for excitations to several discrete states; in addition a broad peak which to me appears to be equivalent to a collective excitation at about 16 eV was seen; for the solid, the structures were broadened, and the major peak for the collective excitations had shifted to 21 eV.

For energy losses well above that of the plasmon or collective excitation peak, the collision cross section decreases smoothly until the ionization energy of the next electron shell is reached and a new peak is superimposed on the smooth function (e.g. the K-shell in carbon at $\epsilon \approx 285eV$, or the L-shell in silicon at $\approx 100eV$, Fig. 2; in gold, though, the lowest N-shell excitations appear only

as a change in slope, but the lowest M-shell produces a distinct peak at $\approx 2200eV$, Ahn and Krivanek, 1983).

While there are large changes in the structure of the excitation of the valence electrons as the atoms are coalesced into a solid (Fig.1), changes are less important for the inner shells. The main reason for this is that the binding energies of these shells are quite large, while the energies associated with the chemical changes are small. As an example, consider carbon. For an amorphous, thin carbon film, the K-edge occurs at 284 eV (Ahn and Krivanek, 1983), and K-shell excitation shows essentially just one peak at $\approx 296eV$. For three molecules important in biology: adenine, uracil and thymine, Isaacson (1972) found several peaks, all located between 284 and 300 eV. Thus there is at most a small change in the energy of the K-shell excitation, but the number of peaks as well as their locations change considerably. For aluminum, above the K-edge (1.56 keV), Pianetta and Barbee (1988) observed a structure with several peaks, each separated from the previous one by about 40 eV. They describe this structure as extended x-ray absorption fine structure (EXAFS); it is caused by the presence of nearby cores which backscatter the photoelectrons and thus change their wavefunctions. The discrete excitations of the atom below the K-edge disappear completely. Solid state and chemical effects thus are very important for valence shell electrons, less so for inner shell electrons.

While the *angular dependence* of the differential collision cross sections can be determined from the quantum mechanical calculations, it is sufficient for many purposes to relate θ, the angle of deflection of the incident electron, to the energy loss with the simple classical kinematic relation $\theta \approx (\epsilon/E)^{1/2}$, and to forget about the spread in angle. Then it is sufficient to consider the cross section $\sigma(\epsilon)$ differential in energy loss ϵ only. This approach has been used in many studies. The kinematic calculation (Nieminen, 1988) has also been used to obtain the angle of emission of the secondary electron, but it should only be used for large ϵ. For smaller ϵ, an isotropic distribution for the secondary electrons might be more appropriate (Kim, 1983).

A variety of models have been used to obtain theoretical $\sigma(\epsilon)$ for bound electrons. Here, four of them will be described and compared with each other. They are expressed in terms of a factor modifying the Coulomb cross section. First, though, it is useful to define the moments of $\sigma(\epsilon)$.

3 Moments of the differential collision cross section $\sigma(\epsilon)$

The moments of $\sigma(\epsilon)$ are defined by (Bichsel, 1988):

$$M_\nu \equiv \int \epsilon^\nu \sigma(\epsilon) d\epsilon \qquad (2)$$

In general, $\sigma(\epsilon)$ will have finite values over a certain range of values of ϵ and will be zero elsewhere, thus no limits need to be defined for the integral.

The moment M_0 is usually called the total collision cross section and determines the average number μ of collisions in an absorber of thickness x : $\mu = x \cdot M_0$. The inverse of M_0, λ, is the mean free path between collisions, and is used in Monte Carlo calculations.

The moment M_1 is called the stopping power dE/dx. The mean energy loss Δ in a thin absorber of thickness x is $\Delta = x \cdot dE/dx$.

The second moment, M_2, is related to the width of straggling functions. Higher moments are not very useful (Bichsel, 1970) except for special applications (Tschalär, 1968).

It must be kept in mind that for incident electrons, because of multiple scattering, these moments are useful only if one follows the paths of different electrons and averages over energy loss along these paths.

