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Angular distribution of electrons elastically backscattered from non-crystalline solid surfaces

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Abstract. In the energy range 100 eV–2 keV, we applied the Monte Carlo method to analyse the elastic reflection coefficient and the angular distribution of electrons elastically backscattered from the solid surface of an isotropic and homogeneous medium. Results indicated that elastically backscattered electrons arose substantially from only a few scatterings with a single scattering event contributing to approximately half of these electrons. Thus, neither the multiple scattering model nor the single scattering model is sufficient to describe the angular distribution. To improve these models, we evaluated the contribution from one, two and three scatterings exactly and higher scatterings by the P_1 -approximation, an approximate method to solve the Boltzmann transport equation assuming multiple elastic scattering of electrons in the solid. This approach allowed us to derive analytical formulations for the elastic reflection coefficient and the angular distribution of elastically backscattered electrons. Results calculated based on these formulations were in good agreement with those using Monte Carlo simulations and experimental data.

1. Introduction

Analysis of the elastic peak of electrons escaped from solid surfaces is important in surface sensitive electron spectroscopies [1, 2]. This analysis of elastically backscattered electrons in elastic peak electron spectroscopy (EPES) [3–8], for instance, has attracted much attention since it extracted important information on the properties of solid surfaces. A common application of EPES is the determination of electron inelastic mean free paths (IMFPs) in solids [9, 10]. The features of elastic peak electron spectra are characterized by the elastic reflection coefficient and the angular distribution of elastically backscattered electrons.

Several theoretical approaches are available to evaluate the elastic peak electron spectra. The simplest one is the single elastic scattering model proposed by Jablonski [9]. He assumed that electrons backscattered from solid surfaces underwent a large-angle elastic scattering. Other approaches assumed the multiple-scattering process [11, 12]. For instance, Schilling and Webb [11] proposed a fitting model to estimate the angular distribution of elastically backscattered electrons using the forward scattering approximation for two elastic scatterings and a uniform scattering assumption for more scatterings. For low-energy electrons, the forward scattering approximation

may lead to a significant error due to the broad angular distribution of electron differential elastic cross sections. In addition, the uniform scattering assumption for escape angles does not seem plausible. Tofterup [12] derived the angular distribution of elastically backscattered electrons using the Boltzmann transport equation and the P_1 -approximation. This approximation is, however, inadequate to describe the contribution from only a few elastic scatterings. Therefore, Tofterup's results failed to account for the experimentally measured angular distributions [13–17]. Because of theoretical deficiencies, a more realistic and accurate model for the calculation of the angular distribution of elastically backscattered electrons is needed.

In this work, we first applied the Monte Carlo (MC) method to analyse the elastic reflection coefficient and the angular distribution of electrons elastically backscattered from solid surfaces. Results indicated that elastically backscattered electrons were substantially contributed by only a few scatterings with a single scattering event accounting for approximately half of these electrons. Thus, neither the multiple scattering model nor the single scattering model was sufficient to describe the angular distribution. To improve these models, we evaluated the contribution from one, two and three scatterings exactly and higher scatterings by the P_1 -approximation. This approach allowed us to derive analytical formulations for the elastic reflection coefficient and the angular distribution of elastically backscattered electrons. Since we assumed

