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ELASTIC SCATTERING OF ELECTRONS BY ATOMS

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Abstract

Elastic scattering is defined as scattering in which the incident particle or radiation does not give up any of its energy to the scatterer. Electrons are elastically scattered in atoms by both the nucleus and the atomic electrons which screen the nuclear charge. When considering only nuclear scattering the first Born approximation quantum mechanical cross section and the classical Rutherford cross section are identical. The effects of the atomic electrons can be taken into account by a simple screening term or by more exact treatments based on Hartree-Fock or Dirac-Fock wave functions. The partial wave expansion can be used to go beyond the first Born approximation but it is only really appropriate for isolated atoms. All elastic scattering cross sections at energies of interest in electron microscopy are strongly forward peaked. Mott cross sections calculated by solving the radial Dirac equation incorporate more than just the relativistic changes in mass and wave vector and so cannot be compared directly to Rutherford cross sections.

Introduction

Elastic scattering is defined as scattering in which the incident particle gives up no energy to the scattering particle. Strictly the scattering should be referred to the centre of mass frame, but as the mass of the lightest atom is a few thousand times the mass of an electron, it is not usual to consider the recoil of the scattering particle.

The electron is a charged particle and both the positively charged nucleus and the negatively charged atomic electron cloud influence the scattering distribution. The scattering is described by the cross section which has units of area. It is derived by considering the probability, $I(\theta)$, of a particle being observed at a particular angle, θ , after scattering through a slab of thickness t with N atoms/unit volume.

$$I(\theta) = N\sigma(\theta)t \quad 1 \text{ (a)}$$

$$\sigma_T = \int_0^\pi \sigma(\theta) 2\pi \sin\theta d\theta \quad 1 \text{ (b)}$$

The cross section shown here is a differential cross section and assumes a spherically symmetric potential. To obtain the total probability of elastic scattering, it is necessary to integrate over all scattering angles. The use of a cross section presupposes a classical or semi-classical description in that it only relates intensities. Any wave properties of the electron are explicitly excluded though they may have been used to derive the cross section.

It is easiest to treat the scattering from only the nucleus. The electron experiences an electrostatic potential given by

$$\frac{Ze^2}{4\pi\epsilon_0 r} \quad (2)$$

where Z is the atomic number, e is the electronic charge and r is the distance of the electron from the nucleus.

Classically by conservation of energy and momentum, the cross section is

$$\sigma(\theta) = \left(\frac{Ze^2}{4\pi\epsilon_0}\right)^2 \frac{1}{4E^2 \sin^4(\theta/2)} \quad (3)$$

where E is the electron energy. This cross section is called the Rutherford cross section. It diverges at small scattering angles because the

KEY WORDS: Elastic scattering, Rutherford cross section, Mott cross section, Born approximation, Lippman-Schwinger equation, electron scattering factor, Dirac equation, partial wave expansion, perturbation theory, Fermi-Thomas model.

electron can experience the nuclear potential no matter how far it is from the nucleus.

List of Symbols

$\sigma(\theta)$	= differential scattering cross section (m^2)
σ_T	= total scattering cross section (m^2)
t	= specimen thickness (m)
N	= number of atoms/unit volume (m^{-3})
Z	= atomic number
E	= electron energy (J)
e	= electronic charge (C)
ϵ_0	= permittivity of free space ($\text{C}^2\text{J}^{-1}\text{m}^{-1}$)
h	= Plank's constant (Js)
m	= electron mass (kg)
$\underline{r}, \underline{r}'$	= electron coordinates (m)
\underline{c}	= speed of light (ms^{-1})
$\chi(\underline{r})$	= total wave function
$u(\underline{r})$	= initial wave function
$v(\underline{r})$	= scattered wave function
$f(\theta)$	= electron scattering factor (m)
H	= Hamiltonian (J)
$V(\underline{r})$	= potential (J)
$G(\underline{r}, \underline{r}')$	= scattering Greens function ($\text{J}^{-1}\text{m}^{-3}$)
\underline{K}_f	= final state wave vector (m^{-1})
\underline{K}_i	= initial state wave vector (m^{-1})
\underline{K}_z	= z component of wave vector (m^{-1})
K	= magnitude of wave vector (m^{-1})
q	= scattering wave vector (m^{-1})
v	= electron velocity (ms^{-1})
a	= typical atomic dimension (m)
$P_\ell(\cos\theta)$	= Legendre polynomial
$j_\ell(kr)$, $n_\ell(kr)$	= Bessel functions
δ_ℓ	= scattering phase shift
ψ	= wave function
ψ_ℓ	= solution of radial Shrödinger equa.
u	= screening constant (m^{-1})
$\rho(\underline{r})$	= electron density (m^{-3})
$f_x(q)$	= x-ray scattering factor
A_j, B_j	= parameters
$g(\theta)$	= solution of Dirac equation
p	= momentum operator (kgms^{-1})
σ	= spinor
$I(\theta)$	= probability
r	= distance of electron from nucleus
β	= v/c
α	= $Z/137$