4 Coulomb cross section and simple derived models

Since the inelastic interactions of electrons with matter are mainly due to collisions with electrons in the material, it is useful to write down the cross section for the collision of a charged particle with a free electron at rest. This (nonrelativistic) Coulomb cross section $\rho(\epsilon)$ for an energy loss ϵ in the collision of a charged particle with charge ze, rest mass M and speed v with a free electron with charge $-e$ and rest mass m, in the laboratory system is given by (Evans, 1967; Bichsel, 1968; Inokuti, 1971):

$$\rho(\epsilon) = \frac{2\pi z^2 e^4}{mv^2\epsilon^2} = \frac{k}{\beta^2\epsilon^2} \quad \text{with} \qquad (3)$$

$$k = \frac{2\pi z^2 e^4}{mc^2} = 2\pi r_o^2 mc^2 z^2 = 2.54955 \cdot 10^{-19} z^2 eV\,cm^2, \qquad (4)$$

$r_o = e^2/mc^2$ the classical electron radius, $\beta = v/c$ and c the speed of light. Since the secondary electron receives all the energy ϵ lost by the incident particle, the momentum transfer K is determined by $\epsilon = K^2/2m$. $\rho(\epsilon)$ does not depend on M, and gives a good approximation for the collisions with bound electrons if the energy transfered to these electrons greatly exceeds their binding energy (Fig. 2).

For electrons bound in matter with binding energy B, average kinetic energy U and an average speed u, a useful approximation for the cross section follows from binary encounter theory:

$$\sigma_L(\epsilon) = \rho(\epsilon) \cdot (1 + \frac{4}{3}\frac{U}{\epsilon}). \qquad (5)$$

It is valid for $v \gg u$ and $\epsilon > 10U$ (Inokuti, 1971), and agrees well with quantum mechanical calculations (Bichsel, 1988).

For collisions of electrons with electrons, the exchange effect must also be taken into account. This can be achieved with the Møller formula (Uehling, 1954). For free elec-

trons, the nonrelativistic expression is given by:

$$\sigma_M(\epsilon) = \rho(\epsilon)[1 - \frac{\epsilon}{E-\epsilon} + (\frac{\epsilon}{E-\epsilon})^2] \qquad (6)$$

The exchange effect is small for small ϵ, and σ_M will counteract σ_L with increasing ϵ; for example, with $B = 16eV$, $\epsilon = 160eV$, and $E = 1keV$, the increase of σ_L over ρ is approximately equal to the decrease of σ_M, while for $E = 10keV$, the decrease of σ_M is small. Kim (1975) gave an expression combining Eqs. (5) and (6). The Bhabha cross section for the scattering of positrons on electrons is also given in Uehling (1954).

It appears advisable to compare any cross section based on given theoretical assumptions with the asymptotic cross sections σ_L and σ_M (e.g. at E=10 or 20 keV). Furthermore, since $\rho(\epsilon)$ has a very simple functional form ($\rho \sim \epsilon^{-2}$), it will be useful to plot the ratio $\sigma(\epsilon)/\rho(\epsilon)$ for a cross section $\sigma(\epsilon)$ (Fig. 2).

Gryzinski's binary encounter model

By considering the orbital speeds of electrons in a material, and using classical mechanics and the Coulomb cross section, Gryzinski (1965) was able to refine Eq. (5) and obtain total collision cross sections M_0 and stopping powers M_1 which agreed quite well with experimental data. The following expression is a modification of the equation which has been used by Valkealahti and Nieminen (1983) to describe the Gryzinski cross section:

$$\sigma(\epsilon) = \rho(\epsilon)\sum_i n_i[1 + (B_i/E)]^{-3/2} \cdot (1 - \frac{\epsilon}{E})^{B_i/(\epsilon+B_i)} \cdot \left\{(1 - \frac{B_i}{E}) + \frac{4}{3}\frac{B_i}{\epsilon}\ln\left[2.7 + (\frac{E-\epsilon}{B_i})^{\frac{1}{2}}\right]\right\} \qquad (7)$$

where E is the energy of the incident electrons, and n_i the number of electrons in the shell i, with binding energy B_i. For energy losses below B_i, the approximation given by these authors is used: for $\epsilon < B_i$, $\sigma(\epsilon) = \sigma(B_i)$ (this is their model MC-2). Note that for large energies of the electrons, we have $1 + (B_i/E) \to 1, [1 - (\epsilon/E)]^{B/(\epsilon+B)} \to 1$ and $1 - (B_i/E) \to 1$, and then Eq. (7) is the same as Eq. (5), except that the logarithm is greater than 1.

