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INELASTIC AND ELASTIC MULTIPLE SCATTERING OF FAST ELECTRONS DESCRIBED BY THE TRANSPORT EQUATION

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ABSTRACT

A method for solving the transport equation for the propagation of electrons in the primary energy range of interest in electron beam technology has been developed which is based on discretizing the related integral equation. The integral equation is solved by a collocation procedure yielding a system of linear equations.

The elementary scattering processes were described for elastic scattering by quantum mechanical differential cross sections and for inelastic scattering by Gryzinski type semi-empirical excitation functions for core and outer electrons separately.

From the electron flux density calculated, angular and energy distributions of transmitted and backscattered electrons were derived for various elements (Al, Cu, Ag, Au) and film thicknesses. The results agree with experimental data, including finer details as e.g. the dependence of the elastic backscattering peak on scattering angle and atomic number.

Keywords: Transport equation, elastic electron scattering, inelastic electron scattering, electron multiple scattering, electron matter interaction, electron energy deposition, electron backscattering, energy distribution of backscattered electrons.

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INTRODUCTION

The scattering of keV-electrons in films of solid matter is of great practical importance in electron microscopy, electron probe microanalysis, and electron beam lithography. Many efforts have been made to obtain a useful theoretical description of the electron transport problem using different approaches: Analytical models (Thümmel 1974, Dudek 1980, Niedrig 1982) involve strong simplifications of the transport problem or are restricted to describe partial aspects only of the problem. Simulation of electron trajectories by Monte-Carlo calculations has been successfully applied (Berger 1963, Krefting and Reimer 1973, Shimizu et al. 1976, Ichimura et al. 1980) to describe the major effects. Small effects, however, i.e. the effects of rare events, are practically excluded from being reproduced by the statistics.

Solving the transport equation directly by numerical methods (Brown et al. 1969, Strickland et al. 1976, Fathers and Rez 1979, Schmoranzer and Hoffmann 1980, Lanteri et al. 1981) has proved to be an advantageous alternative. The procedure commonly used is to reduce the number of co-ordinates to the ones pertinent to the distributions in question and to discretize a system of first-order differential equations.

In this work a numerical procedure was developed (Schmoranzer and Hoffmann 1980, Hoffmann and Schmoranzer 1981, Hoffmann and Schmoranzer 1982) which solves the transport equation via an integral equation which, according to experiences from neutron transport calculations (I. Kuscer, private communication; J. Wick, private communication), can be expected to have some numerical advantages over the differential equation. The method will be described and selected results of various calculations will be presented: e.g. the angular and energy distribution of transmitted and backscattered electrons for Al, Cu, Ag, and Au.

MATHEMATICAL PROCEDURE

We start out from the transport equation written in general form, an integro-differential equation:

$$\frac{\partial \psi(\vec{r},\vec{v},t)}{\partial t} + \vec{v} \cdot \nabla \psi(\vec{r},\vec{v},t) + v\, \sigma(\vec{r},\vec{v})\, \psi(\vec{r},\vec{v},t) = q(\vec{r},\vec{v},t) + \int d\vec{v}'\, \sigma(\vec{v}' \rightarrow \vec{v},\vec{r})\, v'\, \psi(\vec{r},\vec{v}',t) \qquad (1)$$

