Inelastic High-Energy Electron Spectra from Plane-Parallel Layer-Nonhomogeneous Surfaces

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We consider inelastic backscattering of electrons with initial energy of tens and hundreds of keV by plane-parallel homogeneous and sandwiched targets. Basing on the invariance principle, we find expressions that describe the dynamics of the changes in the energy spectra of electrons reflected into a given solid angle that occur with increase of the thickness of films of different materials on substrates of finite and infinite thickness. We substantiate a procedure of linearizing the equations for the reflection function obtained by the method of invariant imbedding. We obtain an analytical solution of linearized equations in the form of a series in Legendre polynomials. A comparison with experimental data shows that the theory developed gives an adequate description of the process of electron backscattering.

Introduction

Electron spectroscopy is one of the most efficient methods to analyse solid surfaces [1]. The present paper deals with the interpretation of the signals observed in the spectra of electrons reflected nonelastically within a given body angle (in the *doubly differential spectra of nonelastically reflected electrons, DD SNRE*). Such experiments are done with an electron gun and an energy analyzer. However, in constrast to Auger spectroscopy, SNRE measurements do not require such a high resolution; the signal is large and is measured with a Faraday cup (instead of a second electron multiplier used in Auger spectroscopy). The information one can obtain by decoding the SNRE comes from depths of the target of the order of the electron transport range $l_{\mathrm{tr}} \simeq 2\,\langle\chi^2\rangle$, where $\langle\chi^2\rangle$ is the mean square scattering angle [2].

As will be shown, from this information one can make conclusions about the composition and depth profiles of the target.

The process of electron scattering is described by an integro-differential transport equation [2, 3] for which the solutions for various boundary conditions are known in many cases [2–12]. A consistent solution of the boundary problem about the reflection of electrons from a half-infinite homogeneous medium has been obtained in [6] by using the method of invariant imbedding.

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In the present paper, basing on invariance principles, we find the DD SNRE in the case where electrons are scattered by planar layers and by layer-nonhomogeneous surfaces. We substantiate a linearisation procedure that leads to an analytical solution of the Dashen equation [6], which describes the DD SNRE in the case of half-infinite homogeneous targets.

1. Energy Spectra of Electrons Reflected from Layer-Nonhomogeneous Targets

A detailed derivation of the formulae we use in this section and their analysis is presented in [11, 12], so here we will explain only the main idea of this derivation.

Let us define the reflection r and transmission t functions of electrons by the expressions

$$I_{\mathrm{r}}(\boldsymbol{q}, E) = \int_0^{-1} \mathrm{d}\cos\vartheta_0 \cdot \int_0^{2\pi} \mathrm{d}\phi \cdot \int_E^{E_0} \mathrm{d}E_0 \cdot r(x, \boldsymbol{q}_0, E_0, \boldsymbol{q}, E) \cdot I_0(\boldsymbol{q}_0, E_0)\,, \quad (1)$$

$$I_{\mathrm{t}}(\boldsymbol{q}, E) = \int_0^{1} \mathrm{d}\cos\vartheta_0 \cdot \int_0^{2\pi} \mathrm{d}\phi \cdot \int_E^{E_0} \mathrm{d}E_0 \cdot t(x, \boldsymbol{q}_0, E_0, \boldsymbol{q}, E) \cdot I_0(\boldsymbol{q}_0, E_0)\,. \quad (2)$$

Here I_0, I_{r}, and I_{t} are respectively the incident electron flux, the reflected electron flux, and the electron flux that has passed through the layer, $\boldsymbol{q} = \{\theta, \phi\}$ is a unit vector in the direction of the normal to the surface into the depth of the layer and x_i is the thickness of the i-th layer.

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To reduce the equations, in addition to the detailed form of (1) and (2) we will also use the matrix notations

$$I_{r,t} = r, t, I_0 .$$

R will denote the function of reflection from a half-infinite medium.

