

# Backscattering of 10–35 keV Electrons from Thick Targets at Normal Incidence

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Energy distributions of backscattered electrons were obtained by Monte Carlo simulations for a wide range of elements for electron beams at normal incidence and with energies  $E_0$  between 10 and 35 keV. These spectra were compared with experimental data and with a theoretical expression. An analytical expression was fitted to the simulated data. An expression for the backscattered electron coefficient  $\eta$  as a function of atomic number  $Z$  and incident energy  $E_0$  was obtained. A model for  $\eta$  and the energy distribution of backscattered electrons for multi-element samples is suggested.

## INTRODUCTION

Packwood and Brown<sup>1</sup> showed that the depth distribution of characteristic x-ray production  $\phi(\rho z)$  can be described by a Gaussian expression:

$$\phi(\rho z) = \gamma_0 \exp[-\alpha^2(\rho z)^2] \times \left[ 1 - \left( \frac{\gamma_0 - \phi_0}{\gamma_0} \right) \exp(-\beta \rho z) \right] \quad (1)$$

which requires an accurate knowledge of the parameters  $\alpha$ ,  $\beta$ ,  $\gamma_0$  and  $\phi_0$ ,  $\rho z$  being the mass depth.

Tirira and Riveros<sup>2</sup> showed that the parameters  $\gamma_0$  and  $\phi_0$  can be described by

$$\gamma_0 = \frac{2 \int_1^{v_0} Q(U) dU}{Q(U_0) \int_1^{v_0} dU} + \frac{2}{Q(U_0)} \int_1^{v_0} Q(U) \frac{d\eta}{dU} dU \quad (2)$$

and

$$\phi_0 = 1 + \frac{2}{Q(U_0)} \int_1^{v_0} Q(U) \frac{d\eta}{dU} dU \quad (3)$$

where  $U$  is the overvoltage,  $U_0$  is the incidence overvoltage and  $Q(U)$  is the ionization cross-section.

In order to assess Eqns (2) and (3), a model for  $d\eta/dU$  must be used. Czyzewski and Szymanski<sup>3</sup> developed an expression for this, but it is a complicated function of  $U$  and makes the integration difficult. For this reason, a simple analytical expression to describe  $d\eta/dU$  spectra was fitted in this work. An evaluation of the use of the complete expression given by Eqns (2) and (3) will be the subject of a subsequent paper.

In order to carry out this fitting, a set of data from Monte Carlo simulations was used because of the difficulties in obtaining experimental spectra. For this purpose, the MC program<sup>4</sup> was used.

This simulation model uses Bethe's law<sup>5</sup> for continuous energy loss along a segmented path. Each segment or step represents the distance between two single elastic interactions and is taken as a stochastic variable in addition to both angles involved in each deflection. In determining the step length and polar scattering angle, a single screened Rutherford cross-section is used.

In this simulation model, approximately 10 000 paths are required in order to obtain a mean square deviation of about 1% in the maximum of the backscattered electron energy distribution. Thus, about 8 computer hours (PC-AT, 8 MHz) are needed in order to obtain each spectrum.

## EXPERIMENTAL AND RESULTS

### Backscattering coefficient at normal incidence

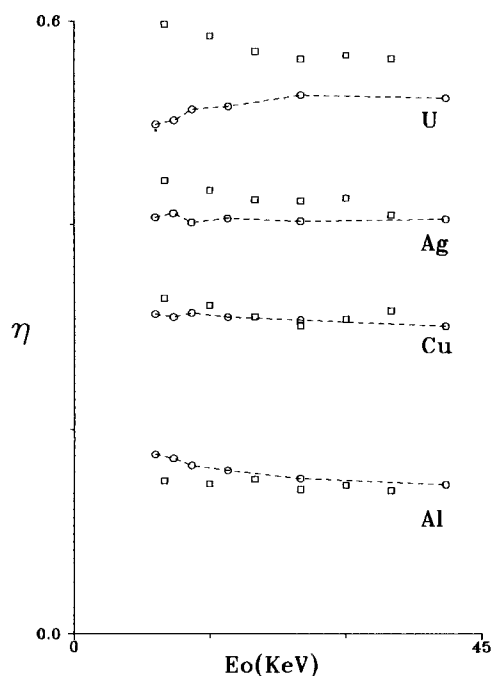
A large number of backscattered spectra (138) for normal beam incidence at energies  $E_0$  of 10, 15, 20, 25, 30 and 35 keV were obtained for atomic numbers  $Z$  from 13 to 92. The simulations were performed for electron energies between  $E_0$  and 1 keV. Thus, the fraction of backscattered electrons neglected (with energies lower than 1 keV) is lower than 0.3% (see, e.g., Ref. 6).