where

LIST OF SYMBOLS

ψ	= Probability density of finding an electron at $\vec{r}$, $\vec{v}$, t
$\vec{r}$	= Electron position
x,y,z	= Spatial co-ordinates
$\vec{v}$, $\vec{v}'$	= Electron velocity
v_x, v_y, v_z	= Velocity components
t	= Time
$\sigma(\vec{r}, \vec{v})$, $\sigma(z,v)$	= Macroscopic total scattering cross section
q	= Electron source density
$\sigma(\vec{v}' \rightarrow \vec{v},\vec{r})$, $\sigma\left(v', v, \frac{\vec{v}' \cdot \vec{v}}{v' v}, z\right)$	= Macroscopic scattering cross section
$\vec{j}$	= Differential electron flux density
Θ	= Spatial polar angle
$\hat{z}$	= Unit vector in z-direction
Q	= Electron source term in the integral equation
R,R′	= Integration variables
Z	= Atomic number
$d\sigma_{RMB}$	= Differential elastic scattering cross section after Riley et al. 1975
$d\sigma_{scR}$	= Screened Rutherford cross section
E	= Electron energy
E_0	= Primary electron energy
W	= Relative electron energy normalized to E_0
η_B	= Backscattering coeffient
I	= Mean ionization energy
η_T	= Transmission coefficient
N(Θ)	= Electron current normalized to solid angle and primary current

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$\psi(\vec{r},\vec{v},t)$ = probability density of finding an electron at a point $\vec{r}$ with velocity $\vec{v}$ at time t

$\vec{j}(\vec{r},\vec{v},t) = \vec{v}\psi(\vec{r},\vec{v},t)$ = differential flux density

$q(\vec{r},\vec{v},t)$ = source density

$\sigma^{-1}(\vec{r},\vec{v})$ = mean free path for total scattering

$\sigma(\vec{v}' \rightarrow \vec{v},\vec{r})$ = macroscopic scattering cross section (scattering probability per unit path length)

The solution describes the differential flux density from which all quantities for characterizing the propagation of electrons in matter can be derived (by integration).

In establishing equation 1 the following assumptions have been included:

1. The scattering centers are randomly distributed and at rest.
2. No interactions between propagating electrons.
3. The electron during its passage through the scattering medium interacts with one scattering center only at one time.
4. The electron trajectory is a straight line between the collisions, i.e., a zigzag line over-all.

Now, instead of discretizing the integro-differential equation which yields a system of difference equations, we aim at transforming equation 1 into an integral equation first before applying numerical procedures.

The following simplifications can be made which are compatible with many practical applications:

1. The problem is stationary with respect to time.
2. $\sigma(\vec{v}' \rightarrow \vec{v},\vec{r})$ possesses rotational invariance, i.e.

$$\sigma(v' \rightarrow \vec{v},\vec{r}) = \sigma\left(v', v, \frac{\vec{v}' \cdot \vec{v}}{v' v}, \vec{r}\right)$$

3. Plane symmetry: If the z-axis is chosen perpendicular to and the x,y-plane parallel to the surface of the target layer, then
   a) the quantities $\sigma(\vec{r},\vec{v})$, $\sigma(\vec{v}' \rightarrow \vec{v},\vec{r})$ and $q(\vec{r},\vec{v})$ depend on one of the spatial co-ordinates only, i.e., z,
   b) the angular dependence of $q(\vec{r},\vec{v})$ is on $\cos\theta = \frac{\vec{v}}{v} \cdot \hat{z}$ only,
   c) any boundary conditions must depend on z and $\cos\theta$ only.

In other words, the macroscopic cross sections depend on the magnitudes of the electron velocity vectors before ($\vec{v}'$) and after ($\vec{v}$) the individual scattering process and on the cosine of the angle between $\vec{v}'$ and $\vec{v}$, i.e. the polar scattering angle, whereas they do not depend on the azimuthal scattering angle. The target is semi-infinite and flat. The source, too, is infinite and constant in the x,y co-ordinates. In realistic cases, the source is often point-like with respect to the x,y co-ordinates, so that the above assumption is equivalent with integrating the electron flux density over the x,y co-ordinates. Of course the information on the x,y-dependence of the electron flux density is lost therewith. It should be noted that this is also the case in all other methods mentioned above which solve the transport equation numerically.