As an example, values of $\sigma(\epsilon)$ and its moments were calculated for silicon, with two sets of parameters for the binding energies and the number of electrons in each shell. The first set was chosen to give peaks in $\sigma(\epsilon)$ where they appear in the Bethe model: $B_K = 1840eV, n_K = 2, B_L = 150eV, n_L = 8, B_M = 17eV, n_M = 4$. The second set was used by Valkealahti and Nieminen (1983): $B_K = 1840eV, B_L = 112eV, B_M = 8eV$, with the same number of electrons in each shell. These cross sections, calculated for 10 keV electrons, are shown in Fig. 2; and moments for three different electron energies are shown in Table I. The moments for the model MC-1 (where $\sigma(\epsilon) = 0$ for $\epsilon < B_i$) of Valkealahti and Nieminen (1983) are also given. Clearly, in Eq. (7), the choice of the smallest B_i (for silicon, this is for the M-shell), influences the value of M_0 greatly, it is of lesser importance for M_1 or M_2. The inclusion of the K-shell electrons influences M_0 very little, but changes M_1 by $\approx 5\%$ and M_2 by 15%.

Table I. Moments of $\sigma(\epsilon)$

Moments of the singly differential collision cross section spectrum $\sigma(\epsilon)$, for electrons with initial energy E(keV) in silicon, calculated with different theories of $\sigma(\epsilon)$. M_0 is given in units of number of collisions per μm, M_1 is the stopping power in MeV/cm, M_2 is the moment related to straggling, in units of keV^2/cm. The mean energy loss per collision, $< \epsilon >$, is given by M_1/M_0, and is in units of eV. Sources for the calculations are: Si - Bethe model, described in section 5; DOS - model of Eq. (23); G-17 - Gryzinski model, Eq. (7), with $B_M = 17$ eV, n=4 for the M-shell, $B_L = 150$ eV, n=8 for the L-shell, $B_K = 1840$ eV, n=2 for the K-shell; G-8 is calculated with the parameters used by Valkealahti and Nieminen (1983) ($B_M = 8eV$, $B_L = 112$ eV, $B_K = 1840$ eV). For G-L, the K-shell was excluded. For G-0, the contribution for $\epsilon < E_B$ was excluded (model MC 1 of Valkealahti and Nieminen, 1983, but the K-shell contribution is included again), and the changes in M_0 and M_1 are very large. For comparison, the stopping power given in ICRU-37 (1984) is also shown.

E	source	M_0	M_1	M_2	$< \epsilon >$
100	Si	0.105	7.51	27,600	71.3
	G − 8	0.237	8.43	32,000	35.6
	ICRU		7.61		
10	Si	0.646	37.5	24,100	58.1
	DOS	0.707	39.5	25,200	55.9
	G − 17	0.749	41.1	28,200	54.8
	G − 8	1.502	44.6	27,500	29.7
	G − L	1.5	42.9	23,800	28.6
	G − 0	0.642	34.5	26,200	53.8
	ICRU		39.4		
5	Si	1.15	60.8	23,500	52.8
	G − 8	2.74	74.5	26,700	27.2

5 Bethe model of cross sections

Using the first Born approximation, Bethe (1930) derived a quantum mechanical expression for $\sigma(\epsilon)$. In its nonrelativistic form it can be written as the Coulomb cross section modified by a factor (called the "inelastic form factor"), which represents the probability of exciting the atomic electrons (Fano, 1963; Inokuti, 1971):

$$d\sigma(\epsilon, Q) = \rho(Q) \mid F(\epsilon, \mathbf{K}) \mid^2 d\epsilon dQ, \qquad (8)$$

where $\mathbf{K}$ is the momentum transferred from the incident particle to the absorber, $\mid F(\epsilon, \mathbf{K}) \mid$ the transition matrix element for the excitation, and $Q = (\hbar\mathbf{K})^2/2m$. Q is

the energy which would be given to a free electron with momentum K. For an atom with Z electrons, $\mid F(\epsilon, \mathbf{K}) \mid$ is defined for the transition from the ground state $\mid 0 >$ to an excited state $\mid n >$, with energy transfer ϵ and momentum change $\hbar\mathbf{K}$ (Inokuti, 1971) by:

$$F(\epsilon, \mathbf{K}) =< n \mid \sum_{j=1}^{Z} exp(i\mathbf{K} \cdot \mathbf{r_j}) \mid 0 > . \qquad (9)$$

If we assume that the atoms are oriented at random or that the ground state is spatially symmetric, $\mid F(\epsilon, \mathbf{K}) \mid^2$ is a function of the scalar variable K. The cross section differential in ϵ is obtained by integrating Eq. (8) over Q. In our current understanding, this approach to the calculation of $\sigma(\epsilon)$ is closest to reality.