The quantum mechanical treatment will also give the same expression for the cross section. The theory will be given for a completely general potential which includes the effects of the atomic electrons. The incident wave function will be denoted by $u(\underline{r})$ and the wave function after scattering will be denoted by

$$\chi(\underline{r}) = u(\underline{r}) + v(\underline{r}) \quad (4)$$

The scattered part of the wave function $v(\underline{r})$ will eventually become $f(\theta)\exp(i\mathbf{K}\cdot\mathbf{r})/r$, which is a spherical outgoing wave multiplied by $f(\theta)$, the electron scattering factor.

The incident wave function $u(\underline{r})$ is a solution of the Shrödinger equation for an electron of energy E in free space:

$$H u = E u \quad (5)$$

The wave function after scattering has to satisfy the Shrödinger equation with the scattering potential.

$$(H + V(\underline{r}))\chi(\underline{r}) = E\chi(\underline{r}) \quad (6)$$

Making use of equation 5 and rearranging gives

$$(H - E)v(\underline{r}) = V(\underline{r})\chi(\underline{r}) \quad (7)$$

which can be solved by using the Greens function $G(\underline{r}, \underline{r}')$, which is a solution of the homogeneous equation

$$(H - E)G(\underline{r}, \underline{r}') = 0 \quad (8a)$$

$$G(\underline{r}, \underline{r}') = \frac{-m}{2\pi\hbar^2} \frac{\exp(i\mathbf{K}(\underline{r} - \underline{r}'))}{|\underline{r} - \underline{r}'|} \quad (8b)$$

The wave function after scattering then becomes

$$\chi(\underline{r}) = u(\underline{r}) + \int G(\underline{r}, \underline{r}')V(\underline{r}')\chi(\underline{r}')d^3r' \quad (9)$$

(see Schiff, 1955)

This integral equation, which must be solved iteratively for an exact solution, is called the Lippman-Schwinger equation.

It is possible to derive an approximate solution by making the following simplifications. The solution is only required at a point \underline{r} far from the scattering centre and $r \gg r'$ so the Greens function becomes

$$\frac{\exp(i\mathbf{K}_f \cdot \underline{r})}{r} \exp(-i\mathbf{K}_f \cdot \underline{r}') \quad (10)$$

where \mathbf{K}_f is the wave vector of the outgoing spherical wave. The incident wave function $u(\underline{r})$ is a plane wave of wave vector \mathbf{K}_i and it will be assumed that the scattering is sufficiently weak so that $\chi(\underline{r})$ is not much different from $u(\underline{r})$. This weak scattering approximation is the first Born approximation. The outgoing spherical wave is then

$$\begin{aligned} v(\underline{r}) &= \frac{\exp(i\mathbf{K}_f \cdot \underline{r})}{r} f(\theta) \\ &= \frac{-m}{2\pi\hbar^2} \frac{\exp(i\mathbf{K}_f \cdot \underline{r})}{r} \int \exp(-i\mathbf{K}_f \cdot \underline{r}') V(\underline{r}') \\ &\quad \exp(i\mathbf{K}_i \cdot \underline{r}') d^3r' \end{aligned} \quad (11)$$

If the scattering only comes from the nucleus, the potential is given by equation 2 and the electron scattering factor becomes

$$f(\theta) = \left(\frac{Ze^2}{4\pi\epsilon_0} \right) \frac{1}{2E\sin^2(\theta/2)} \quad (12)$$

making use of the relation

$$|\mathbf{K}_i - \mathbf{K}_f| = q = \left(\frac{2mE}{\hbar^2} \right)^{1/2} 2\sin(\theta/2) \quad (13)$$

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The differential cross section is the modulus squared of the scattering factor and is again given by equation 3.