Let us find an analytical expression for the reflection function of a layer made of material 1 with thickness x_1 spray-covered with a film made of material 2 with thickness x_2, i.e.

$$r_{12}(x_2+x_1; \boldsymbol{q}_0, E_0, \boldsymbol{q}, E) . \tag{3}$$

The function r_{12} is given by a formula that follows from the definitions (1) and (2):

$$r_{12}(x_2+x_1) = r_2(x_2)+t_2(x_2)\, r_1(x_1)\, t_2(x_2) + t_2(x_2)\, r_1(x_1)\, r_2(x_2)\, r_1(x_1)\, t_2(x_2)+ \ldots . \tag{4}$$

Let us write down this formula for a target made of material 2 with thickness x_1+x_2 in which a boundary is mentally drawn at the depth x_2:

$$r_2(x_2+x_1)=r_2(x_2)+t_2(x_2)\, r_2(x_1)\, t_2(x_2) + t_2(x_2)\, r_2(x_1)\, r_2(x_2)\, r_2(x_1)\, t_2(x_2)+ \ldots . \tag{5}$$

By subtracting (5) from (4), we obtain

$$\begin{aligned} r_{12}(x_2+x_1) &= r_2(x_2+x_1) \\ &\quad + t_2(x_2)\cdot [r_1(x_1)-r_2(x_1)]\cdot t_2(x_2) \\ &\quad + t_2(x_2)\cdot [r_1(x_1)\, r_2(x_2)\, r_1(x_1) \\ &\quad - r_2(x_1)\, r_2(x_2)\, r_2(x_1)]\cdot t_2(x_2)+ \ldots . \end{aligned} \tag{6}$$

By neglecting, in accordance with [11, 12], the alternating-sign series that begins from the third term on the r.h.s. of (6), we obtain

$$r_{12}(x_2+x_1) = r_2(x_2+x_1)+t_2(x_2)\cdot [r_1(x_1)-r_2(x_1)]\cdot t_2(x_2) . \tag{7}$$

The detailed form of this formula is

$$\begin{aligned} r_{12}(x_2+x_1, \boldsymbol{q}_0, \boldsymbol{q}, E_0-E) &= r_2(x_2+x_1, \boldsymbol{q}_0, \boldsymbol{q}, E_0-E) \\ &\quad + \int_E^{E_0} \mathrm{d}E_1 \int_E^{E_0} \mathrm{d}E_2 \int_0^1 \frac{1}{\mu_2}\, \mathrm{d}\mu_2 \cdot t_2(x_2, \boldsymbol{q}, \boldsymbol{q}_1, E_1-E) \\ &\quad \cdot [r_1(x_1, \boldsymbol{q}_1, \boldsymbol{q}_2, E_2-E_1)-r_2(x_1, \boldsymbol{q}_1, \boldsymbol{q}_2, E_2-E_1)] \\ &\quad \cdot t_2(x_2, \boldsymbol{q}_2, \boldsymbol{q}_1, E_0-E_2), \end{aligned} \tag{8}$$

where $\mu_i = \cos\theta_i$.

Formulae (7) and (8) enable one to solve the problem of electron reflection from a layer-nonhomogeneous plane-parallel structure if one knows the laws of scattering from homogeneous layers.

The problem of calculating the transmission function t for homogeneous layers was solved in a number of papers [7–12]. The double differential transmission function $t(x, \boldsymbol{q}_0, \boldsymbol{q}, E_0-E)$ has been found in [11] by the method of invariant imbedding in the case of scattering by a thin layer.

Good agreement with experimental data is reached by an approach based on the assumption that the transmission function can be factorized with respect to angular and energy variables:

$$t(x, \boldsymbol{q}_0, \boldsymbol{q}, E_0-E) = t_E(x, E_0-E)\, \mu_0\, \mu\, t_q(x, \boldsymbol{q}_0, \boldsymbol{q})\, f(x) . \tag{9}$$

The function $t_E(x, E_0-E)$ is calculated within the Landau theory [7]. In those cases where electrons hit the layer at an angle close to normal, the function $t(x, \boldsymbol{q}_0, \boldsymbol{q})$ is calculated according to the Moliere formula [9].

As was shown in [11], the supposition that the total flux is conserved, under which solutions t_E and t_q have been found, violates the invariance principle. For this reason the r.h.s. of (9) contains a factor $f(x)$ that makes allowance for the dissipation of the electron flux. The function $f(x)$ is calculated from the Bugger law

$$f(x) = \exp[-x/I_0] .$$

The best agreement with the experimental data is given by I_0 presented in [13].