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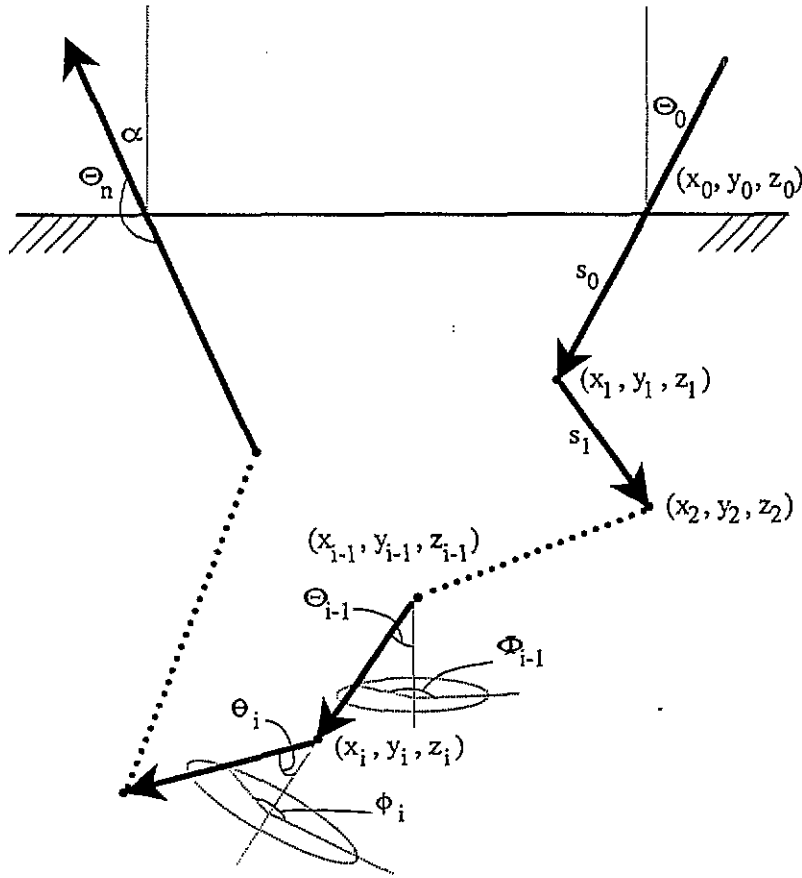


Figure 1. A diagram of the 'zigzag' trajectory of an electron inside the solid before the electron is elastically backscattered from the surface.

an isotropic and homogeneous medium for electron elastic scatterings, the present work was valid for amorphous and polycrystalline solids only. Results calculated based on these formulations were in good agreement with those using MC simulations and experimental data.

2. Monte Carlo simulations and analyses

The MC method has been widely employed to study electron transport properties in solids [18–21]. Here we apply this method to compute the elastic reflection coefficient and the angular distribution of electrons elastically backscattered from solid surfaces for later use. In MC simulations, electron elastic scatterings are described by the probability density function.

$$P(\theta) = \frac{2\pi \sin \theta \, d\sigma_e}{\sigma_e \, d\Omega} \quad (1)$$

where θ is the polar scattering angle, $d\Omega = 2\pi \sin \theta \, d\theta \, d\phi$ is the differential solid angle around (θ, ϕ) , ϕ is the azimuthal scattering angle, $d\sigma_e/d\Omega$ is the elastic scattering differential cross section, and $\sigma_e = \int (d\sigma_e/d\Omega) \, d\Omega$ is the elastic scattering cross section. The probability density function of the electron free path against elastic scatterings s is described by the Poisson stochastic process. It is given by

$$P(s) = \frac{1}{\lambda_e} \exp\left(-\frac{s}{\lambda_e}\right) \quad (2)$$

where $\lambda_e = (N\sigma_e)^{-1}$ is the electron elastic mean free path and N is the atomic density in solids. Further, the probability of electrons traversing a path length R without inelastic interactions is proportional to $\exp(-R/\lambda_i)$, where λ_i is the electron IMFP.

As illustrated in figure 1, electron trajectories in a solid may be traced by recording values of s , θ and ϕ repeatedly until all electrons leave the solid. For each step in MC simulations, electron position at the $(i + 1)$ th scattering, i.e. x_{i+1} , y_{i+1} and z_{i+1} , may be determined from its previous position at the i th scattering according to

$$x_{i+1} = x_i + s_i \sin \Theta_i \cos \Phi_i \quad (3)$$

$$y_{i+1} = y_i + s_i \sin \Theta_i \sin \Phi_i \quad (4)$$

and

$$z_{i+1} = z_i + s_i \sin \Theta_i \quad (5)$$

where Θ_i is the angle between electron velocity and the surface normal after the i th scattering and Φ_i is the angle between electron velocity projection on the surface plane and the x -axis. Furthermore, Θ_i and Φ_i may be related to scattering angles through

$$\cos \Theta_i = \cos \Theta_{i-1} \cos \theta_i - \sin \Theta_{i-1} \sin \theta_i \cos \phi_i \quad (6)$$

$$\cos \Phi_i = [\cos \Phi_{i-1} (\sin \theta_i \cos \phi_i \cos \Theta_{i-1} + \cos \theta_i \sin \Theta_{i-1}) - \sin \Phi_{i-1} \sin \theta_i \sin \phi_i] / [\sin \Theta_i] \quad (7)$$