Frequently, $F(\epsilon, K)$ is replaced by the generalized oscillator strength (GOS) $f(\epsilon, K)$, defined by

$$f(\epsilon, K) = \frac{\epsilon}{Q} \mid F(\epsilon, K) \mid^2, \quad \text{thus} \qquad (10)$$

$$d\sigma(\epsilon, Q) = \frac{k}{\beta^2} \frac{f(\epsilon, K)}{\epsilon} \frac{dQ}{Q} d\epsilon. \qquad (11)$$

Extensive studies have been made of the GOS for atoms with hydrogenic electron orbits (Bethe, 1930; Walske, 1952, 1956; Khandelwal, 1982; Bichsel, 1983), and these data should be used as a baseline for comparison with other calculations. A model of $f(\epsilon, K)$ for H-atoms is shown in Fig. 10 of Inokuti (1971). Further numerical calculations of GOS, Eq. (10) have been made with wave functions approximating real atoms more closely than the hydrogenic functions. For example, for aluminum and silicon, the ground state functions and the atomic potential were taken from the Herman-Skillman calculations, while for the continuum states, numerical integrations of the Schrödinger equation were performed (Manson, 1972; Bichsel, 1988). For silicon, for a few values of ϵ, results of these calculations are compared with hydrogenic calculations in Fig. 3. A detailed study of $\sigma(\epsilon, Q)$ for all shells of solid silicon has been made by Bichsel (1988), and $\sigma(\epsilon)$ calculated with Eq. (11) for 10 keV electrons is shown in Fig. 2. Moments are given in the Table. I believe that with the inclusion of the shell corrections, Eq. (21) below, these calculations should give adequate cross sections for energies as low as 1 keV. This has not been done so far, though. In order to get $\sigma(\epsilon)$, it is necessary to calculate the complete functions for the generalized oscillator strength. Problems related to the valence electrons (Bichsel, 1988) have been solved with the methods proposed by Ritchie, who will discuss them at this conference. I estimate that for $E > 10keV$ the calculation of $\sigma(\epsilon)$ for Si with Eq. (11) is accurate to $\pm 5\%$ or better for most ϵ, while the moments are accurate to about $\pm 1\%$ (Bichsel, 1988).

Bethe derivation for the first moment

In order to get the moments M_ν, Eq. (2), from $d\sigma(\epsilon, K)$ of Eq. (11), it is necessary to perform double integrals for each particle speed (e.g. Walske, 1956). Bethe showed that for large particle speeds it is not necessary to know GOS in order to get the stopping power, M_1. Instead, sum rules can be used. Bethe et al. (1950) noted that the GOS, $f(\epsilon, K)$, is constant near $K = 0$ (Fig. 3), and they used the following clever approach to simplify the calculation. With Eq. (11), the integral over Q, used to obtain the singly differential cross section $\sigma(\epsilon)$, is divided into three parts:

$$\begin{aligned} \frac{\epsilon\beta^2}{k}\sigma(\epsilon) &= \int_{Q_m}^{\infty} f(\epsilon, K)\frac{dQ}{Q} = \int_{Q_m}^{Q_1} f(\epsilon, 0)\frac{dQ}{Q} \\ &+ \int_{Q_1}^{\infty} f(\epsilon, K)\frac{dQ}{Q} \\ &+ \int_{Q_m}^{Q_1} [f(\epsilon, K) - f(\epsilon, 0)]\frac{dQ}{Q} \end{aligned} \qquad (12)$$

with $Q_m \approx \epsilon^2/2mv^2$. Q_1 is chosen such that the GOS at K_1 differs little from that at $K = 0$ (Fig. 3). Frequently, the collisions with $Q < Q_1$ are referred to as "distant", those with $Q > Q_1$ as "close".