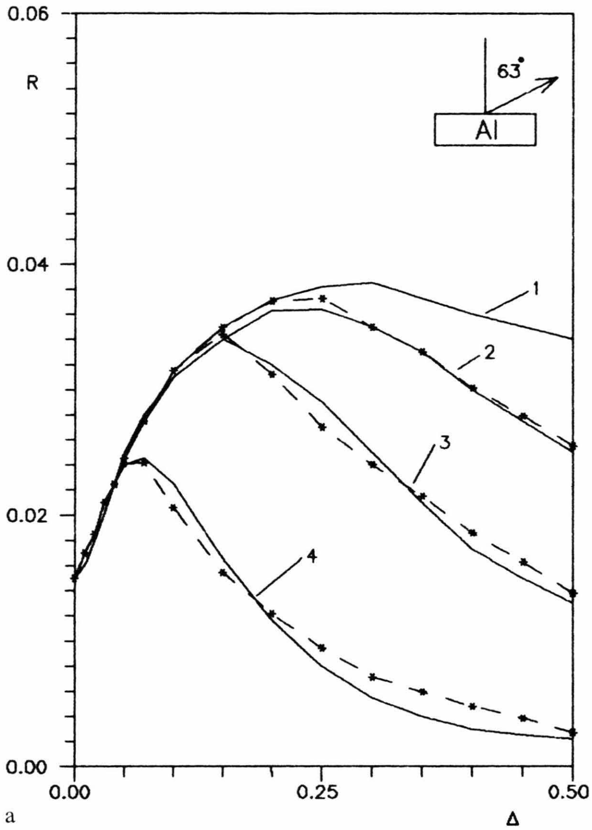
Let us use (4) to calculate the DD SNRE in the case where electrons are reflected by homogeneous layers of finite thickness, supposing that we know the reflection function for a half-infinite target R. In this case

$$r(x) = R - t(x)\, R\, t(x) . \tag{10}$$

The detailed form of (10) is reconstructed by analogy with (8). $t(x)$ is calculated by using (9).

The results of the calculation, presented in Fig. 1, are in good agreement with the experimental data of [14] in a wide range of scattering angles, $97^\circ \leq \vartheta \leq 147^\circ$, and values Z for the target, $13 \leq Z \leq 79$.

Formula (8) was used to calculate the DD SNRE in the case of a two-layer structure, viz. for Al film (material 2) with varying thickness placed upon an Au substrate (material 1) with thickness $x_1 = 600$ Å. The results of the calculation are shown in Figure 2. One clearly sees how the DD SNRE changes both qualitatively and quantitatively with variation of x_2 (the thickness of the Al film).



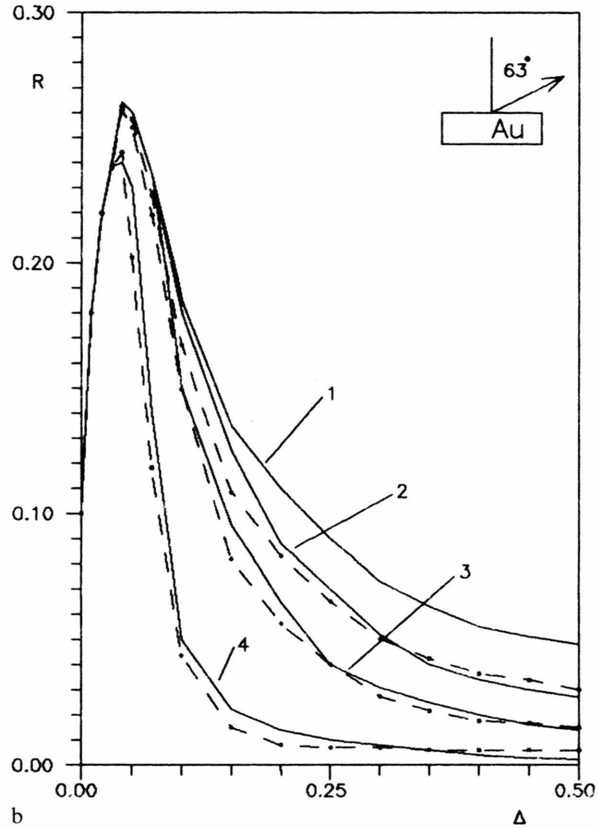


Fig. 1. Energy spectra of 25-keV electrons reflected from thin Al (a) and Au (b) layers with reduced thickness d (dashed lines, calculation according to (10); solid lines, experiment [14]). a) for Al. 1, Al target; 2, $d=440$ μg/cm²; 3, $d=310$ μg/cm²; 4, $d=160$ μg/cm². b) for Au. 1, Au target; 2, $d=340$ μg/cm²; 3, $d=245$ μg/cm²; 4, $d=120$ μg/cm².