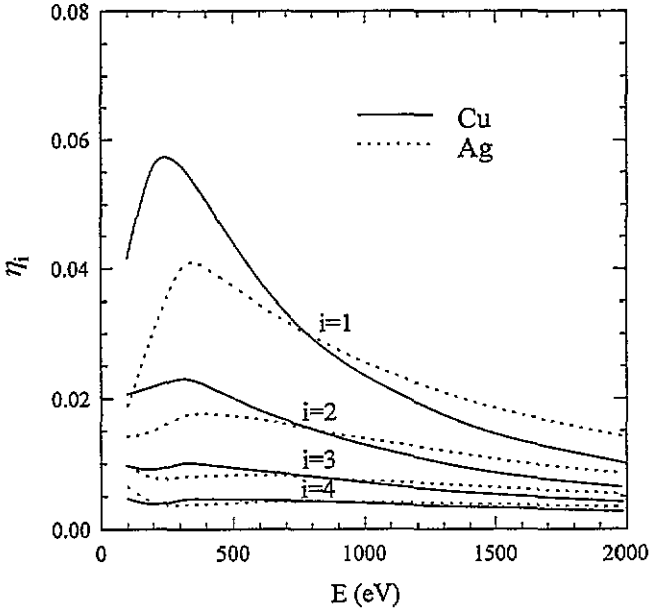


Figure 2. MC results of the elastic reflection coefficient contributed by one to four elastic scatterings for electrons elastically backscattered from Cu and Ag surfaces as a function of electron energy.

and

$$\sin \Phi_i = [\sin \Phi_{i-1} (\sin \theta_i \cos \phi_i \cos \Theta_{i-1} + \cos \theta_i \sin \Theta_{i-1}) + \cos \Phi_{i-1} \sin \theta_i \sin \phi_i] / [\sin \Theta_i]. \quad (8)$$

Note that Θ_0 and Θ_n ($n \geq 1$) represent the incident and exist angles of electrons with respect to the surface normal respectively and $\alpha = \pi - \Theta_n$ is the escape angle with respect to surface plane, shown in figure 1.

In this work, we first apply MC simulations to compute the elastic reflection coefficient and the angular distribution of electrons elastically backscattered from Cu and Ag surfaces. In order to compare MC results with experimental data, we choose incident electrons normal to the surface, i.e. $\Theta_0 = 0$. Input data on electron IMFPs are calculated using the extended Drude dielectric function [22] for volume excitations of valence electrons. Data on electron elastic scattering differential cross sections are obtained using the partial wave expansion method with the Hartree-Fock-Wigner-Seitz (HFWS) scattering potential [23]. Figure 2 shows MC results of the elastic reflection coefficient contributed by one to four elastic scatterings for electrons elastically backscattered from Cu (full curves) and Ag (broken curves) surfaces as a function of electron energy. Here η_i denotes the elastic reflection coefficient contributed by i scatterings for backscattered electrons with escape angles between 0 and $\pi/2$. It is seen that this coefficient drops rapidly with an increasing number of elastic scatterings—as electrons encountered more elastic scatterings in the solid, they travelled longer path lengths and thus were further attenuated.

Figure 3 shows MC results on η_{i+1}/η_i as a function of the number of elastic scatterings for electrons elastically backscattered from Cu (full circles) and Ag (open circles) surfaces. The full and broken curves are interpolation results for easy inspection of the dependence of this

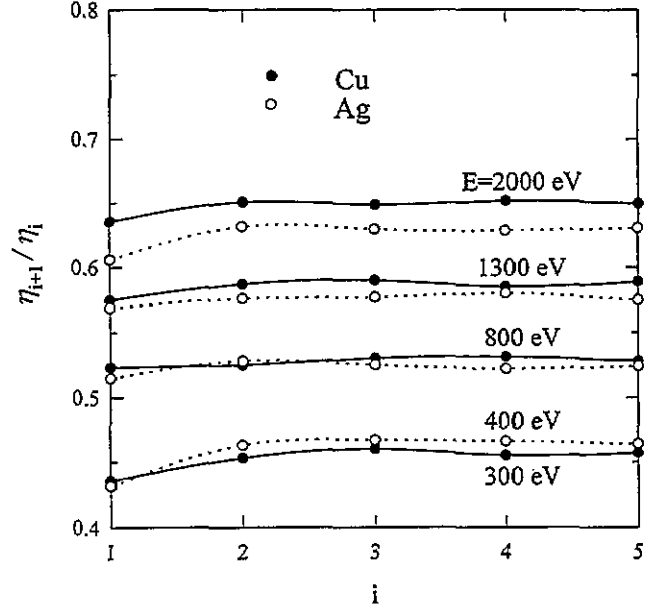


Figure 3. MC results of η_{i+1}/η_i corresponding to different electron energies for electrons elastically backscattered from Cu and Ag surfaces as a function of the number of elastic scatterings. The data points are MC results. The curves are interpolation results to guide the eye.