Note that the scattering factor in the first Born approximation is real.

The first Born approximation is only valid when the scattering potential is weak, i.e.

$$v(0) \ll 1$$

The condition for the validity of the first Born approximation can be expressed as

$$Z \left(\frac{e^2}{4\pi\epsilon_0 \hbar c} \right) \left(\frac{c}{v} \right) \log e(Ka) \ll 1 \sim \frac{Z(c)}{137(v)} \ll 1 \quad (14)$$

(See Schiff, 1955)

This is clearly acceptable for light elements for electron energies of 30 kV but not for heavy elements.

The Lippman-Schwinger scattering equation can be solved exactly by means of partial waves. The incident wave is defined to lie along the z direction and the wave function after scattering $\chi(\underline{r})$

$$\chi(\underline{r}) = \exp(iK_z \cdot \underline{r}) + \frac{f(\theta)}{r} \exp(iK \cdot \underline{r}) \quad (15)$$

can be expanded as a partial wave series if the potential is spherically symmetric.

$$\chi(\underline{r}) = \sum_{\ell} (2\ell + 1) i^{\ell} \frac{\psi_{\ell}(r)}{r} P_{\ell}(\cos\theta) \quad (16)$$

where $\psi_{\ell}(r)$ are solutions of the radial Schrödinger equation and $P_{\ell}(\cos\theta)$ are Legendre polynomials.

The solutions to the radial Schrödinger equation can be expressed in terms of the spherical Bessel functions $j_{\ell}(kr)$ and $\eta_{\ell}(kr)$ with an appropriate phase shift

$$\frac{\psi_{\ell}(r)}{r} = \exp(i\delta_{\ell}) (\cos\delta_{\ell} j_{\ell}(kr) - \sin\delta_{\ell} \eta_{\ell}(kr)) \quad (17)$$

Far from the scattering centre this becomes

$$\frac{\psi_{\ell}(r)}{r} = \exp(i\delta_{\ell}) \frac{\sin(kr - \frac{1}{2}\ell\pi + \delta_{\ell})}{kr} \quad (18)$$

which is an outgoing spherical wave with a phase shift. The electron scattering factor is

$$f(\theta) = \frac{1}{2K} \sum_{\ell} (2\ell + 1) (\exp(2i\delta_{\ell}) - 1) P_{\ell}(\cos\theta) \quad (19)$$

and the differential cross section is

$$\sigma(\theta) = \frac{1}{K^2} \left| \sum_{\ell} (2\ell + 1) \exp(i\delta_{\ell}) \sin\delta_{\ell} P_{\ell}(\cos\theta) \right|^2 \quad (20)$$

To calculate the cross section in the general case by this method, it is necessary to solve the radial Schrödinger equation numerically and derive the phase shifts by matching at an arbitrary boundary. The higher the electron energy, the greater the number of phase shifts that are needed. The maxi-

mum value of ℓ should be of the order ka where a is an atomic distance.

It is possible to see the relationship between this theory and the first Born approximation by substituting the appropriate partial wave expansions for $u(\underline{r})$ and $\chi(\underline{r})$ in equation 9.