Figure 2 indicates that the DD SNRE can be used efficiently for studying the growth dynamics of various films in situ. Unfortunately, we were not able to find experimental data adequate to the situation presented in Figure 2. The reliability of the dependences shown in this figure can, however, be concludes from the similarity of their calculcation to those which lead to the verified results shown in Figure 1.

2. Double Differential Spectra of Electrons Reflected Nonelastically from Homogeneous Half-Infinite Targets

In order to find the reflection function $R(\boldsymbol{q}_0, \boldsymbol{q}, E_0-E)$ that describes the DD SNRE, one has to solve the boundary problem for the transport equation. A consistent solution of the boundary problem according to the method of invariant imbedding leads to a nonlinear equation for the function $R(\boldsymbol{q}_0, \boldsymbol{q}, E_0-E)$ [6, 11, 12] which, in the matrix notations (3) has the form

$$\begin{aligned}
&[\sigma(E_0)\,\mu_0^{-1}+\sigma(E)\,\mu^{-1}]\cdot R(\boldsymbol{p}_0,\boldsymbol{p})\\
&\quad=\omega_{\mathrm{el}}(E_0,\boldsymbol{q}_0,-\boldsymbol{q})+\omega(E_0,\boldsymbol{p}_0,\boldsymbol{p}_1)\cdot R(\boldsymbol{p}_1,\boldsymbol{p})\\
&\qquad+R(\boldsymbol{p}_0,\boldsymbol{p}_1)\cdot\omega(E_1,\boldsymbol{p}_1,\boldsymbol{p})\\
&\qquad+R(\boldsymbol{p}_0,\boldsymbol{p}_1)\,\omega_{\mathrm{el}}(E_1,\boldsymbol{q}_1,-\boldsymbol{q}_2)\cdot R(\boldsymbol{p}_2,\boldsymbol{p})\,,
\end{aligned}\tag{11}$$

where $\sigma(E)=\sigma_{\mathrm{inel}}(E)+\sigma_{\mathrm{el}}(E)$ is the total scattering cross-section, σ_{inel} and σ_{el} are the total elastic and inelastic cross-sections and ω denotes the corresponding differential cross-section

$$\begin{aligned}
\omega(E,\boldsymbol{p}_0,\boldsymbol{p})=\omega_{\mathrm{inel}}(E,E_0-E)\cdot\delta(\boldsymbol{q}-\boldsymbol{q}_0)\\
+\omega_{\mathrm{el}}(E,\boldsymbol{q}_0,\boldsymbol{q})\cdot\delta(E_0-E)\,.
\end{aligned}$$

The vector $\boldsymbol{p}=(\boldsymbol{q};E)$ denotes a set of angular and energy variables.