The first integral can be evaluated directly, and is equal to:

$$f(\epsilon, 0) \cdot \ln(Q_1/Q_m) = f(\epsilon, 0) \cdot \ln(Q_1 \cdot 2mv^2/\epsilon^2) \qquad (13)$$

and contains the particle speed, v, explicitly. The contribution to M_1 from the distant collisions, i.e. the integral over ϵ of Eq. (13) therefore is

$$\begin{aligned} {}_dM_1 &= \frac{k}{\beta^2} \int f(\epsilon, 0) \ln(Q_1 \cdot 2mv^2/\epsilon^2) d\epsilon \\ &= \frac{k}{\beta^2} \ln(Q_1 \cdot 2mv^2) \int f(\epsilon, 0) d\epsilon \\ &- 2\frac{k}{\beta^2} \int f(\epsilon, 0) \ln \epsilon d\epsilon. \end{aligned} \qquad (14)$$

For the integral over $f(\epsilon, 0)$, the sum rule for DOS provides the value Z (see Eq. (17)), and the last integral defines the average logarithmic excitation energy, I:

$$Z \ln I \equiv \int f(\epsilon, 0) \ln \epsilon d\epsilon. \qquad (15)$$

Therefore we can write

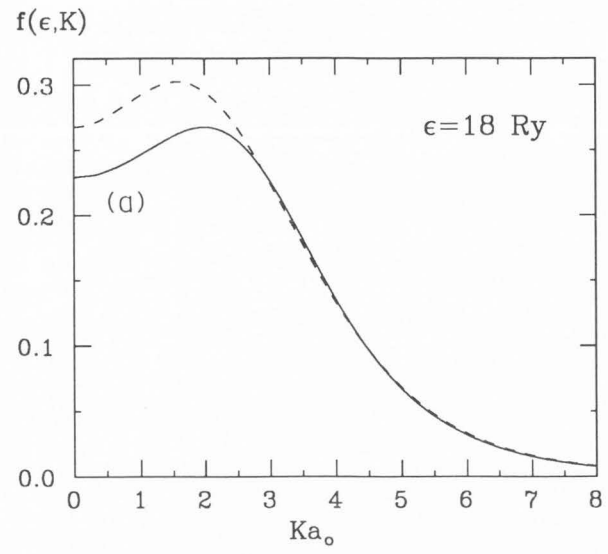
$${}_dM_1 = \frac{k}{\beta^2}[Z \ln(Q_1 \cdot 2mv^2) - 2Z \ln I] = \frac{k}{\beta^2} Z \ln \frac{Q_1 \cdot 2mv^2}{I^2}. \qquad (16)$$

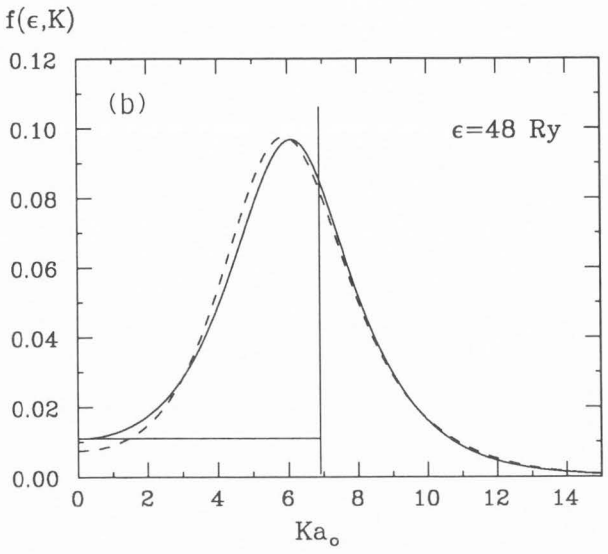
The contribution to M_1 from close collisions is given by the integral over ϵ of the second integral in Eq. (12), it is independent of particle speed. The sumrule

$$\int_0^{\infty} f(\epsilon, Q) d\epsilon = Z \quad \text{(Bethe, 1930; Inokuti, 1971)} \qquad (17)$$

simplifies the calculation of this integral greatly, we can change the order of integration and get:

$$\begin{aligned} \int_0^{\infty} d\epsilon \int_{Q_1}^{\infty} f(\epsilon, Q)\frac{dQ}{Q} &= \int_{Q_1}^{Q_M} \frac{dQ}{Q} \int_0^{\infty} f(\epsilon, Q) d\epsilon \\ &= Z \int_{Q_1}^{Q_M} \frac{dQ}{Q} = Z \ln \frac{Q_M}{Q_1} \end{aligned} \qquad (18)$$