The exact scattering factor is

$$f(\theta) = \frac{-2m}{\hbar^2} \sum_{\ell} (2\ell + 1) P_{\ell}(\cos\theta) \int V(r) j_{\ell}(kr) \frac{\psi_{\ell}(r)}{r} r^2 dr \quad (21)$$

In the first Born approximation $\chi(\underline{r})$ is also a plane wave and

$$f(\theta) = \frac{-2m}{\hbar^2} \sum_{\ell} (2\ell + 1) P_{\ell}(\cos\theta) \int V(r) j_{\ell}^2(kr) r^2 dr \quad (22)$$

The condition for the first Born approximation implies that the scattering phase shifts must be small and the Born approximation phase shift can be identified as

$$\delta_{\ell} = \frac{-2mK}{\hbar^2} \int V(r) j_{\ell}^2(kr) r^2 dr \quad (23)$$

To understand the physical meaning of the quantum mechanical solution, it is necessary to go back to the Lippman-Schwinger equation.

The solution can be formally represented as

$$\chi = u + GV\chi \quad (24)$$

where χ and u are vectors and GV is an operator. The first Born approximation corresponds to

$$\chi = u + GVu \quad (25)$$

The partial wave solution corresponds to the full series expansion

$$\chi = u + GVu + GVG Vu + GVG VG Vu + \text{etc.} \quad (26)$$

which can be visualised as multiple scattering from an isolated atom (i.e. an atom in a gas). The question then arises as to whether going beyond the first Born approximation is justified in a solid as the various terms in the series should consider scattering by atoms on different sites. This is exactly the problem of scattering in a crystal which can be solved using the dynamical theory of electron diffraction. In this theory, the wave function is expressed as a Fourier series (sum of the different Bragg beams) and the Schrödinger equation becomes equivalent to a series of first order coupled differential equations in which the terms coupling one beam to another are proportional to the Fourier coefficients of the crystal potential. (See Hirsch et al., 1963.) Formally these coefficients are equivalent to the first Born approximation (see equation 11), and this has caused some confusion.

Howie and Whelan (1961) derived the dynamical equations using a wave optical theory involving the electron scattering factors. It was then suggested that scattering factors in the first Born approximation were not accurate enough and that higher order terms should be included in the Born series expansion. As can be seen from equation

19, this made the scattering factor complex which leads to an "absorption" from the elastic wave field. This is obviously incorrect and the problem arose because of a confusion between the first Born and the Fourier coefficients of scattering potential, which happen to be identical. (The argument given above strictly only applies to primitive crystal lattices with one atom/unit cell. When the unit cell contains more than one atom, the Fourier coefficients of the potential are related to the structure factor, which can be Hermitian in the general case.) In fact Fujiwara (1959, 1961) showed that for electron scattering in a crystal, a summation of the Born series including all possible scattering events between atoms on different sites leads to a result identical to the dynamical theory in which the coupling coefficients appear to be related to those given by the first Born approximation. An amorphous material could be treated in exactly the same way as a crystal using a Fourier transform instead of a Fourier series and then summing over the ensemble of atomic configurations to get the average scattering. The partial wave expansion recognises the failure of treating an electron as a particle, but is only appropriate for isolated atoms. The correct procedure would be to solve the Schrödinger (or Dirac) equation for the solid so that the coherent nature of electron scattering is explicitly taken into account.

So far the detailed treatment of electron scattering has not included the scattering by atomic electrons. As a first approximation, the atomic electrons screen the nuclear charge, making the effective potential

$$V(r) = \frac{Ze^2}{4\pi\epsilon_0 r} \exp(-\mu r) \quad (27)$$

The differential cross section in the first Born approximation becomes

$$\sigma(\theta) = \left(\frac{Ze^2}{4\pi\epsilon_0} \right)^2 \left(\frac{m}{\hbar} \right)^2 \frac{1}{(q^2 + \mu^2)^2} \quad (28)$$

Note that this does not diverge for small scattering angles. The parameter μ is to some extent arbitrary, but can be derived from a Fermi-Thomas model of the atom. This model assumes that the maximum kinetic energy at a position r is $-eV(r)$, which can be used to derive the number density of electrons. The number density is also related to the potential by Poisson's equation and a differential equation for the electron density can be derived. The result is that the screening distance $1/\mu$ is of the order $a_0/Z^{1/3}$ where a_0 is the Bohr radius. It is not difficult to solve for the atomic scattering factors using an exact electron density calculated from a Hartree-Fock procedure. If the electron density is $\rho(r)$ the scattering potential is

$$\frac{e^2}{4\pi\epsilon_0 r} (Z - \rho(r)) \quad (29)$$

and the electron scattering factor becomes

$$f(q) = \left(\frac{e^2 m}{4\pi\epsilon_0 \hbar} \right) \frac{(Z - f_x(q))}{q^2} \quad (30)$$

where $f_x(q)$ is the x-ray scattering factor which is the Fourier transform of the charge density.