The number of variables has been reduced thus from seven (x,y,z,v_x,v_y,v_z,t) to three (z, cos θ, v). The reduced transport equation is then transformed into a Fredholm type integral equation of the second kind.

$$\psi(z,v,\cos\theta) =$$

$$= \frac{1}{v} Q(z,v,\cos\theta) + \frac{1}{v} \int_0^\infty dR \left\{ \exp\left[- \int_0^R dR' \right.\right.$$

$$\left.\sigma(z - R'\cos\theta, v)\right] *$$

$$* \int d\vec{v}'\, v'\, \sigma\left(v', v, \frac{\vec{v}' \cdot \vec{v}}{v' v}, z - R\cos\theta\right) \psi(z - R\cos\theta, v',$$

$$\left.\cos\theta')\right\} \qquad (2)$$

where

$$Q(z,v,\cos\theta) = \int_0^\infty dR \{\exp[-\int_0^R dR' \sigma(z-R'\cos\theta, v)]^* \\ *q(z-R\cos\theta, v, \cos\theta)\}$$

A collocation method (see e.g. Baker 1977) using triangular basis functions was specially developed for numerical solution of the above integral equation. The collocation method is particularly suited for computer application because of its simple mathematical structure. One obtains a system of linear equations, of which the solution, i.e. the coefficients of the triangular basis functions, yield an approximation of the electron flux density. The system of linear equations, which usually consists of more than 10^4 equations, is solved by iteration using group relaxation.

The amount of computer time needed compares favorably with Monte-Carlo-methods (e.g. 15 min on a TR 440, Gibson-Mix 1.23 μs) for the flux density of a given layer.

CROSS SECTIONS FOR ELEMENTARY SCATTERING EVENTS

The computer code has been written in such a way that differential cross sections for elastic and inelastic scattering can be used in closed form as well as in tabulated form including interpolation specifications.

For the elastic scattering tabulated cross sections (Riley et al. 1975) were employed which have been calculated in central static potential approximation by partial wave expansion of the Dirac equation by means of Hartree-Fock wave-functions for $Z \leq 35$ and relativistic wave-functions for $Z \geq 36$. Comparison of these cross sections with the familiar screened Rutherford type cross section (Fig. 1) shows that there is little difference only for small atomic numbers (e.g. Z = 13) and scattering angles greater than 30°. For smaller scattering angles the discrepancy is in principle larger but not significant for transport considerations because of the small solid angle involved.

For larger atomic numbers the more accurate theoretical cross sections $d\sigma_{RMB}$ deviate more from the screened Rutherford ones $d\sigma_{scR}$, particularly also for large scattering angles. A similar behavior in the backscattering region has also been found for theoretical Mott cross sections (Doggett and Spencer 1956, Reimer et al. 1971) and has been experimentally investigated previously by our group (Schmoranzer and Grabe 1976, Grabe 1979). For large atomic numbers where elastic scattering is dominating, the angular distributions calculated on the basis of pure elastic electron-atom scattering have turned out to describe well the experimental results (Schmoranzer and Hoffmann 1980), particularly in the backward direction (in forward direction inelastic effects appear to be less negligible).

In a more general approach, inelastic effects have been included, too. The most widely used continuous slowing-down approximation inherent to the Bethe stopping power is known to yield discrepancies in the energy distribution of electrons transmitted through thin films (Shimizu et al. 1975, Adesida et al. 1980). It is important not to exclude the random nature of the inelastic scattering events in order not to lose important details, as e.g. the backscattering at primary

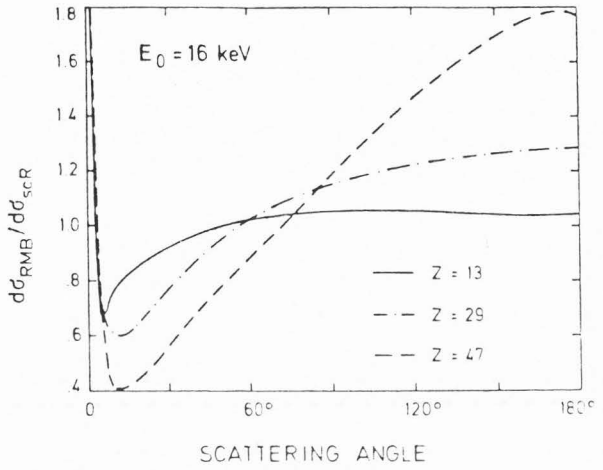

Fig. 1. Comparison of differential elastic scattering cross sections after Riley et al. (1975) $d\sigma_{RMB}$ with screened Rutherford ones $d\sigma_{scR}$.

