Energy distribution of light ions backscattered from a solid

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The energy distribution of backscattered ions is obtained in analytical form as a result of approximate solution of the Boltzmann equation. The elastic-scattering cross section is taken in a form corresponding to the screened Coulomb potential which allows investigation of the cases from hard-sphere to Rutherford scattering. Inelastic energy losses are assumed to be proportional to some power of energy. The theory is valid for the case of normal ion incidence on the target surface and for the mass of a target atom much more than the mass of an ion. The first term of expansion of the final result gives the energy distribution obtained in the single-collision approximation and it coincides with the solution known from the previous theories. The theoretical results are verified by their comparison with computer simulation data. [S0163-1829(99)00533-0]

I. INTRODUCTION

If the surface of a solid is bombarded by electrons or by an ion beam then investigation of the angular and energy distributions of reflected particles gives the information about chemical and physical properties of the solid. This information is of essential importance for electron microscopy, nuclear fusion research, and radiation damage problems. Theoretical treatment of ion reflection represents the first and necessary step towards theoretical treatment of sputtering. For these reasons at present the process of reflection is intensively investigated by experimental[1] and computer simulation methods.[2] Analytical theories of reflection for the case of normal ion incidence on the target surface can be divided in two main categories of the theories based on the single-collision approximation and the theories which take into consideration multiple-collision effects.

The supposition of reflection by a single collision is generally used at high energies of incident ions where the reflection coefficient is relatively small. This model assumes two straight-line trajectories between the surface and the point of elastic collision. On their straight-line path the ions lose energy due to inelastic collisions, and they can also lose energy as a result of elastic collisions if the ratio of target atom to ion mass is finite. The single-collision theories started from the cases of the Rutherford elastic scattering and inelastic energy losses proportional to ion velocity.[3] Subsequent calculations considered more general cases of the screened elastic cross section[4] and inelastic losses proportional to some power of energy[5] that led to expressions valid in a wider range of energies. In Refs. 6 and 7 the theory was extended for heavier ions by taking into account elastic energy losses, and in Ref. 8 the effect of beam attenuation was added to the results.[6,7]

At lower ion energies the effects of multiple scattering become essential and the theory requires solution of the Boltzmann equation. In Ref. 9 the theory was based on the infinite-medium approximation with the target surface being considered as some reference plane which, unlike reality, may be crossed by the ion several times.

In recent years theoretical work has been devoted to the general case of multiple scattering in a half infinite medium. The theory in Ref. 10 assumes that the elastic-scattering cross section is isotropic in the laboratory system that represents an extremely rare case in reality. The theory in Ref. 11 is based on the approximate solution of the Boltzmann equation by the method of spherical harmonics. Within the limits of this method the delta function in the boundary condition is represented by a finite number of Legendre polynomials that leads to the problem of negative number of scattered ions and makes the final results questionable. Moreover, the theory in Ref. 11 disregards the correlation between scattering and energy loss in a single collision. For these reasons the theory in Ref. 11 achieved satisfactory results for the integral characteristic of backscattered ions—the reflection coefficient—but failed for the differential characteristic—the energy distribution.

In the previous works[12,13] the authors applied another method of approximate solution of the Boltzmann equation—the Chandrasekhar method of discrete streams[14]—in which the problem of negative number of scattered ions does not arise. In Refs. 12 and 13 the energy distribution of reflected ions was obtained in an analytical form for a power-law elastic-scattering cross section with inelastic energy losses being neglected. In the present work the inelastic energy losses are taken into consideration.

II. BASIC EQUATIONS

We consider the scattering and slowing down of ions in a half infinite target consisting of randomly distributed immovable atoms with the number density N. We assume that the ion beam of energy E_0 impinges perpendicularly on the target surface and that the ion mass is much less than the mass of the target atom. In this case the ions are scattered due to elastic collisions with atoms and they lose their energy as a result of electronic stopping between elastic collisions. We seek for the energy distribution, integrated over all exit angles, of those ions which underwent a number of elastic collisions and approached again to the target surface.

For the electronic stopping cross section we use the power-law expression

$$S_e(E)=K_pE^p, \tag{1}$$

where E is the ion energy, K_p is a constant, and the parameter p varies between $p=\frac{1}{2}$ for low energies and $p=-1$ for high energies.