The results confirmed a strong dependence of  $\eta$  on the atomic number of the specimen and a weaker dependence on the incidence energy,  $E_0$ ; this behaviour has been found previously by other workers.<sup>6-8</sup> This can be seen in Fig. 1, where  $\eta$  values obtained by MC and by Drescher *et al.*<sup>8</sup> are plotted as a function of  $E_0$ .

A simple fitting analytical expression for these data as a function of  $Z$  and  $E_0$  was developed:

$$\eta(Z, E_0) = aZ^{0.02} + bE_0 + c \quad (4)$$

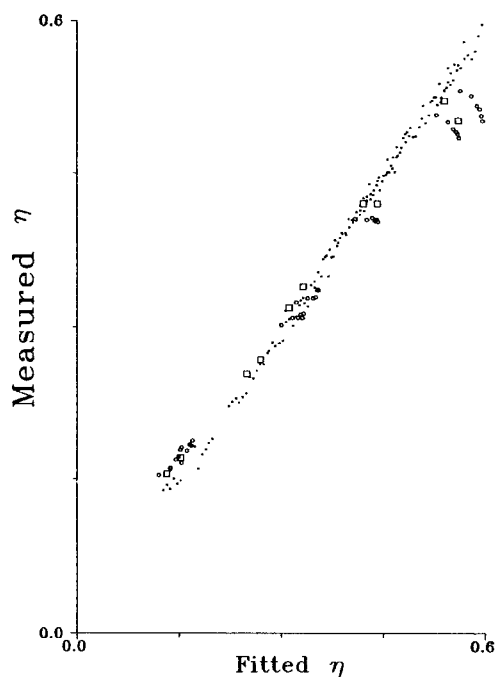
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**Figure 1.** Experimental and simulated backscattering coefficient  $\eta$  as a function of beam energy.  $\circ$ , Drescher *et al.*,<sup>8</sup>  $\square$ , this work.

where  $a = 10.50 \pm 0.05$ ,  $b = -0.00102 \pm 0.00007$  and  $c = -10.911 \pm 0.007$ , which fits all 138 data with an r.m.s. error  $\sigma$  of 0.007 and a multiple regression coefficient of 0.9983. Nevertheless, it is necessary to keep in mind that this is merely a fit, and therefore it must not be used away from the range of values for which it was developed.

In Fig. 2 a plot of  $\eta$  given by the Monte Carlo method *vs.* the corresponding  $\eta$  values predicted by the fitting can be observed. Experimental data from Bishop<sup>9</sup> and Drescher *et al.*<sup>8</sup> were also plotted against the corresponding  $\eta$  values given by Eqn (4). It can be seen that the Monte Carlo data are closely distributed around a straight line of slope unity, showing the quality of this fitting. The experimental and simulated data show very good agreement, with deviations only for high  $\eta$ . These deviations are more clearly showed in Fig. 1; a systematic trend for the experimental data to remain below the simulated data occurs at high  $Z$ , especially for low incident energies. The most important fact is that also in these cases a difference in the behaviour of the data is observed. For simulated data  $\eta$  decreases for increasing  $E_0$ , whereas for the experimental data  $\eta$  increases. These effects, also noted by Bishop,<sup>9</sup> could be due to either the use of Born's approximation in the elastic cross-section expression or the continuous energy loss approximation. The first possibility is sustained by the fact that Born's approximation fails for large deflection angles, since the scattering probability at large angles is high both when the electron to be scattered has low energy and when the atomic nucleus is heavy. Both conditions are given precisely when the largest deviations between the simulated and experimental data are observed. Perhaps this problem can be elucidated using more accurate cross-sections in the simulation model, e.g., the cross-sections reported by Mott and Massey.<sup>10</sup>



**Figure 2.** Experimental and simulated backscattering coefficient  $\eta$  as a function of the corresponding values predicted by Eqn (4).  $\circ$ , Drescher *et al.*,<sup>8</sup>  $\square$ , Bishop,<sup>9</sup>  $\bullet$ , this work.