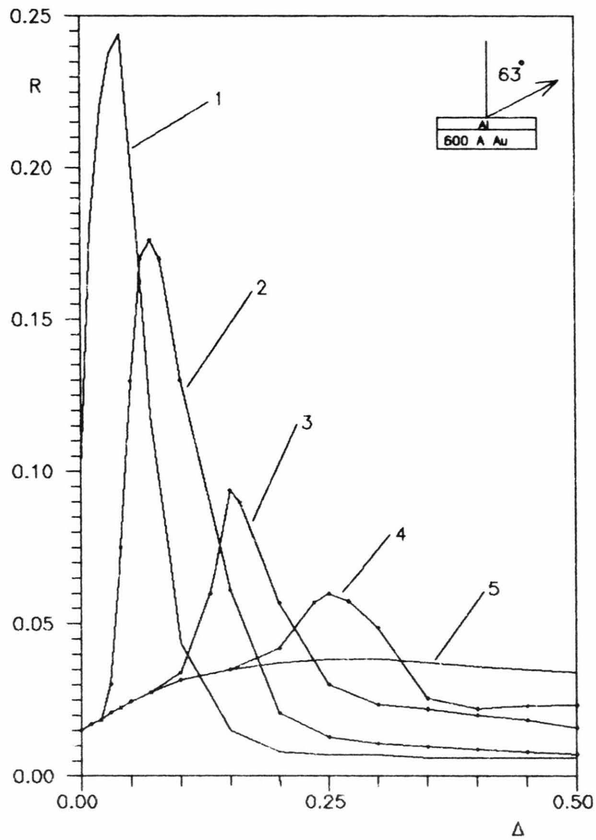


Fig. 2. Energy spectra of 25-keV electrons reflected from a two-layer system (Au layer 600 Å thick, Al layer of thickness d). 1, $d=0$ Å; 2, $d=3000$ Å; 3, $d=9000$ Å; 4, $d=15\,000$ Å; 5, $d=\infty$.

It is impossible to solve (11) for cross-sections ω of arbitrary form. It is, however, possible to construct an effective procedure for solving a shortened, or linearized, equation obtained from (11) by omitting the last, nonlinear, term. The linearization procedure is justified by the fact that the elastic scattering has a sharp maximum at small scattering angles. Whereas the processes described by the second and third terms on the r.h.s. of (11) are determined by small-angle scattering, the last term is associated with backward scattering, and, in the first approximation, its contribution can be neglected. Let us assume that the elastic cross-section is described by the Rutherford law

$$\omega_{\rm el}(E, \boldsymbol{q}_0, \boldsymbol{q}) = (Z e^2/2E)^2 \cdot (1 + 2\eta - (\boldsymbol{q}_0 \cdot \boldsymbol{q}))^2 . \quad (12)$$

Then the ratio of the contribution given by the nonlinear term to those of the second and the third terms on the r.h.s. of (11) is given by the expression

$$[1 + (\tfrac{\pi}{2} - \vartheta_0)^2/4\eta]^{-1} + [1 + (\tfrac{\pi}{2} - \vartheta)^2/4\eta]^{-1} = A . \quad (13)$$

From here it follows that the linearization procedure, which is valid for $A \ll 1$, is applicable within a wide range of incidence and scattering angles if only $\eta \ll 1$. The latter inequality holds in first approximation if the electron energy $E \gg 10 \cdot Z^{2/3}$ (E is measured in eV and Z is the atomic number of target material). We assume that

$$\sigma(E) = \sigma(E^*) = \text{const} , \quad (14)$$

where E^* is the mean energy of reflected electrons.

Basing on (14), one can suppose that R depends only on the energy difference $\Delta = E_0 - E$: $R(\boldsymbol{p}_0, \boldsymbol{p}) = R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)$. Considering what we have said above and regrouping the terms in (11) we obtain a linearized equation for the function $R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)$:

$$\begin{aligned}
&\omega_{\rm el}(E_0, \boldsymbol{q}_0, -\boldsymbol{q})\, E_0\, \delta(\Delta) \\
&+ \int_I^{\Delta - I} \mu_0^{-1}\, d\varepsilon\, \omega_{\rm inel}(E_0, \varepsilon)\, [R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta - \varepsilon) - R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)] \\
&+ \int_I^{\Delta - I} \mu^{-1}\, d\varepsilon\, \omega_{\rm inel}(E^*, \varepsilon)\, [R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta - \varepsilon) - R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)] \\
&+ \int d\boldsymbol{q}_1\, \omega_{\rm el}(E_0, \boldsymbol{q}_0, \boldsymbol{q}_1)\, [\mu_1^{-1} R(\boldsymbol{q}_1, \boldsymbol{q}, \Delta) - \mu_0^{-1} R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)] \\
&+ \int d\boldsymbol{q}_1\, \omega_{\rm el}(E^*, \boldsymbol{q}_1, \boldsymbol{q})\, [\mu_1^{-1} R(\boldsymbol{q}_0, \boldsymbol{q}_1, \Delta) - \mu^{-1} R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta)] \\
&= 0 .
\end{aligned}$$