Both the electron scattering factor and the x-ray scattering factor have been parameterised by Doyle and Turner (1968) as

$$f(q) = \sum_{j=1}^4 A_j \exp(-B_j q^2) \quad (31)$$

There is no reason to believe that this parameterisation is superior to others that could have been derived. It is probably better to use the parameterised x-ray scattering factors in equation 30 rather than the parameterised electron scattering factors as then the electron scattering factor will have the right asymptotic form for large q . The electron density in crystals is slightly different from that in isolated atoms due to bonding, but this usually does not have much effect in electron scattering factors as only the outermost electrons are affected. The main effect is to make a change of no more than a few percent for scattering factors with q less than 0.5 \AA^{-1} .

The differential cross sections calculated using equations 30 and 31 have been plotted for scattering by 5 kV electrons from gold (Fig. 1). The main feature to note is that the cross section is dominated by the $1/\sin^4(\theta/2)$ term and the cross section falls rapidly with scattering angle. At 180° it is about 10^{-4} of its value at 10° . The cross section is therefore replotted (Fig. 2) on a log scale and compared to the cross section for 25 kV electrons. This, as expected, is even more strongly forward peaked than the 5 kV cross section.

The cross sections for scattering of 5 kV electrons for aluminum and gold divided by Z are plotted on a log scale as Fig. 3 and not surprisingly the aluminum cross section is more forward peaked. It is interesting to note that when the cross sections are normalized by Z they are roughly equal for small scattering angles.

Relativistic effects can be incorporated in the theory in a number of ways. The simplest is to relativistically correct the mass and velocity of the electrons in the classical or first Born approximation formula. To be exact in the framework of quantum mechanics it is necessary to solve the Dirac equation. Physically the Dirac equation takes into account electron spin and creation of electron-hole pairs as well as relativity. It is not necessary to consider the positive and negative energy states for energies below about 10 MeV and for unpolarized beams the spin effects are averaged out. The cross section can be expressed as

$$\sigma(\theta) = |f(\theta)|^2 + |g(\theta)|^2 \quad (32)$$

where $f(\theta)$ and $g(\theta)$ are derived from the radial Dirac equation. This cross section is called a Mott cross section, and partial wave methods such as those described for solving the radial Schrödinger equation can be used. To compare the Dirac equation with the Schrödinger equation, it is easiest to use the second order wave equation given by Mott (1929)

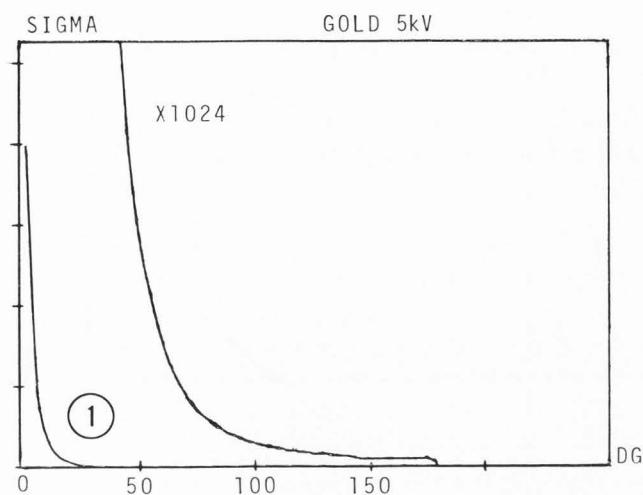


Fig. 1. Elastic scattering cross section for scattering of 5 kV electrons by gold.