### Energy distribution of backscattered electrons

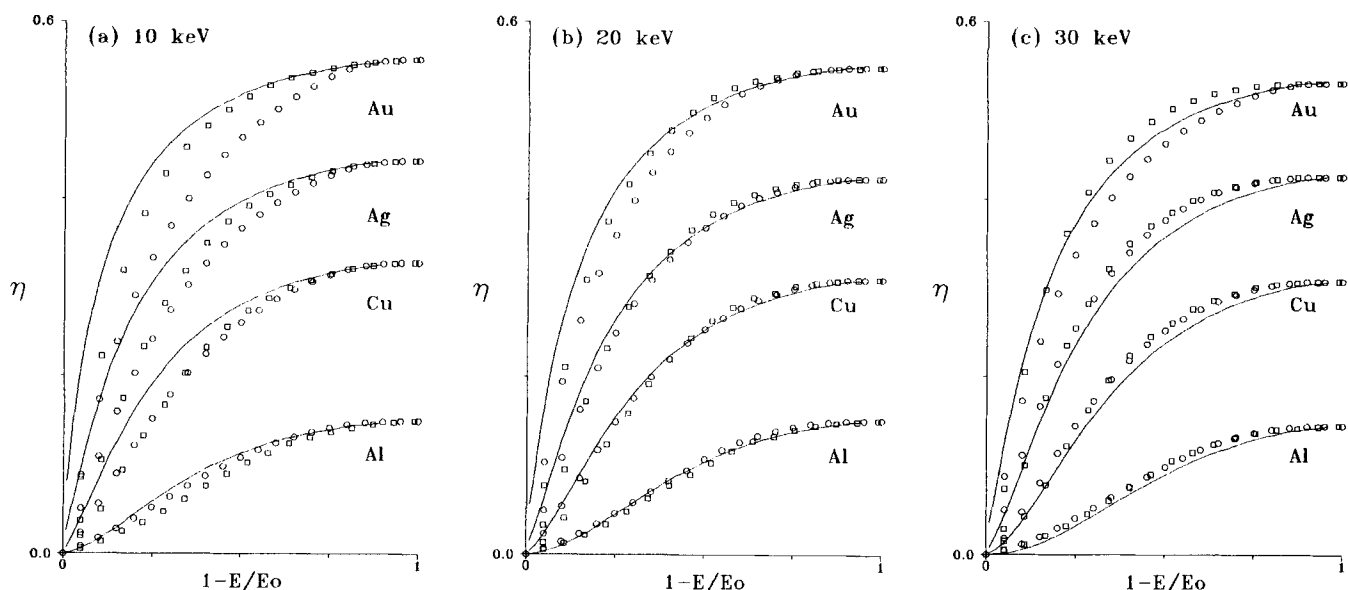
As indicated above, spectra with energies between  $E_0$  and 1 keV were obtained from simulations, and this range was divided into 51 intervals.

In Fig. 3, MC data are compared with the experimental values published by Darlington<sup>6</sup> and with those predicted by Czyzewski and Szymanski<sup>3</sup> for aluminium, copper, silver and gold at 10, 20 and 30 keV. Close correspondence between the experimental and simulated data can be observed; only for all the gold spectra and the silver spectrum at 10 keV are the experimental data slightly below those produced by the MC program in the intermediate region. This agreement shows that the set of MC data used to fit an analytical expression is adequate; nevertheless, the deviations of  $\eta$  with respect to the experimental data mentioned in the previous section, also do affect the shape of the differential energy distribution of backscattered electrons in the same cases. On the other hand, in all cases the MC data approach the experimental values more closely than (or, in a few cases, as closely as) those given by Czyzewski and Szymanski.<sup>3</sup>

In analysing the behaviour of simulated spectra, typical Poisson shapes were observed. This behaviour is in agreement with the relationship found between the most probable value and the arithmetic mean value of  $p = 1 - W$  ( $W = E/E_0$ ) for  $d\eta/dp$ . This ratio was found to be approximately 0.5, as expected for a Poisson distribution. For this reason, the following trial function, normalized to unity, was chosen:

$$\frac{d\hat{\eta}}{dp} = \frac{p \exp(-p/\lambda)}{\lambda^2} \quad (5)$$

Since the spectra obtained by simulations may be regarded as the sum of an actual distribution and a