By making the Laplace transformation with respect to Δ and using the diffusion approximation [2] in the elastic scattering channel, from (15) we come to the equation

$$\omega_{\rm el}(E_0, \boldsymbol{q}_0, -\boldsymbol{q})\, E_0 - W(s)\, r(\boldsymbol{q}_0, \boldsymbol{q}, s) + \frac{\langle \chi^2 \rangle}{4} [\nabla^2_{\vartheta_0, \phi_0} + \nabla^2_{\vartheta, \phi}]\, r(\boldsymbol{q}_0, \boldsymbol{q}, s) = 0 , \quad (16)$$

where

$$r(\boldsymbol{q}_0, \boldsymbol{q}, s) = (\mu_0^{-1} + \mu^{-1} \xi^{\alpha})\, R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta); \quad (17)$$

$$\xi = E_0/E^*,$$

$$W(s) = \int_I^{\Delta - I} d\varepsilon\, \omega_{\rm inel}(E_0, \varepsilon)\, [1 - \exp(-s\varepsilon)] . \quad (18)$$

Let us find the solution of (16) in the form of a series in spherical functions

$$r(\boldsymbol{q}_0, \boldsymbol{q}, s) = \sum_{l, m} r_l(s)\, \gamma_{lm}(\vartheta, \phi)\, \gamma^*_{l, m}(\vartheta_0, \phi_0) . \quad (19)$$

By analogy with (19), let us present the differential cross-section of elastic scattering as

$$\begin{aligned}
\omega_{\rm el}(E, \boldsymbol{q}_0, \boldsymbol{q}) &= \sum_{l, m} \omega_{\rm el}\, \gamma_{lm}(\boldsymbol{q})\, \gamma^*_{lm}(\boldsymbol{q}_0) \quad (20) \\
&= \sigma_{\rm el}\, \delta(\boldsymbol{q}_0 - \boldsymbol{q}) - \sum_{l, m} \sigma_{\rm el}\, \gamma_{lm}(\boldsymbol{q})\, \gamma^*_{lm}(\boldsymbol{q}_0) .
\end{aligned}$$

Since $\omega_{el}(E, \boldsymbol{q}_0, -\boldsymbol{q})$ is associated with backwards scattering of electrons, the term $\sigma_{el}\,\delta(\boldsymbol{q}_0-\boldsymbol{q})$ in (20) should be omitted. By virtue of the addition theorem of spherical functions we have

$$\sigma_l = 2\pi \int_0^{\pi} (1-P_l(\cos\theta)) \cdot \omega_{el}(E^*, \cos\theta)\, \mathrm{d}\cos\theta\,,$$

where

$$\cos\theta = \cos\vartheta\cos\vartheta_0 + \sin\vartheta\sin\vartheta_0\cos(\phi_0-\phi)$$

and

$$\sigma_1 = \sigma_{tr} = \langle\chi^2\rangle/2\,.$$

After substituting the expansions (19) and (20) into (16) and considering the above remarks, we obtain

$$\sigma_l E_0 + W(s)\cdot r_l(s) + (\sigma_{tr}/2)\, l(l+1)\, r_l(s) = 0\,. \tag{21}$$

The coefficients $r_l(\Delta)$ are found from (21) by reverse Laplace transformation

$$r_l(\Delta) = -\frac{\sigma_l E_0}{2\pi i}\int_C \exp(s\Delta)\,\mathrm{d}s/[W(s)+\sigma_{tr}\, l(l+1)]\,, \tag{22}$$

and with account of (17), (19), and (20), the solution of (15) in the diffusion approximation for the elastic scattering channel can be presented in the form

$$R(\boldsymbol{q}_0, \boldsymbol{q}, \Delta) = -\frac{\mu\mu_0}{\mu+\mu_0\,\xi^{\alpha}}\sum_{l,m}\frac{\sigma_l E_0}{2\pi i}\int_C \frac{\exp(s\Delta)\,\mathrm{d}s}{W(s)+\sigma_{tr}\, l(l+1)}\gamma_{lm}(\boldsymbol{q})\gamma^*_{lm}(\boldsymbol{q}_0)\,. \tag{23}$$

By directly substituting (23) into (11) one can check the validity of the preliminary estimates we made for the contribution of the nonlinear term.

With increase of the screening parameter and breaking down of condition (13), solution (23) can be regarded as an effective first approximation in the iteration procedure by means of which one finds the solution of the nonlinear equation (11) [15].