function on the number of scatterings. The results indicate that

$$\frac{\eta_3}{\eta_2} \approx \frac{\eta_4}{\eta_3} \approx \frac{\eta_5}{\eta_4} \approx \dots \quad (9)$$

These relations confirm the prediction of Schilling and Webb [17] and Schmid [24]. Since $\eta = \sum_{i=1}^{\infty} \eta_i$, we let

$$\eta = \left(\sum_{i=1}^3 \eta_i \right) + \frac{\eta_3^2}{\eta_2 - \eta_3} \quad (10)$$

where the last term in the above equation is the result of an infinite geometric series representing the sum over all fourth- and higher-order scattering terms. Equation (10) suggests that the elastic reflection coefficient may be determined by its individual contributions from one, two and three elastic scatterings.

To study the angular distribution of elastically backscattered electrons, we consider the elastic reflection coefficient contributed by i scatterings for backscattered electrons with escape angles between α and $\alpha + d\alpha$, i.e. $\eta_i(\alpha, \alpha + d\alpha)$. Note that $\eta_i \equiv \eta_i(0, \pi/2)$. The normalized angular distribution of elastically backscattered electrons contributed by i scatterings is defined as

$$I_i(\alpha) d\Omega_\alpha = \frac{\eta_i(\alpha, \alpha + d\alpha)}{\eta_i} \quad (11)$$

where $d\Omega_\alpha = 2\pi \sin \alpha d\alpha$ is the differential solid angle around α . If we adopt the P_1 -approximation, this normalized angular distribution becomes [12]

$$I^{P_1}(\alpha) = \frac{1}{2\pi} \left(\frac{3}{2} \cos^2 \alpha + \cos \alpha \right). \quad (12)$$

The histograms in figure 4 are MC results on the normalized angular distribution contributed by one to four scatterings,

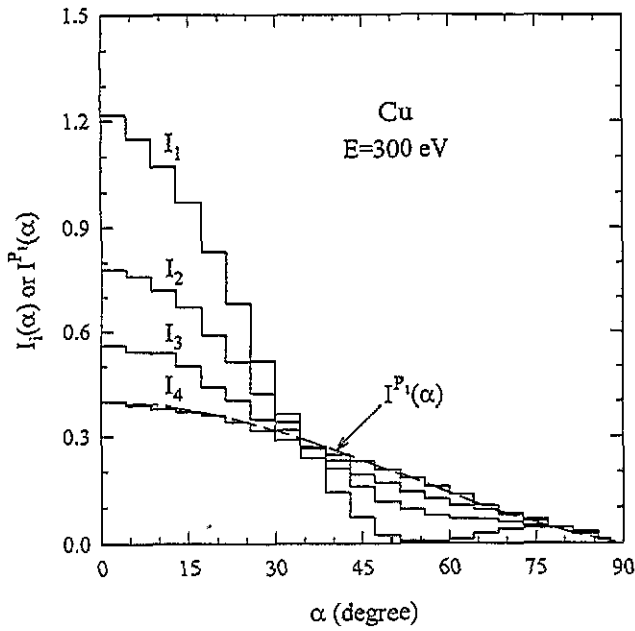


Figure 4. A plot of the normalized angular distribution contributed by one to four scatterings $I_i(\alpha)$ ($i = 1, 2, 3, 4$) using MC simulations (histograms) and using the P_1 -approximation (broken curve) for 300 eV electrons elastically backscattered from a Cu surface as a function of escape angle.

i.e. $I_i(\alpha)$ ($i = 1, 2, 3, 4$) given in equation (11), for 300 eV electrons elastically backscattered from Cu surface as a function of escape angle. The broken curve shows the results of $I^{P_1}(\alpha)$ in equation (12) under the P_1 -approximation. A similar plot for 400 eV electrons elastically backscattered from Ag surface is shown in figure 5. It can be seen that $I_4(\alpha)$ closely resembles $I^{P_1}(\alpha)$ at all escape angles. This resemblance holds true for all $I_i(\alpha)$ with $i \geq 4$. Hence it suggests that the angular distribution of elastically backscattered electrons may be determined by a combination of the P_1 -approximation for $i \geq 4$ and exact formulations for $i = 1, 2$ and 3 .