For the elastic cross section we take the expression

$$\sigma(E,\omega)=\frac{2\eta(1+\eta)\sigma_0(E)}{(1+2\eta-\cos\omega)^2}, \tag{2}$$

where the scattering angle ω can be found from the equation

$$\cos\omega=\cos\theta\cos\theta_1+\sin\theta\sin\theta_1\cos(\varphi-\varphi_1), \tag{3}$$

and (θ_1,φ_1) and $(\theta,\ \varphi)$ are the normalized polar and azimuthal components of the ion velocity before and after collision.

Cross sections of the type (2) arise for the truncated Coulomb potential[15,16] and also as a result of quantum-mechanical solution of the scattering problem in the first Born approximation for the screened Coulomb potential.[17] As well as the Lindhard parameter m in the power-law cross section,[18] the screening parameter η in Eq. (2) depends on the ion energy. For low energies we have $\eta\gg1$, and Eq. (2) describes the hard-sphere interaction. For high energies $\eta\ll1$, and $\sigma(E,\omega)$ characterizes the Rutherford scattering. The advantage of the cross section (2) in comparison with the power-law cross section is the fact that its total cross section is finite,

$$\int_0^{\pi}\sigma(E,\omega)\sin\omega d\omega=\sigma_0(E), \tag{4}$$

and no problems arise in formulation of the Boltzmann equation.

In the case of normal ion incidence the problem is azimuthally symmetrical and integration of the cross section (2) over azimuthal angle gives the probability of ion scattering from the state $\mu_1=\cos\theta_1$ to the state $\mu=\cos\theta$:

$$dW(\mu_1,\mu)=\frac{d\sigma(\mu_1,\mu)}{\sigma_0}=\frac{2\eta(1+\eta)(1+2\eta-\mu\mu_1)d\mu_1}{[4\eta(1+\eta-\mu\mu_1)+(\mu-\mu_1)^2]^{3/2}}. \tag{5}$$

Let us denote by $f(z,E,\mu)dEd\mu$ the flux of ions at a depth z with energy (E,dE) and direction of motion $(\mu,d\mu)$. For the function $f(z,E,\mu)$ we can write the forward Boltzmann equation in a form similar to Refs. 11–13:

$$\mu\frac{\partial f}{\partial z}(z,E,\mu)=N\sigma_0\int_{-1}^{1}f(z,E,\mu_1)dW(\mu_1,\mu)-N\sigma_0f(z,E,\mu)+N\frac{\partial}{\partial E}[S_e(E)f(z,E,\mu)]. \tag{6}$$

The term in the left-hand side of Eq. (6) describes the space variation of the flux between elastic collisions. The first two terms in the right-hand side represent the collision integral for elastic scattering, and the third term allows for inelastic energy losses.

Equation (6) should be solved with the boundary condition

$$f(0,E,\mu)=\delta(E-E_0)\delta(1-\mu)\quad\text{for }\ \mu>0, \tag{7}$$

which means that the ion beam of energy E_0 impinges perpendicularly on a target surface $z=0$.

Our aim is to find the flux of those ions which are approaching to the target surface from inside, $f(0,E,\mu)$ for $\mu<0$, to integrate the flux over angle variable μ and to obtain the energy distribution of backscattered ions.

Before starting the solution of the system of Eqs. (6) and (7) it is convenient to define two parameters: the mean free path length of ions with respect to elastic collisions $\lambda_0=(N\sigma_0)^{-1}$, and the total inelastic ion range

$$R_T=\int_0^{E_0}\frac{dE}{NS_e(E)}=\frac{E_0^{1-p}}{(1-p)NK_p}. \tag{8}$$

Then the ratio

$$h=\lambda_0/R_T \tag{9}$$

will characterize the average number of elastic collisions of the ion in the target: the values $h\gg1$ describe the single-collision situation and the values $h\ll1$ correspond to the case of multiple scattering.