Figure 3 shows the DD SNRE for targets made of Ag and Pt, calculated according to (23). The elastic scattering channel was described by the Rutherford formula (12). The scattering cross section in the elastic channel was described by the approximate formula

$$\omega_{incl}(E_0, \varepsilon) = \frac{\pi Z e^2}{E_0}\varepsilon^{-2}.$$

A comparison between the calculation according to (23) and the experimental data [16] shows that the model developed gives an adequate description to the process of electron reflection within a wide range of scattering angles ($97^\circ \leq \vartheta \leq 147^\circ$) and values of Z for the target.

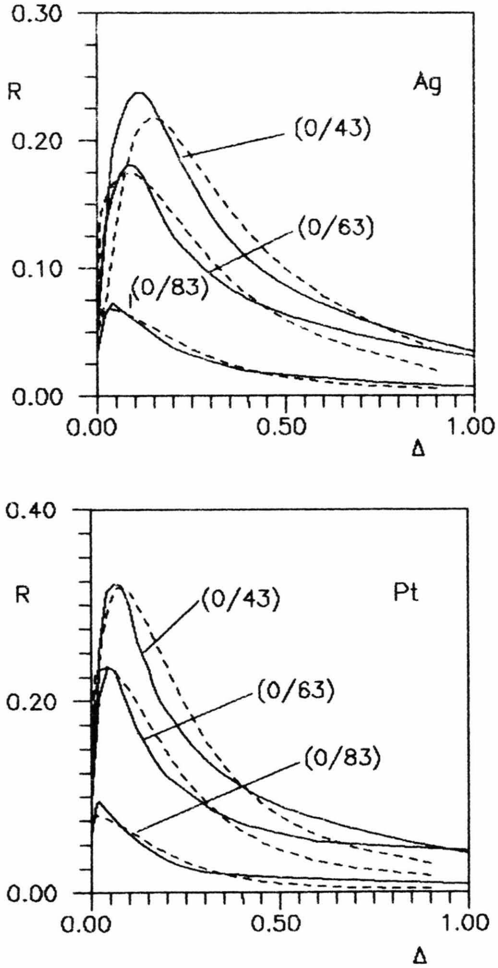


Fig. 3. Energy spectra of 30-keV electrons reflected from Ag and Pt targets for the scattering angles 43°, 63° and 83° (dashed lines, calculated according to formula (213); solid lines, experiment [16]).

3. DD SNRE in the Case of Electron Scattering by Massive Layer-Nonhomogeneous Targets

Let us use (8), (9) and (23) to calculate the DD SNRE in closed form in the case of electron scattering by massive multilayer samples. Figure 4 shows the dynamics of DD SNRE variation with increase of the thickness of an Au film on a half-infinite Al substrate.

For calculating the spectra of two-layer targets we used formula (8) twice. First it was used to find $R_{12}(x_2)$,