3. Theory

The probability density function of the electron elastic free path s_0 without inelastic interactions is given by the Poisson stochastic process as

$$P_s(s_0) ds_0 = \frac{1}{\lambda_e} \exp\left(-\frac{s_0}{\lambda_e}\right) \exp\left(-\frac{s_0}{\lambda_i}\right) ds_0 \quad (13)$$

where the first and second exponentials represent the probability of electrons surviving elastic and inelastic interactions over the distance s_0 respectively. Considering the single elastic scattering event, the probability density function of electrons elastically backscattered into the differential solid angle $d\Omega_1$ and (θ_1, ϕ_1) is given by

$$P_\Omega(\theta_1, \phi_1) d\Omega_1 = \frac{1}{\sigma_e} \left(\frac{d\sigma}{d\Omega}\right)_{\theta_1} d\Omega_1 \quad (14)$$

where $(d\sigma/d\Omega)_{\theta_1}$ is the electron elastic differential cross section at polar scattering angle θ_1 , $d\Omega_1 = \sin\theta_1 d\theta_1 d\phi_1$,

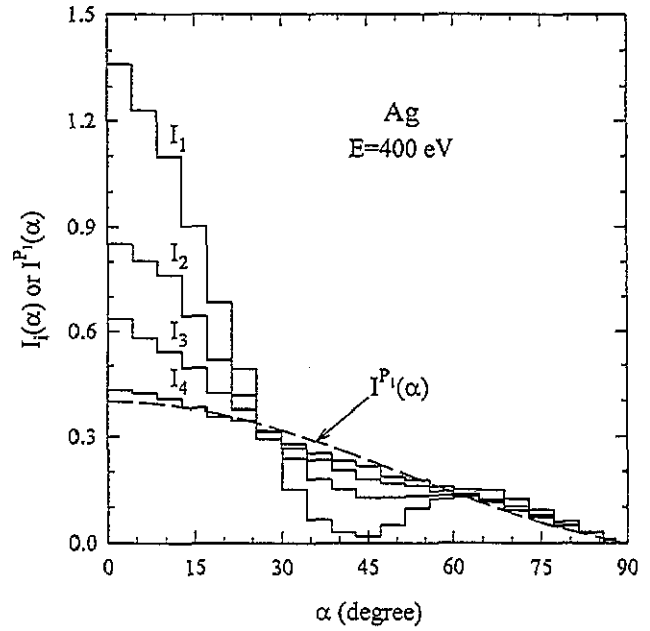


Figure 5. A plot of the normalized angular distribution contributed by one to four scatterings $I_i(\alpha)$ ($i = 1, 2, 3, 4$) using MC simulations (histograms) and using the P_1 -approximation (broken curve) for 400 eV electrons elastically backscattered from a Ag surface as a function of escape angle.

and ϕ_1 is the azimuthal scattering angle. Furthermore, the probability density function for electrons to leave the solid with an exit angle Θ_1 after one elastic scattering is given by

$$P_z(z_1) = \exp\left(-\frac{z_1}{\lambda_e \cos(\pi - \Theta_1)}\right) \times \exp\left(-\frac{z_1}{\lambda_i \cos(\pi - \Theta_1)}\right) \quad (15)$$

where $z_1 = s_0 \cos\Theta_0$ is the electron depth at the point of elastic scattering and Θ_1 is determined from Θ_0 , θ_1 and ϕ_1 through equation (6).