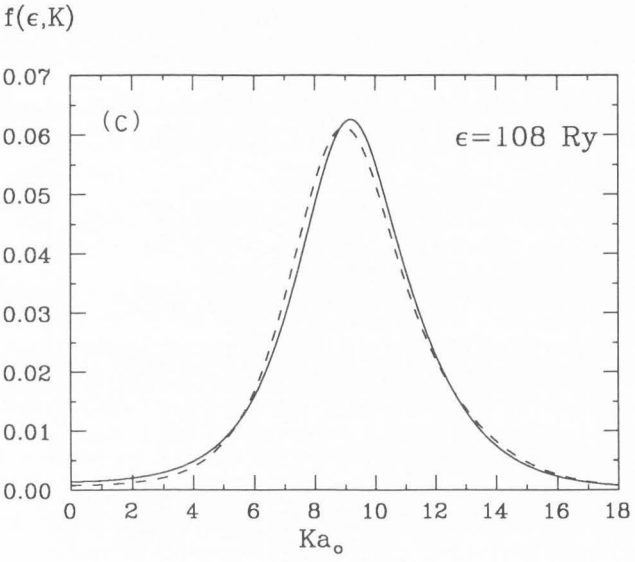


Fig. 3. Generalized oscillator strength (GOS) functions $f(\epsilon, K)$(in Ry^{-1}) for longitudinal excitations of the 2p-shell of silicon atoms (with a binding energy $B_L = 8Ry$). The abscissa represents the momentum transfer K (in atomic units) occurring in the collision. For comparison, the function calculated with the hydrogenic approximation is also given (broken line). Functions are given for $\epsilon = 18Ry$, Fig. 3a; $\epsilon = 48Ry$, Fig. 3b; and $\epsilon = 108Ry$, Fig. 3c. If $K_1 a_0$ [corresponding to Q_1 in Eq. (12)] is chosen to be equal to 1, the third integral of Eq. (12) will be very small for $\epsilon = 108Ry$, but larger for smaller ϵ. Note that the maxima of $f(\epsilon, K)$ are located at $(Ka_0)^2 \approx \epsilon - B_L$. In the DOS model, Eq. (23), the GOS is replaced by a δ-function at $Ka_0 = \epsilon^{1/2}$ and by DOS for $0 < Ka_0 < \epsilon^{1/2}$. This model is shown in Fig. 3b (the δ-function is shown schematically).

where we had to replace the upper limit ∞ of the integral over Q by $Q_M = \epsilon_M$, the maximum energy loss; for electrons, $\epsilon_M = E/2$, for heavier ions, $\epsilon_M = 2mv^2$ (further details in Fano, 1963). Thus the contribution ${}_cM_1$ from close collisions is:

$$ {}_cM_1 = \frac{k}{\beta^2} Z \cdot \ln(Q_M/Q_1). \qquad (19) $$

In the third integral of Eq. (12), for small Q_m, the lower limit can be replaced by zero because $f(\epsilon, K)$ differs little from $f(\epsilon, 0)$ (see Fig. 3; Khandelwal, 1982; Peek, 1983). With the change in the order of integration, the integral over ϵ is zero. Then the total stopping power is

$$ M_1 = {}_d M_1 + {}_c M_1 = \frac{k}{\beta^2} Z \ln \frac{Q_M \cdot 2mv^2}{I^2} \qquad (20) $$

which is the well known asymptotic Bethe result. It must be understood clearly that the approximation made in the derivation of this equation is that the integral

$$ \int_0^{Q_M} [f(\epsilon, K) - f(\epsilon, 0)] \frac{dQ}{Q} \qquad (21) $$

added to Eq. (12) (by replacing Q_m by zero, see below Eq. (19)) is negligibly small. Whenever this is not the case, M_1 of Eq. (20) will be too large, and the contribution from Eq. (21) produces a reduction of M_1, known as the "shell correction". A further correction term related to the behaviour of the function in the neighbourhood of $Q = \epsilon_M$ (see Eq. (18)) was discussed by Fano (1963), and is also considered to be part of the shell corrections.

General structure of the equation for stopping power, M_1

Fano (1963) showed that the shell corrections are proportional to v^{-2}, in first order. For Si and lighter elements, this approximation should give reliable values of

M_1 for $E > 3keV$, while for $1keV < E < 3keV$, corrections of the order of 10 % must be expected. Thus if a parameter fit to experimental stopping power data is attempted, a function with the structure including the major dependence on v, derived from Eq. (20), should be used, i. e.

$$M_1 \approx (g/v^2)[\ln(v^2/h) - j/v^2] \qquad (22)$$

where g, h and j are parameters which are adjustable within reasonable limits. In particular, g, which for large particle speeds is proportional to the atomic number Z, can be chosen to include a factor $Z_{eff} < Z$ in consideration of the fact that, for smaller speeds, some of the atomic electrons do not contribute to the energy loss. For gold, for example, the K- and L-shell electrons only contribute to the energy loss for incident electrons with energies exceeding about 15 keV (or protons with energies above 25 MeV), and one could choose $Z_{eff} = 69$ in calculating g. Correspondingly, h would be reduced from the Bethe value, and the approximation with v^{-2} for the shell corrections would be appropriate to lower speeds.