energy which will be demonstrated later (see Fig. 6-Fig. 8). As to the inelastic scattering, Gryzinski's (Gryzinski 1965) excitation function has been chosen to describe the different kinds of energy losses. Distinction is made between core ionization and outer electron excitation, where the latter has to include inelastic solid state effects as e.g. interband transitions and plasmon excitation (Shimizu and Everhart 1978).

For inner shell ionization Gryzinski's formula has already been successfully applied in several Monte-Carlo calculations of other authors (Krefting and Reimer 1973, Shimizu et al. 1976, Ganachaud and Cailler 1979, Ichimura et al. 1980). For the outer electrons the set of parameters entering Gryzinski's formula was determined similarly to a procedure proposed earlier (Shimizu and Everhart 1978). Assuming the Bethe law as an over-all empirical fact, the sum of the Gryzinski type stopping powers for core and outer electron excitation must yield the Bethe stopping power (see Fig. 2). The number of outer electrons cannot be easily determined, particularly not for compound material (Ichimura et al. 1980). For a more universal application, it appeared suitable to choose a convenient energy parameter first and then to match the number parameter so that the required over-all agreement of core plus outer electron stopping powers with the Bethe law was obtained. Indeed the parameters of the outer electron stopping power and excitation function are physically less meaningful than in the core electron case, but the advantage lies in the general applicability of the fitting procedure.

Finally, the angular deflection of electrons upon the inelastic scattering event was accounted for approximately by multiplying the elastic scattering cross section of small atomic number Z by $(Z + 1)/Z$ (see e.g. Brown et al. 1969 and further references therein).

RESULTS AND DISCUSSION

The method was applied to a number of elements for which detailed experimental results are available for comparison.

Transmission coefficient and backscattering coefficient can be obtained by integration of the outgoing electron flux density over angle and energy. For electrons of 20 keV primary energy and normal incidence the absolutely calculated coefficients as a function of film thickness can be compared for Al with the measurements (Fig. 3). The agreement is as good as in recent Monte-Carlo work (Adesida et al. 1980).

As mentioned earlier the method yields energy and angular distributions of transmitted and backscattered electrons as well. For Al e.g. the energy distribution of transmitted electrons (Fig. 4) agrees rather well with the measurements.

The angular distribution of transmitted electrons for Al (Fig. 5) shows satisfactory agreement with the early experimental data (Cosslett and Thomas 1964).

For backscattered electrons the calculated energy distributions for Al, Cu, Ag, and Au are shown in Fig. 6. The film thicknesses were chosen such as to yield a total backscattering coefficient close to the one of the bulk material according to the measurements (Niedrig and Sieber 1971). The distributions for the lighter elements show a broad maximum with increasing height, increasing energy of the maximum, and decreasing energy width with increasing atomic number. For Au the distribution looks monotonic. This behavior is well known from experiments (Kulenkampff and Spyra 1954, Kanter 1957, Darlington 1975, Matsukawa et al. 1974, Bauer 1979). It should be noted that a peak of elastically backscattered electrons is reproduced by the present calculations, whereas it has been missing in most other calculations so far because of too coarse an energy grid, the continuous slowing down model or the small probability of the event in the case of Monte-Carlo calculations. The only theoretical work in which to our knowledge the elastic peak has been quoted, too, is the one of Brown, Wittry and Kyser (1969). To these authors, however, it was not clear whether the tendency of their theoretical curves to turn up for energies approaching E_0 was a real effect or an artefact. Our results are in good agreement with recent extensive measurements by Bauer (1979) and earlier experiments (Boersch et al. 1967, Sommerkamp 1970). The ratio of elastic to inelastic scattering increases with increasing atomic number.