The elastic reflection coefficient contributed by a single elastic scattering for backscattered electrons with escape angles between α and $\alpha + d\alpha$ may be obtained using

$$\eta_1(\alpha, \alpha + d\alpha) = \int \int_0^\infty P_s(s_0) P_\Omega(\theta_1, \phi_1) P_z(s_0 \cos\Theta_0) \times G(\Theta_1) ds_0 d\Omega_1 \quad (16)$$

where

$$G(\Theta_1) = u(\pi - \Theta_1 - \alpha)u(\alpha + d\alpha - \pi + \Theta_1) \quad (17)$$

and

$$u(x) = \begin{cases} 1 & \text{for } x > 0 \\ 0 & \text{for } x < 0. \end{cases} \quad (18)$$

Carrying out the integral in equation (16) over s_0 , we find

$$\eta_1(\alpha, \alpha + d\alpha) = N\lambda_r \int \left(\frac{d\sigma}{d\Omega}\right)_{\theta_1} \frac{G(\Theta_1) \cos\Theta_1}{\cos\Theta_1 - \cos\Theta_0} d\Omega_1 \quad (19)$$

where λ_r is the electron total mean free path determined by

$$\lambda_r^{-1} = \lambda_e^{-1} + \lambda_i^{-1}. \quad (20)$$

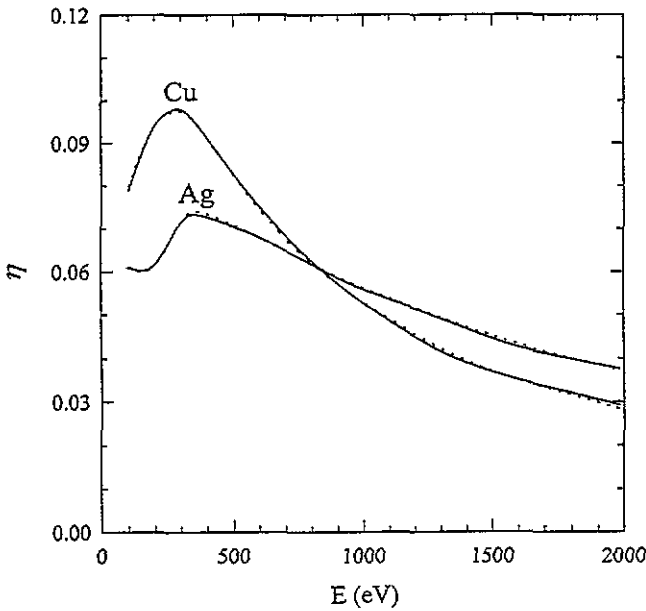


Figure 6. A comparison of the elastic reflection coefficient calculated using present formulations (full curves) and MC simulations (broken curves) for electrons elastically backscattered from Cu and Ag surfaces as a function of electron energy.

Equation (19) is analogous to the one-scattering formula of Jablonski [9] for normally incident electrons. The leading factor in equation (19), i.e. $N\lambda_i$, is, however, different from that used by Jablonski, i.e. $N\lambda_i$. This is because we included elastic scatterings in the probability density function of equation (13).

Similarly, we can derive the elastic reflection coefficient contributed by two elastic scatterings. This coefficient for backscattered electrons with escape angles between α and $\alpha + d\alpha$ may be expressed as

$$\begin{aligned} \eta_2(\alpha, \alpha + d\alpha) = & (N\lambda_i)^2 \int \left(\frac{d\sigma}{d\Omega} \right)_{\Theta_2} d\Omega_2 \\ & \times \int \left(\frac{d\sigma}{d\Omega} \right)_{\Theta_1} \frac{G(\Theta_2) \cos \Theta_2}{\cos \Theta_2 - \cos \Theta_1} \\ & \times \left[\frac{F(\Theta_1) \cos \Theta_2}{\cos \Theta_2 - \cos \Theta_0} \right. \\ & \left. + B(\Theta_1) \left(\frac{\cos \Theta_2}{\cos \Theta_2 - \cos \Theta_0} - \frac{\cos \Theta_1}{\cos \Theta_1 - \cos \Theta_0} \right) \right] d\Omega_1 \end{aligned} \quad (21)$$

where Θ_2 is the exit angle, $F(x) = u(x)u(\pi/2 - x)$ and $B(x) = u(x - \pi/2)u(\pi - x)$. The first term inside the square brackets of equation (21) corresponds to electrons moving away from the surface after the first scattering, i.e. $0 < \Theta_1 < \pi/2$, whereas the second term corresponds to electrons moving toward the surface after the first scattering, i.e. $\pi/2 < \Theta_1 < \pi$.