Next we consider an approximate model for $\sigma(\epsilon)$ which does not require the calculation of the GOS.

Model approximating the Bethe ridge with a delta function

Allison and Cobb (1980) and Liljequist (1983) approximated the GOS of Fig. 3 by placing a delta function at $Q = \epsilon$ and by replacing the GOS $f(\epsilon, Q)$ for $Q < \epsilon$ by the dipole oscillator strength $f(\epsilon, 0)$ (see Fig. 3b). Then the integral over Q of Eqs. (11) and (12) extends from Q_m to ϵ rather than to Q_1. Sum rules were used to obtain the residual contribution for the delta functions. The collision cross section then is given by (for $\epsilon < \epsilon_M$):

$$\sigma(\epsilon) = \rho(\epsilon)[\epsilon f(\epsilon, 0)\ln(2mv^2/\epsilon) + \int_0^{\epsilon} f(\epsilon', 0)d\epsilon'] \qquad (23)$$

(Allison and Cobb, 1980; relativistic terms are omitted); for $\epsilon > \epsilon_M$, $\sigma(\epsilon) = 0$. For large ϵ, $f(\epsilon, 0) \approx \epsilon^{-3.5}$, thus the first term in the bracket decreases rapidly with increasing ϵ, the integral $\rightarrow Z$, and we get $\sigma(\epsilon) \approx Z\rho(\epsilon)$. This of course is only true for $\epsilon \gg B_K$. This model has the advantage that it is only necessary to determine the DOS $f(\epsilon, 0)$ for the absorber, or, equivalently, the imaginary part of the inverse of the complex dielectric function, $Im(-1/\kappa)$. Data for κ can be extracted from a variety of optical measurements. In particular, $Im(-1/\kappa)$ can be obtained from electron energy loss measurements (Raether, 1980). The cross section calculated with this model is given in Fig. 2. It differs by as much as 50% from the Bethe result, but the moments differ by at most 10% (Table). It gives a closer approximation to the Bethe model than does the Gryzinski approximation.

6 Conclusions and recommendations

The choice of an appropriate differential collision cross section for applications in transport theory is of great importance. Especially for Monte Carlo calculations, the structure of the cross section for the valence electrons is relevant because it influences the moment M_0 and therefore the number of collisions per unit pathlength greatly. This is evident from the comparisons shown in Fig. 2 and in the Table. The most reliable method to obtain theoretical $\sigma(\epsilon)$ is that described by Eq. (11). If this approach is too time consuming, the approximation defined in Eq. (23) will give much better results than the Gryzinski approach, Eq. (7). Clearly, accurate data for DOS are important in this construction of $\sigma(\epsilon)$. While the parameters in Eq. (7) can easily be adjusted to give the correct stopping power, this is not readily done for the differential collision cross section. No attempt has been made here to achieve this. In general, it appears advisable to first evaluate any proposed theoretical or semi empirical $\sigma(\epsilon)$ for 10 keV electrons in Si and compare it to Fig. 2 before calculations are made for other substances.

It clearly is desirable to perform transport calculations with several models of $\sigma(\epsilon)$ in order to assess the sensitivity of the results to the properties of the model (e.g. Valkealahti and Nieminen 1983; Bichsel, 1988).

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Discussion with Reviewers

J. Schou: How is the agreement between the stopping power that you calculated, and that from Tung, Ashley and Ritchie (e.g. Surf. Science **81**, 427 (1979)?

Bichsel: I am comparing these results with my data in this table:

E(keV)	$M_1(MeV/cm)$		M_0(collisions/μm)	
	Tung	*Bichsel*	*Tung*	*Bichsel*
1	198	160	4.69	4.18
2.5	110	96	2.24	2.04
5	66.6	60.9	1.26	1.15
10	39.2	37.5	0.69	0.646

It must be noted that Tung et al. used the Lindhard statistical model for their calculations. My calculation for protons with energies above 5 MeV agree with the Janni (1982) data to better than 1%.