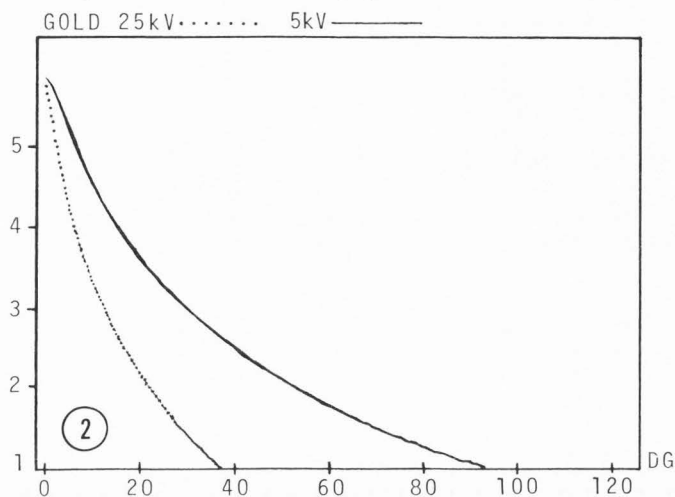


Fig. 2. Comparison of elastic scattering cross sections for 5 kV and 25 kV electrons for gold. (Log scale.)

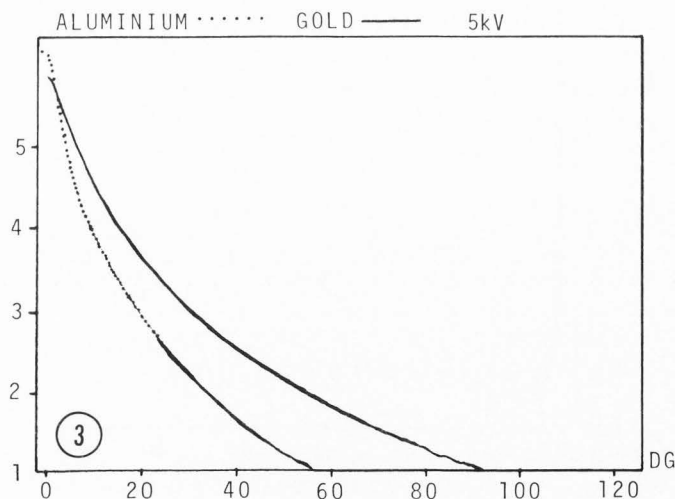


Fig. 3. Comparison of elastic scattering cross section divided by atomic number for 5 kV electrons for gold and aluminum. (Log scale.)

$$[\nabla^2 + 1 + \frac{2\alpha}{r\beta} + \frac{\alpha^2}{r^2} - i\alpha[P,(\sigma,r)]]\psi = 0 \quad (33)$$

where length is in units of $\hbar K$, $\beta = (v/c)$, and

$$\alpha = \frac{Ze^2}{4\pi\epsilon_0\hbar c} = \frac{Z}{137}$$

The first three terms are the same as those of a Shrödinger equation in which the wave vector and mass have been relativistically corrected. The last term is a spin term which is not important for unpolarized beams. The only other term is an effective addition to the potential of order $1/r^2$, which is $0.5\alpha\beta$ smaller than the main coulombic term. This term will tend to enhance the high scattering angle part of the cross section as it falls off as $1/(r^2)$.

This explains why the ratio of the Mott cross section to the relativistically corrected Rutherford cross section increases with scattering angle (see Kessler and Weichert (1968)). Comparisons between Mott and even relativistically corrected Rutherford cross sections can be misleading as the Mott cross section also includes the effects of atomic electrons and represents a partial wave solution rather than a first Born approximation. This is apparent in a comparison made by Reimer and Krefting (1976) and Shimizu et al. (1979), who used an unscreened and screened Rutherford cross section respectively (Figs. 4 and 5). For the energies used in scanning microscopy, it is hard to see why the extra term (which arises because the Dirac equation is a first order equation linear in the potential, not a second order equation like the Shrödinger equation) should have much effect even for heavy elements. It must also be remembered that any differences between Mott and Rutherford cross sections are very small compared to the overall behavior of the cross section as given in Figs. 1-3.