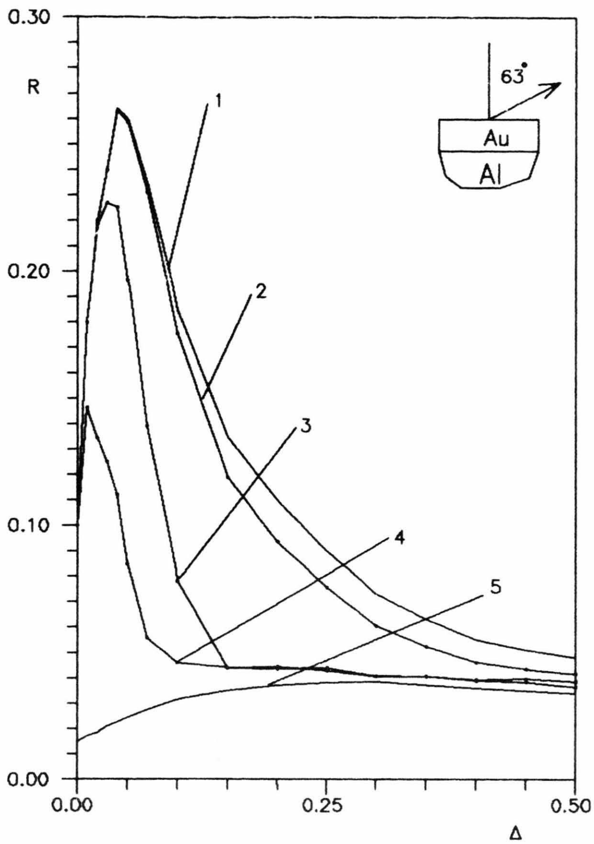


Fig. 4. Energy spectra of 25-keV electrons reflected from a two-layer system (bulk of Al, Au layer of thickness d). 1, $d=\infty$; 2, $d=2000$ Å; 3, $d=5000$ Å; 4, $d=9000$ Å; 5, $d=0$ Å.

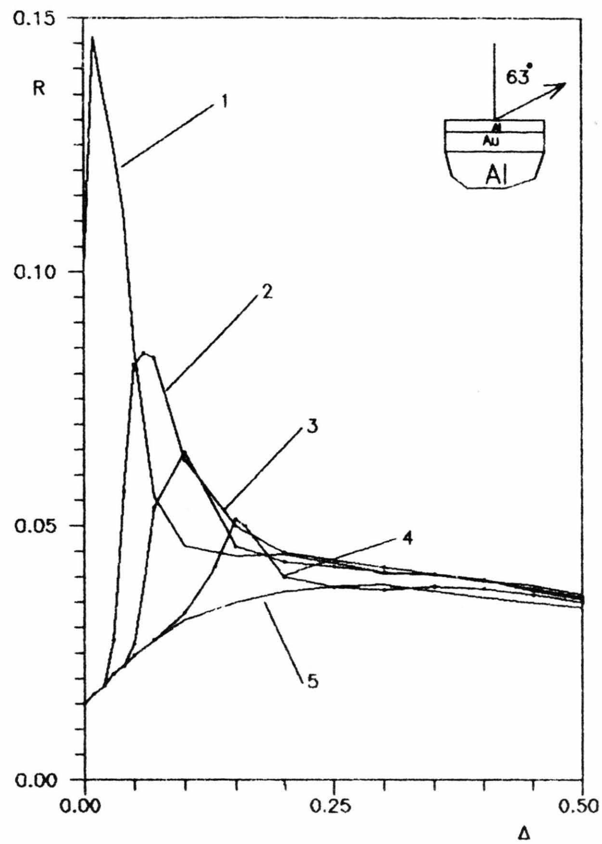


Fig. 5. Energy spectra of 25-keV electrons reflected from a three-layer system (bulk of Al, Au layer of thickness 200 Å, Al layer of thickness d). 1, $d=0$ Å; 2, $d=3000$ Å; 3, $d=5000$ Å; 4, $d=9000$ Å; 5, $d=\infty$.

the spectrum of a one-layer target. Then $R_{12}(x_2)$ was used in formula (8) instead of R_2, the spectrum of the substrate, to calculate $R_{123}(x_3+x_2)$. This calculation scheme has enabled us to calculate e.g. the evolution of DD SNRE which should be observed as a 200 Å thick Au marker moves into the target depth (Figure 5).

Unfortunately we were not able to find experimental data that would correspond to the situations presented, but the possibilities of the DD SNRE method in analyzing in situ the processes that occur with different changes in the layer-by-layer composition of a surface are clearly demonstrated by the calculations.

4. Conclusion

At present the nondestructive layer-by-layer analysis of layer-nonhomogeneous targets is made according to the method of Rutherford backward scattering (RBS). The realization of the latter method involves the creation of expensive megaelectron volt devices. The decoding of RBS spectra is based on the model of one deflection [17] and is relatively simple. The realization of the layer-by-layer analysis based on SNRE requires much less expansive and much more compact apparatus. The SIRE method enables one to make a nondestructive quick analysis in situ of layer-nonhomogeneous structures. It gives especially high resolution (up to fractions of an Ångström) when one analyses thin ($\cong 100$ Å thick) films. The procedure of decoding the SIRE signals is not so trivial as it is in the case of RBS. The spectrum decoding method presented in this paper is realized in the form of convenient high-service programs that work efficiently on the PC's used in modern analytical setups. This method can easily be realized experimentally with an analytical apparatus that includes an electron gun and an energy analyzer (for instance on AES setups).

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