Schou: Is it straightforward to extend your treatment to other elements?

Bichsel: To achieve reasonably accurate results (5-10% in $\sigma(\epsilon)$, 2% in M_0 and M_1), a complete evaluation of the GOS of Eq. (10) must be made. This work is straightforward but time consuming. A detailed description of such an evaluation for silicon is given in Bichsel (1988).

As a first step, the DOS should be determined accurately. The major problem will be to find the function for the valence electrons. It could be obtained from absorption measurements with synchrotron radiation or from electron spectroscopy experiments. Then the approximation described by Eq. (23) may be used to get preliminary results for $\sigma(\epsilon)$, with uncertainties of up to 50% in $\sigma(\epsilon)$, but maybe 5% in M_0 and M_1.

R. Bonham: Eq. (6) certainly is correct for an electron scattered by atomic hydrogen. For electron scattering by He or a hydrogenic atom excited to a quartet state I am not so sure. For elastic scattering from He the cross term may have to be multiplied by 2? The expression is correct as written if one assumes the factorization approximation which assumes that the cross-section can be written as a product of a form factor and the electron-electron scattering cross section. If, on the other hand, one starts

from the first Born approximation, including exchange, and works out all the spin couplings, say for an L-S coupling model for atomic scattering, to obtain the correct expression for the cross section and then uses a high energy approximation to relate the Born-Oppenheimer exchange matrix element to the matrix element for direct scattering, I am not so sure the answer will be the same. I have been trying to do this in a general way but have run afoul of horrendous angular momentum algebra.

Bichsel: Eq. (6) is meant to demonstrate the general nature of the exchange effect; it is valid for the scattering of electrons on free electrons. The problems you mention must certainly be considered for any of the more complex situations. I do not know how serious problems would be for solids as compared to atoms.

Bonham: The comment below Eq. (6) "(e.g. at E = 10 or 20 keV)" is a good one, but the suggested energies may have to be higher; see Phys. Rev. A **38**, 654 (1988). I am personally a little suspicious of this work as my preconceived idea was that the experiment should approach the theory at a lower energy or at least show signs of moving in that direction by 5 keV. Of course we don't know whether the problem is due to the elastic or the inelastic part. I suspect that most of the problem is in the elastic part.

Bichsel: My major concern is with the validity of different theoretical models. What I am suggesting is that the structure of a model should show the behaviour of Eqs. (5) and (6) for the appropriate values of energies and the parameters. For example, it must be noted that Eq. (7) does not show the exchange effects of Eq. (6)! As you point out, in a comparison of experiment and theory, it will be necessary to explore carefully which aspects of either could be faulty.

Bonham: About benzene. I don't believe any plasma or collective excitations in free molecules have ever been documented although there have been papers predicting such effects. The author should make it clear whether or not the prediction that the feature in gaseous benzene at 16 eV is his own prediction or someone elses. This is a very interesting observation and our next experiment will be to study the momentum transfer dependence of this feature to determine whether or not it is actually a collective excitation.

Bichsel: I am judging by appearances. Readers should study Bonham's suggestions in given applications and let us know their conclusions. This raises the question how big the molecule is in the two phases. Maybe this could be determined from the Van-de-Waals terms.

Bonham: Wouldn't a particle in a box model suggest greater delocalization in the solid than in the gas and suggest that the collective excitation should lie at lower energy loss rather than higher in the solid case?

Bichsel: One might argue that more electrons would be involved in the collective motion in the solid than in the gas, therefore more energy would be needed for the excitation.

P. J. Schultz: Powell has done extensive comparisons of inelastic cross sections (e.g. Rev. Mod. Phys. **48**, 33 (1976)). Please comment on the conclusions and degree to which they do or do not agree.

Bichsel: Powell's work certainly must be studied and used if cross sections for various substances are to be determined. I do not believe that Powell made measurements for Si, therefore a direct comparison with my data is not possible. In the ref. mentioned above, Powell was discussing total collision cross sections (i.e. M_0) for inner shell excitations. He did find good agreement with the Bethe theory (without shell corrections) with parameters fitted to experiments for electron energies greater than $4 \cdot B_i$. His procedure is essentially the same as the one I used for Si, thus we agree on the procedure, and I would expect agrement on the data.