**Figure 3.** Experimental and simulated integral spectrum of backscattered electrons for several elements and beam energies. (a) 10; (b) 20; and (c) 30 keV.  $\circ$ , Darlington;<sup>6</sup>  $\square$ , this work. The function obtained by Czyzewski and Szymanski<sup>3</sup> is also plotted (solid lines).

white noise, the integral spectrum  $\eta(p)$ , which represents the probability of an electron being scattered with values ranging from 0 to  $p$ , was used to find the best  $\lambda$  in Eqn (5). Thus the white noise effect is significantly reduced. Then, from Eqn (5), the functional form for  $\hat{\eta}(p)$  is

$$\hat{\eta}(p) = 1 - (1 + p/\lambda)\exp(-p) \quad (6)$$

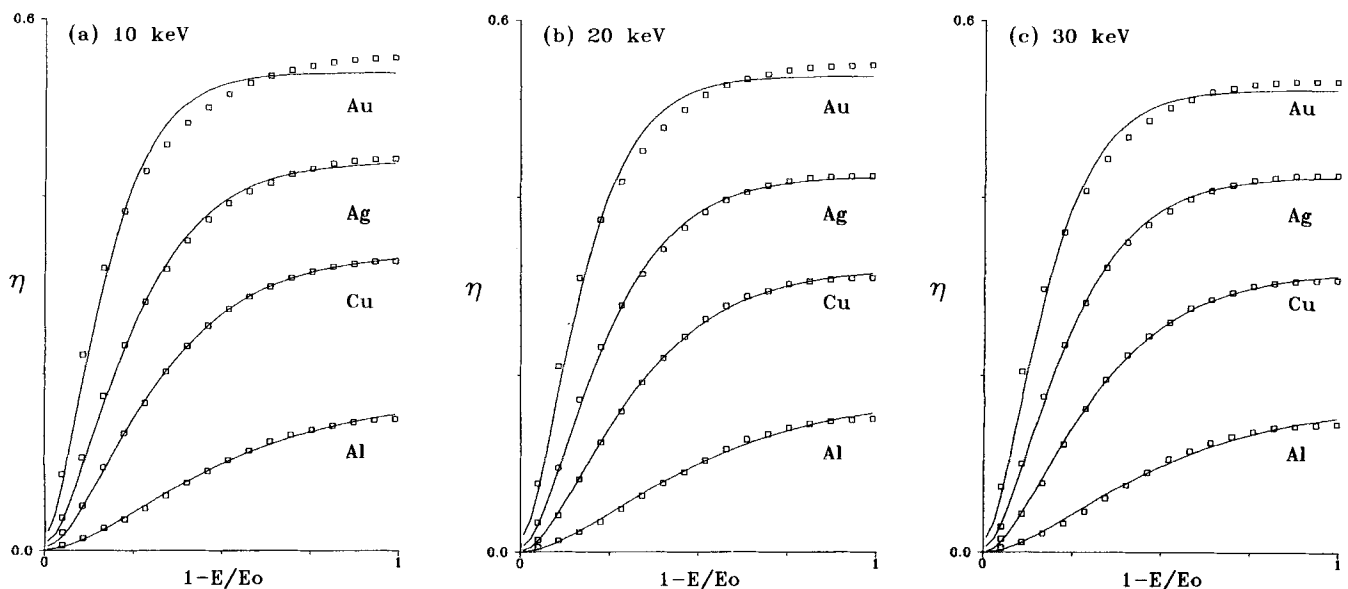
A linear fit of the function  $\eta(p)$ , obtained from the simulations, vs.  $\hat{\eta}(p)$  was performed for each spectrum; the following expression is proposed:

$$\eta(p) = c_1\hat{\eta}(p) + c_2$$

where  $c_1$  and  $c_2$  are constants. In order to carry out this fit,  $\lambda$  was chosen so as to be capable of minimizing the r.m.s. error by means of a numerical method of mini-

mization. It was found that all spectra are appropriately fitted by this trial function (see Fig. 4), producing in the most unfavourable case a linear correlation coefficient  $r = 0.996$  and  $\sigma = 4\%$ , the worst values of  $r$  corresponding to high atomic numbers and low energies; on the other hand, the best case produces  $r = 0.99998$  and  $\sigma = 0.4\%$ . The multiplicative constant  $c_1$  corresponding to each spectrum is very close to the  $\eta$  coefficient for backscattered electrons. Differences greater than 10% were found in only very few cases, all of which also correspond to high atomic numbers, especially at low energies. The greatest additive constant  $c_2$  was  $0.04\eta$ , a typical value being  $0.005\eta$ . These considerations lead to a definition of  $\hat{\eta}'$  as  $\hat{\eta}'(p) = \eta\hat{\eta}(p)$ .