Further details can be seen in the following Figs. 7 and 8 where the energy distributions of backscattered electrons at different angles are shown for e.g. Al and Ag. The elastic peak increases with angle until about 130° and stays constant then. On the other hand the broad inelastic maximum increases with angle, shifting towards larger energy losses. The minimum occurs for Al at scattering angles $\Theta \geq 110°$ and for Ag at $\Theta \geq 130°$.

The width of the so-called peak of elastically backscattered electrons in our calculations intrinsically depends on the mesh size used of the energy grid, i.e. it cannot come out smaller than the mesh size, which is also the energy width assumed for the source of primary electrons. In the calculations discussed here the mesh size was always 200 eV in the energy region of $0.8\ E_0$ to E_0 and wider outside. It should be noted that therefore the grid point next to E_0 already represents inelastically scattered electrons. The number of electrons which have been subjected to purely elastic scattering (single as well as plural) follows from the density $d\eta_B/dW$ at grid point E_0.

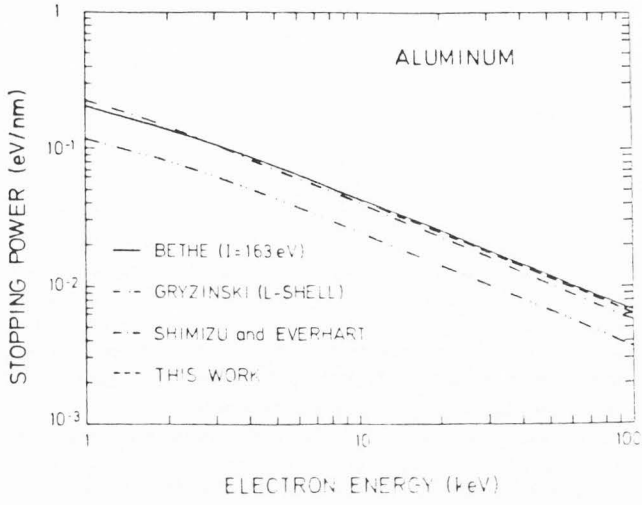

Fig. 2. Stopping powers for aluminum:
——— Bethe law
— · · — Gryzinski formula for L-shell electrons only
— · — sum of inner and outer electron Gryzinski stopping powers after Shimizu and Everhart (1978)
— — — sum of inner and outer electron Gryzinski stopping powers, where outer electron parameters were fitted as explained in text.

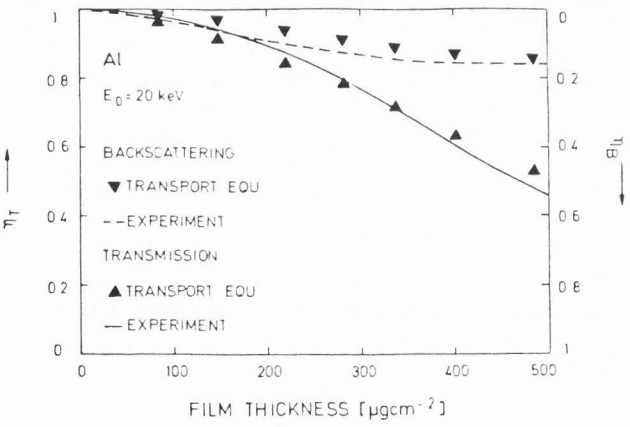

Fig. 3. Transmission coefficient η_T and backscattering coefficient η_B for Al
▼ ▲ transport equation calculations
— — — experimental data taken from Niedrig and Sieber (1971) and Krefting and Reimer (1973)
——— experimental data taken from Krefting and Reimer (1973) and Shimizu et al. (1976).