A similar derivation of the elastic reflection coefficient contributed by three elastic scatterings gives

$$\begin{aligned} \eta_3(\alpha, \alpha + d\alpha) = & (N\lambda_i)^3 \int \left(\frac{d\sigma}{d\Omega} \right)_{\Theta_3} d\Omega_3 \\ & \times \int \left(\frac{d\sigma}{d\Omega} \right)_{\Theta_2} d\Omega_2 \int \left(\frac{d\sigma}{d\Omega} \right)_{\Theta_1} \frac{G(\Theta_3) \cos \Theta_3}{\cos \Theta_3 - \cos \Theta_2} \end{aligned}$$

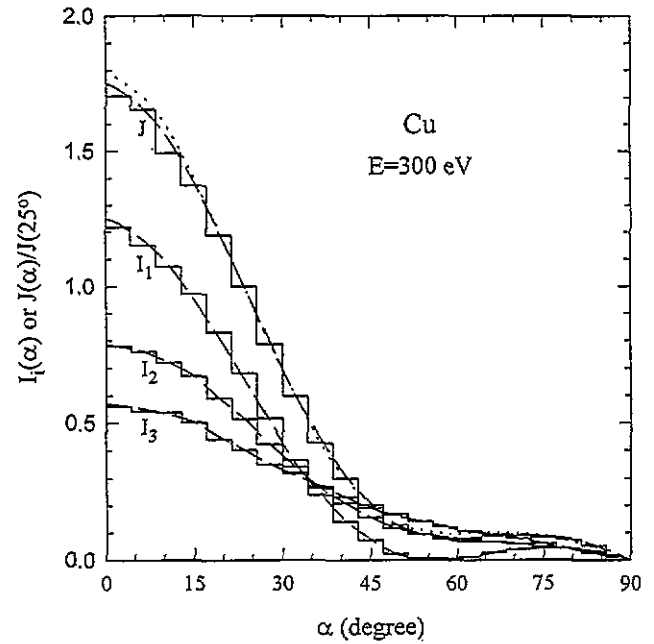


Figure 7. A plot of the normalized angular distribution contributed by one to three scatterings $I_i(\alpha)$ ($i = 1, 2, 3$) using present formulations (broken curves) and MC simulations (histograms) along with the angular distribution $J(\alpha)$ using the present formulations (broken curve), MC simulations (histogram) and experimental data (dotted curve) for 300 eV electrons elastically backscattered from a Cu surface as a function of escape angle. The angular distribution results are normalized to the backscattered electron intensity at an escape of 25° .

$$\begin{aligned} & \times \left\{ F(\Theta_1) \left(\frac{\cos \Theta_3}{\cos \Theta_3 - \cos \Theta_0} \frac{\cos \Theta_3}{\cos \Theta_3 - \cos \Theta_1} \right. \right. \\ & \left. \left. - \frac{B(\Theta_2) \cos \Theta_2}{\cos \Theta_2 - \cos \Theta_0} \frac{\cos \Theta_2}{\cos \Theta_2 - \cos \Theta_1} \right) \right. \\ & \left. + B(\Theta_1) \left[\left(\frac{\cos \Theta_3}{\cos \Theta_3 - \cos \Theta_0} - \frac{\cos \Theta_1}{\cos \Theta_1 - \cos \Theta_0} \right) \right. \right. \\ & \left. \times \frac{\cos \Theta_3}{\cos \Theta_3 - \cos \Theta_1} - B(\Theta_2) \right. \\ & \left. \times \left(\frac{\cos \Theta_2}{\cos \Theta_2 - \cos \Theta_0} - \frac{\cos \Theta_1}{\cos \Theta_1 - \cos \Theta_0} \right) \right. \\ & \left. \times \frac{\cos \Theta_2}{\cos \Theta_2 - \cos \Theta_1} \right] \left. \right\} d\Omega_1 \end{aligned} \quad (22)$$

where Θ_3 is the exit angle.