The resulting values of  $\lambda$  from all the spectra show a strong dependence on atomic number. From Eqn (5) it can be seen that this parameter represents the value of  $p$



**Figure 4.** Fitting (solid lines) of integral spectrum of backscattered electrons for several elements and beam energies (a) 10; (b) 20; (c) 30 keV. MC simulated data are also plotted ( $\square$ ).

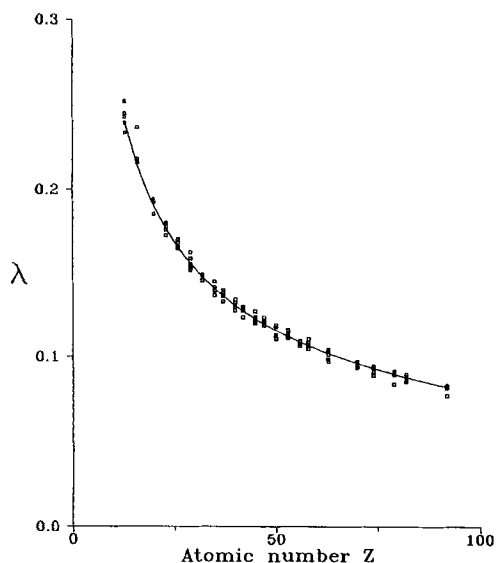


Figure 5. Coefficient  $\lambda$  vs. atomic number  $Z$  from all MC spectra and the fit (solid curve) given by Eqn (7).

for maximum backscattering probability. According to other workers,<sup>6,7</sup> the parameter  $\lambda$  approaches zero for increasing  $Z$ . In other words, most of the backscattered electrons leave the sample with an energy approaching  $E_0$  for increasing  $Z$ . On the other hand, the dependence of  $\lambda$  on the incidence energy was found to be too weak to be clearly determined.

Taking into account these considerations, a fit of  $\lambda$  vs.  $Z$  was performed:

$$\lambda = AZ^{-0.5} + B \quad (7)$$

producing a regression coefficient  $r = 0.9879$  and  $\sigma = 0.006$ , with  $A = 0.907 \pm 0.012$  and  $B = -0.012 \pm 0.006$ . Values of  $\lambda$  predicted by this expression and those produced by the simulations are plotted as a function of  $Z$  in Fig. 5.

Then, from Eqns (5)–(7), we obtain

$$\frac{d\hat{\eta}'}{dp}(p) = \frac{\eta p}{(AZ^{-0.5} + B)^2} \exp[-p/(AZ^{-0.5} + B)] \quad (8)$$

and

$$\hat{\eta}'(p) = \eta \left\{ 1 - \left( 1 + \frac{p}{AZ^{-0.5} + B} \right) \times \exp[-p/(AZ^{-0.5} + B)] \right\} \quad (9)$$

In order to assess the performance of Eqn (9), corresponding r.m.s. errors with respect to simulated data were calculated for each of the 138 spectra, the largest  $\sigma$  being 9% and the smallest 1%; the respective correlation coefficients were 0.994 and 0.999985. These considerations suggest that the trial function  $\hat{\eta}'(p)$  is adequate to express the integral spectrum of backscattered electrons,  $\eta(p)$ . The greatest deviations correspond to high atomic numbers and low incidence energies, as occurs in the fitting of the trial function given by Eqn (6), i.e. without including the expression fitted to  $\lambda$ . It has been seen that in these cases, simulated data apart from experimental data: with improvement of the simulation, perhaps better agreement between the

fit and simulated data would be obtained and, further, both the fitted and simulated data would approach the experimental values.