Another effect that is neglected in all calculations is the effect of exchange between the incident electrons and the atomic electrons. This can be important for very low energies of order 100 eV and methods for dealing with this problem are discussed by Pendry (1974) in his treatment of low energy electron diffraction (LEED).

In conclusion, the elastic scattering of electrons by the nucleus has been described by the Rutherford formula both according to classical mechanics and quantum mechanics in the first Born approximation. The cross section is strongly forward peaked at all energies of interest in scanning electron microscopy. Going beyond the first Born approximation by using a full partial wave solution for the atomic scattering potential is only really justified for an isolated atom. The appropriate procedure for a solid is to solve the relevant wave equation with the potential appropriate for that condensed phase. The divergence in the Rutherford cross section can be removed by considering the scattering by atomic electrons. This can be done by incorporating their charge density in the potential or by assuming they screen the nuclear charge in a simple fashion.

Solving the Dirac equation is equivalent to relativistically correcting the Shrödinger equation and adding on another term which changes the

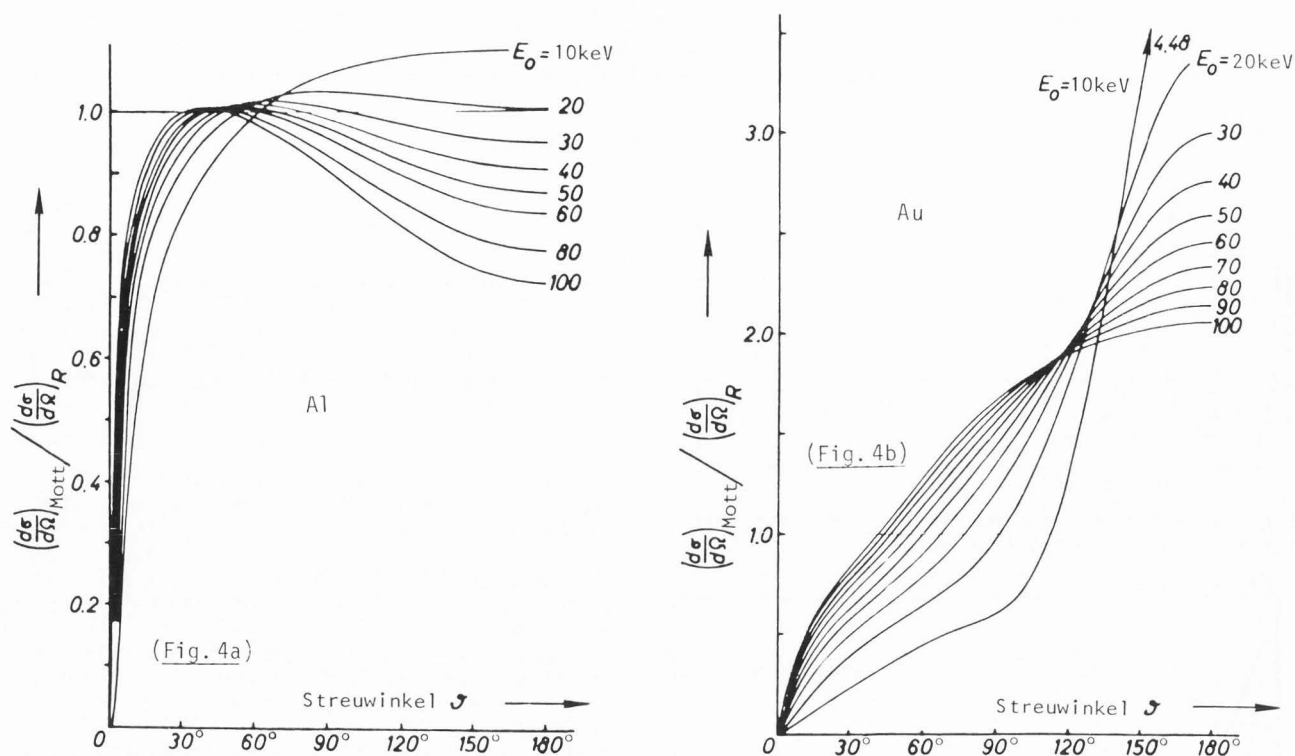


Fig. 4. Ratio of Mott cross sections to Rutherford cross sections for a) aluminum, b) gold from Reimer and Krefting (1976).