Given the angular distribution $J(\alpha) = \sum_{i=1}^{\infty} \eta_i I_i(\alpha)$ we can write

$$J(\alpha) = \sum_{i=1}^3 \eta_i I_i(\alpha) + \frac{\eta_3^2}{\eta_2 - \eta_3} I^{P_1}(\alpha) \quad (23)$$

where η_i and $I_i(\alpha)$ ($i = 1, 2, 3$) are determined using equation (11), (19), (21) and (22) and $I^{P_1}(\alpha)$ using equation (12). Finally, the elastic reflection coefficient of electrons backscattered into the acceptance angles between α_1 and α_2 may be calculated from

$$\eta(\alpha_1, \alpha_2) = \int_{\alpha_1}^{\alpha_2} J(\alpha) d\Omega_\alpha. \quad (24)$$

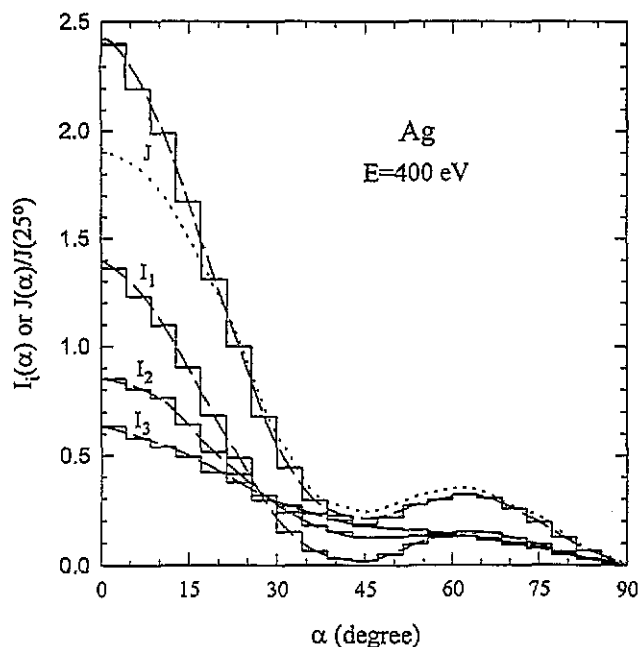


Figure 8. A plot of the normalized angular distribution contributed by one to three scatterings $I_i(\alpha)$ ($i = 1, 2, 3$) using present formulations (broken curves) and MC simulations (histograms) along with the angular distribution $J(\alpha)$ using the present formulations (broken curve), MC simulations (histogram) and experimental data (dotted curve) for 400 eV electrons elastically backscattered from a Ag surface as a function of escape angle. The angular distribution results are normalized to the backscattered electron intensity at an escape of 25° .

4. Results and discussion

Figure 6 shows a plot of the elastic reflection coefficient for electrons elastically backscattered from Cu and Ag surfaces as a function of electron energy. Results of present formulations (full curves) are in close agreement with those of MC simulations (broken curves). Figure 7 shows a plot of the normalized angular distribution contributed by one, two and three elastic scatterings $I_i(\alpha)$ ($i = 1, 2, 3$) and the angular distribution $J(\alpha)$ as a function of escape angle for 300 eV electrons backscattered from a Cu surface. The broken curves, histograms and dotted curve are, respectively, results of present formulations, MC simulations and experimental measurements [14]. Excellent agreement is found for the normalized angular distribution contributed by one to three scatterings between present calculations and MC simulations. The comparison for the angular distribution between present calculations and experimental data is also very good. Note that the angular distribution is normalized to backscattered electron intensity at an escape angle of 25° . Figure 8 shows a similar plot for 400 eV electrons backscattered from a Ag surface. It is again seen that excellent agreement is found between present calculations and MC simulations. The deviation of theoretical results from experimental data [14] on the angular distribution at small escape angles is due to the fact that experimental data at these angles (0 – 25°) were not actually measured but rather extrapolated from results at larger angles.

5. Conclusions

We have derived formulations for the elastic reflection coefficient and the angular distribution of electrons elastically backscattered from solid surfaces. Treating the first three scattering events exactly and applying the P_1 -approximation for higher scattering events, we were able to resolve the deficiencies involved in the single scattering model and the multiple scattering model. Results of present formulations are in good agreement with those of MC simulations and experimental data.

In this work, only volume excitations by electrons are considered. Surface excitations influence the elastic reflection coefficient, especially for low-energy electrons. A detailed discussion about this effect is presented elsewhere [25,26]. Since surface excitations contribute negligibly to the angular distribution of elastically backscattered electrons, we omit this contribution in this work.

Finally, it is noted that the present theory is applicable to amorphous and polycrystalline solids only. The diffraction effect focuses the electron beam in a certain direction depending on the orientation of single crystals. This effect is diminished in non-crystalline solids due to random elastic scatterings which defocus the electron beam [27,28].

Acknowledgment

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