### Backscattering in binary samples

So far only single element samples have been studied and nothing has been stated about multielement samples. The development of an expression that is able to deal with the latter would be of great interest in microanalysis. Since the MC program produced very good results for pure samples, it is necessary to look at the expressions used in the program which involve a dependence on any parameter proper of the considered element (e.g. atomic number). A more effective way of averaging these expressions when more than one element is present in the sample will be considered.

The MC model for electron backscattering involves three magnitudes, depending on the elements that constitute the sample: the stopping power, the differential cross-section for elastic scattering,  $\sigma(\theta)$ , and the total cross-section,  $\sigma_E$ , Rutherford's expressions being used for cross-sections.

Bethe's law for energy loss<sup>5</sup> can be stated as

$$dE/ds = Ng(E) \quad (10)$$

where  $N$  represents the number of atoms per unit volume in the sample and  $g(E)$  is the average stopping per scattering centre taking into account all electronic levels:

$$g(E) = -\frac{2\pi e^4}{E} Z \ln(1.166E/J) \quad (11)$$

where  $e$  is the electron charge and  $J$  is the mean ionization potential.

If the sample is composed of more than one element, then a passing electron loses energy because of the interactions with each of the atoms of these elements; hence Bethe's law may be expressed as

$$dE/ds = N_1 g_1(E) + N_2 g_2(E) \quad (12)$$

where  $N_i$  represents the number of atoms of type  $i$  per unit volume in the sample:

$$N_i = \frac{C_i N_0}{A_i} \rho_{\text{sample}} \quad (13)$$

where  $C_i$  is the mass concentration,  $A_i$  is the atomic weight,  $N_0$  is Avogadro's number and  $\rho_{\text{sample}}$  is the density of the sample. Bearing in mind that the dependence of the stopping power on mean ionization potential is weak, the following approximation can be made:

$$\frac{dE}{ds} = -\frac{2\pi e^4}{E} \bar{Z} N \ln[1.166E/(0.0115\bar{Z})] \quad (14)$$

in which Wilson's<sup>11</sup> ionization potential has been used, and the following definition for  $\bar{Z}$  has been taken:

$$\bar{Z} = \frac{N_1 Z_1 + N_2 Z_2}{N_1 + N_2} \quad (15)$$

In the most unfavourable cases (e.g. a binary sample with  $Z_1 < 15$ ,  $Z_2 > 90$  and  $N_1 = N_2$  for a typical energy of 10 keV), the difference between Eqns (12) and

(14) is about 10%. Therefore, the same expression as for a pure sample, evaluated with the mean atomic number (atomic average), can be used for the stopping power of a binary sample.

It has been shown in Eqn (12) that the stopping power must be atomic averaged in order to obtain an average with physical meaning. In order to arrive at this conclusion, Bethe's original ideas<sup>5</sup> were used. The analysis shows that the approximation made by Reed,<sup>12</sup> according to which the stopping power must be mass averaged, is not adequate. He assumed that the density of a binary sample was a function depending only on the density of each of the components and of their respective concentrations.

The cross-section for elastic scattering,  $\sigma(\theta)$ , represents the probability of an electron being scattered at an angle between  $\theta$  and  $\theta + d\theta$  by a scattering centre. Rutherford's expression is

$$\sigma(\theta) = \frac{Z^2 e^4}{16E^2(\sin^2 \theta/2 + \alpha)} \quad (16)$$

where  $\alpha = 4.34 \text{ eV } Z^{2/3}/E$ .

When there is more than one type of element in the sample, it is necessary to average cross-sections according to the number of scattering centres of each type present in the sample:

$$\sigma(\theta) = \frac{N_1 \sigma_1(\theta) + N_2 \sigma_2(\theta)}{N_1 + N_2} \quad (17)$$

An approximation similar to those carried out in dealing with the stopping power can be adopted:

$$\sigma(\theta) = \frac{\bar{Z}^2 e^4}{16E^2(\sin^2 \theta/2 + \bar{\alpha})} \quad (18)$$

where  $\bar{\alpha} = 4.34 \text{ eV } \bar{Z}^{2/3}/E$ .