As an example for the angular distribution of backscattered electrons the results for Al are displayed in Fig. 9 in a polar diagram. The smaller film thickness (note the change of scale) corresponding to 320 nm clearly shows the maximum contribution around 130°, whereas the distribution approaches the cosine law at 1040 nm.

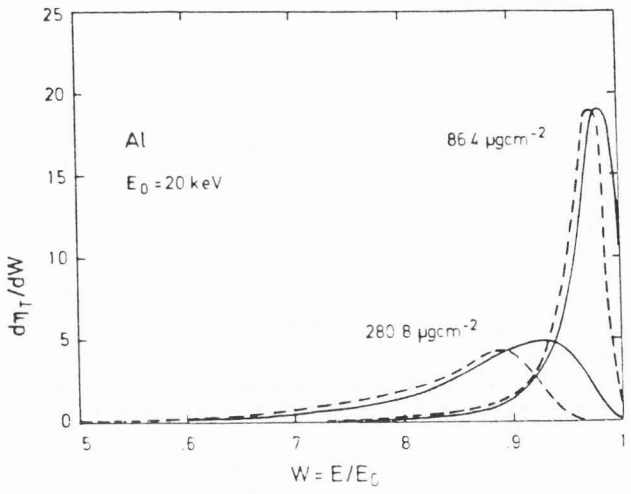

Fig. 4. Energy distribution of transmitted electrons for Al at different film thicknesses
_____ transport equation calculations
– – – experimental data taken from Shimizu et al. (1976).

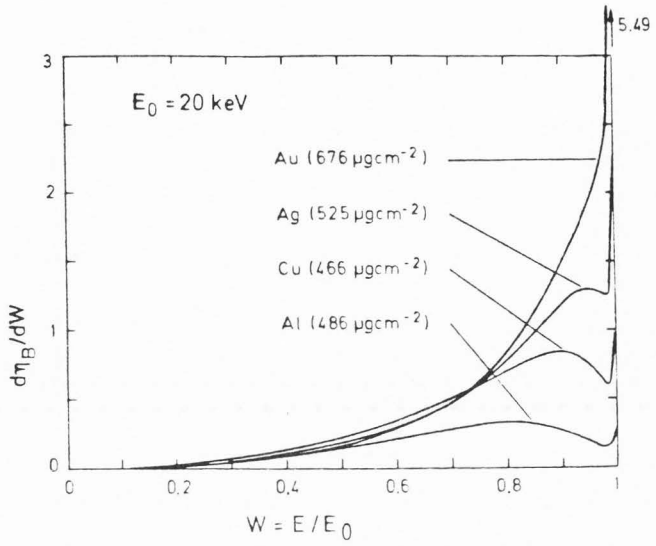

Fig. 6. Energy distributions of electrons backscattered from Al, Cu, Ag, and Au calculated by transport equation.