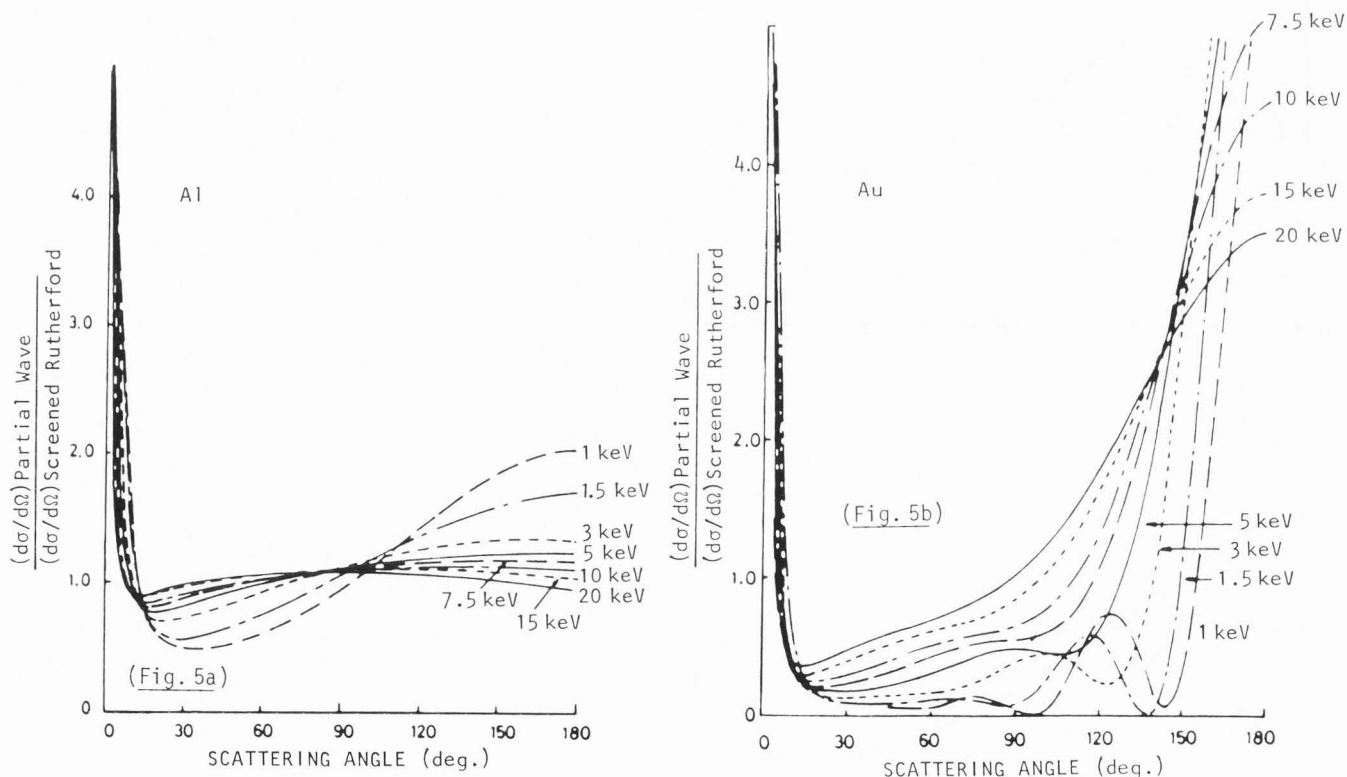


Fig. 5. Ratio of Mott cross sections to Rutherford cross sections for a) aluminum, b) gold from Shimizu et al. (1979).

angular distribution. Mott cross sections calculated by solving the radial Dirac equation incorporate the relativistic changes to mass and wave vector, the extra relativistic term in the wave equation, the effects of the atomic electron and the full partial wave solution to the scattering problem. It is therefore not really fair to compare them to screened or unscreened Rutherford cross sections as it cannot be ascertained which of the additional effects cause any observed difference.

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