In comparing Eqns (17) and (18) small differences are observed; for example, for the unfavourable case considered above, the relative standard deviation is about 10% for the angles in which the scattering probability is not negligible. In Fig. 6 a plot of exact and approximated cross-sections as a function of the scattering angle,  $\theta$ , is shown. Good agreement between both

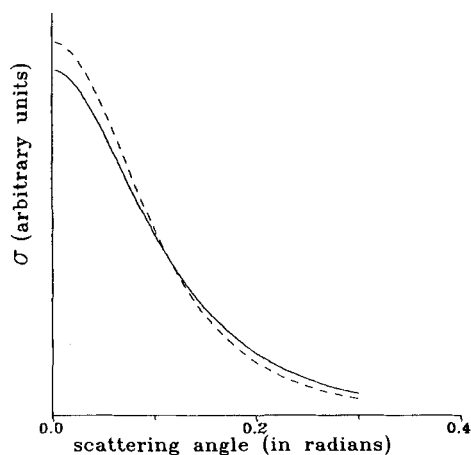


Figure 6. Exact (solid line) and approximated (broken line) cross-sections for elastic scattering,  $\sigma(\theta)$ , for a hypothetical compound 50% U-50% Al (atomic concentrations) at 10 keV as a function of scattering angle.

expressions for cross-section can be observed; this suggests that the approximation is acceptable.

The total cross-section,  $\sigma_E$ , is the integral of  $\sigma(\theta)$  between  $\theta = 0$  and  $\theta = \pi$ :

$$\sigma_E = \frac{Z^2 e^4 \pi}{4E^2 \alpha (1 + \alpha)} \quad (19)$$

From Eqn (17), it is possible to express the total cross-section as

$$\sigma_E = \frac{N_1 \sigma_{E1} + N_2 \sigma_{E2}}{N_1 + N_2} \quad (20)$$

If the same procedure as for the stopping power and the differential cross-section is performed, it can be seen that in the cases considered above, the difference between Eqn (19) evaluated with  $\bar{Z}$  and Eqn (20) is also small (about 12%).

The probability density of scattering for an angle between  $\theta$  and  $\theta + d\theta$ , used by the MC program for the calculation of scattering angles, is defined as the ratio between the differential and total cross-sections. As has been seen, in the expressions of both parameters for a binary sample it is reasonable to approximate the exact expressions by those obtained by evaluating them with atomic averaged values of  $Z$ ; hence, this approximation applied to the probability density should also be good.

Summarizing, if binary samples are considered, in order to give an analytical expression for  $\eta$  or for spectral distribution of backscattered electrons, Eqn (4) or (8) evaluated with atomic average  $\bar{Z}$  may be used. Samples composed of more than two elements represent a straight generalization of the binary case.

## CONCLUSIONS

Analytical expressions for the backscattering coefficient and the spectral distribution have been fitted to data from MC simulations, showing good agreement with experimental data. The range of validity of these expressions depends on the incident energy, the agreement being closer for increasing incident energy. The values predicted by these expressions differ from the experimental data by more than 10% only for elements heavier than silver, for which the MC simulation overestimates the experimental data.

The expressions obtained for the spectrum of backscattered electrons allow an accurate and fairly simple assessment of some parameters involved in microanalysis correction models. An average of spectral distributions for pure elements has been suggested for multielement samples; its validity should be tested with experimental data.

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## REFERENCES

1. R. H. Packwood and J. D. Brown, *X-Ray Spectrom.* **10**, 138 (1981).
2. J. H. Tirira and J. A. Riveros, *X-Ray Spectrom.* **16**, 27 (1987).
3. Z. Czyzewski and H. Szymanski, in Proceedings of the 10th International Congress on Electron Microscopy, Hamburg, Vol. 1, p. 268 (1982).
4. M. del Giorgio, personal communication (1988).
5. H. A. Bethe, *Ann. Phys. (Leipzig)* **5**, 325 (1930).
6. E. H. Darlington, *J. Phys. D* **8**, 85 (1975).
7. H. E. Bishop, PhD Thesis, University of Cambridge (1966).
8. H. Drescher, L. Reimer and H. Seidel, *Z. Angew. Phys.* **29**, 6 (1970).
9. H. E. Bishop, *Br. J. Phys.* **18**, 703 (1967).
10. N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions*. Oxford University Press, Oxford (1949).
11. C. R. Wilson, *Phys. Rev.* **60**, 749 (1941).
12. S. J. B. Reed, *Electron Microprobe Analysis*, p. 203. Cambridge University Press, Cambridge (1975).