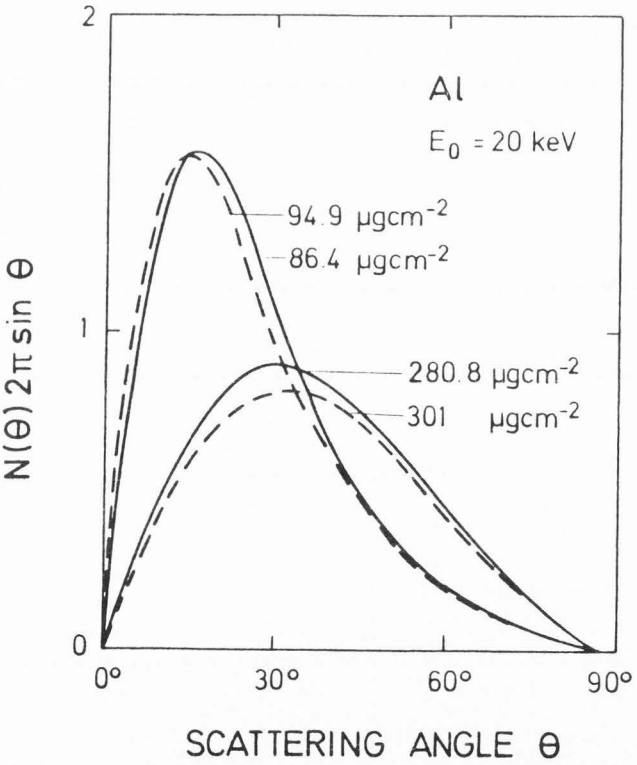

Fig. 5. Angular distribution of transmitted electrons for Al at different film thicknesses
_____ transport equation calculations
– – – experimental data taken from Cosslett and Thomas (1964).

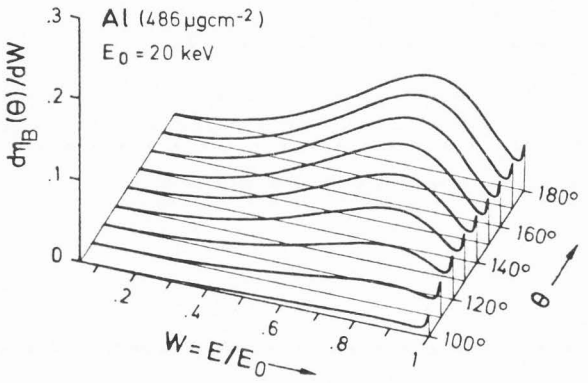

Fig. 7. Energy distribution of backscattered electrons for Al at different scattering angles calculated by transport equation.

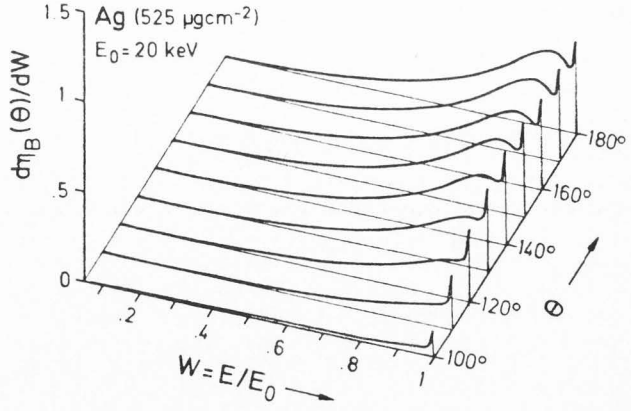

Fig. 8. Energy distribution of backscattered electrons for Ag at different scattering angles calculated by transport equation.

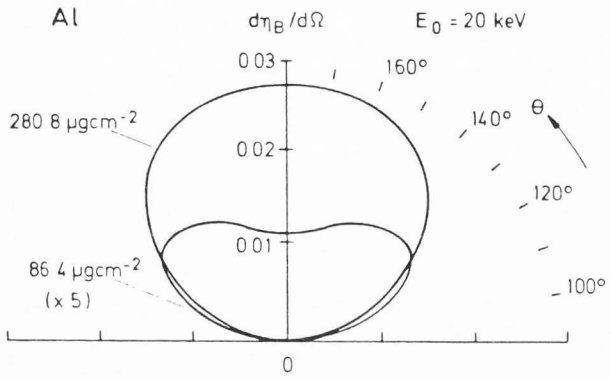

Fig. 9. Angular distribution of backscattered electrons for Al calculated by transport equation.

SUMMARY

In summary the presented method for solving the transport equation for the propagation of electrons in the primary energy range of interest for electron beam technology is based on discretizing the related integral equation by means of a collocation procedure and iterative solution of a system of linear equations. Together with proper choice of differential cross sections for elastic scattering and with adequate representation of the complex energy loss mechanisms the method yields a rather satisfactory theoretical description of angular and energy distributions of electrons under the boundary conditions of plane symmetry and, last but not least, comparing to previous Monte-Carlo and transport equation calculations, the results correctly reproduce some more detailed information.

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