If we also introduce the dimensionless depth variable $x=z/\lambda_0$, the relative ion energy $u=E/E_0$, and the new energy variable

$$t=\frac{1-u^{1-p}}{h}, \tag{10}$$

then for the new unknown function $g(t)$ defined by the transformation $f(u)du=g(t)dt$ instead of Eq. (6) and boundary condition (7) we can write

$$\mu\frac{\partial g}{\partial x}+g+\frac{\partial g}{\partial t}=\int_{-1}^{1}g(\mu_1)dW(\mu_1,\mu), \tag{11}$$

$$g(0,t,\mu)=\delta(t)\delta(1-\mu)\quad\text{for }\ \mu>0. \tag{12}$$

The variable t is proportional to the time of presence of an ion within the target and it is similar to the so-called Fermi age used in the theory of the slowing down of neutrons.[19] This variable gives the possibility to formulate Eq. (11) in a rather general form valid for arbitrary scattering probability $dW(\mu_1,\mu)$ and for arbitrary values of the parameter p in the electronic stopping law (1).

Equation (11) in some different notations was already formulated in the theories[11,20] with following expansion of the solution into series of the Legendre polynomials. Analytical theory[11] used the first two polynomials that may be enough for the infinite-medium problems,[19] but is not sufficient for the half space target. The theory in Ref. 20 considers a large number of polynomials, but it is based upon numerical solution of a system of ordinary differential equations and its results contain only integral and not differential characteristics. The main divergence of the present calculations from the theories[11,20] is another way of approximate solution of Eq. (11).

III. APPROXIMATE SOLUTION

The Laplace transformation

$$G(x,s,\mu)=\int_0^\infty e^{-st}g(x,t,\mu)dt \tag{13}$$

reduces Eqs. (11) and (12) to

$$\mu\frac{\partial G}{\partial x}+(1+s)G=\int_{-1}^{1}G(\mu_1)dW(\mu_1,\mu), \tag{14}$$

$$G(0,s,\mu)=\delta(1-\mu)\quad \text{for}\ \ \mu>0. \tag{15}$$

If from Eqs. (14) and (15) we find the function

$$R(s,\eta)=-\int_{-1}^{0}\mu G(0,s,\mu)d\mu, \tag{16}$$

then inverse transformation of $R(s,\eta)$ together with Eq. (10) will give the energy distribution of backscattered ions.

For two particular cases Eq. (14) can be solved analytically.

The case of $s\gg 1$ describes the single-collision situation when the solution can be found by the methods used in Refs. 3–8:

$$R(s\gg 1,\eta)=\frac{a(\eta)}{2s}, \tag{17}$$

where the function

$$a(\eta)=-\int_{-1}^{0}\frac{2\mu_1 dW(\mu_1,1)}{1-\mu_1}=1-\frac{1+\eta}{\eta}\ln\frac{1+2\eta}{1+\eta} \tag{18}$$

is proportional to the the reflection coefficient R_1 of the ions which suffered a single collision,[8]

$$R_1=\frac{a(\eta)}{2}\left[1-\exp\left(-\frac{1}{h}\right)\right]. \tag{19}$$

The case of $\eta\gg 1$ corresponds to the hard-sphere interaction with the isotropic scattering probability $dW(\mu_1,\mu)=\frac{1}{2}$ for which solution of the system of Eqs. (14) and (15) has the form

$$R(s,\eta\gg 1)=1-\sqrt{1-\alpha}\cdot H(\alpha,1), \tag{20}$$

where $\alpha=(1+s)^{-1}$ and

$$H(\alpha,\mu)=\exp\left[-\frac{\mu}{\pi}\int_0^\infty \ln\left(1-\alpha\frac{\arctan y}{y}\right)\frac{dy}{1+\mu^2y^2}\right] \tag{21}$$

is the Chandrasekhar H function.[14] The function (20) shows the following asymptotical behavior:

$$R(s,\eta\gg 1)=\begin{cases}1-2.9078\sqrt{s} & \text{for}\ \ s\ll 1,\\ \dfrac{1-\ln 2}{2s} & \text{for}\ \ s\gg 1.\end{cases} \tag{22}$$

If we also take into account that $R(s=0,\eta)=1$ for arbitrary scattering probabilities $dW(\mu_1,\mu)$ and for arbitrary η, we can write an approximate solution of the problem (14) and (15) in the form

$$R(s,\eta)=\frac{a}{s+a+[(s+a)^2-a^2]^{1/2}}. \tag{23}$$

The function (23) has the following features: (a) at $s\ll 1$ it behaves in the same way as the solution (22), (b) at $s\gg 1$ it coincides with the solution (17), and (c) at $\eta\gg 1$ it represents approximately the solution (20) with the maximum divergence 6% at $s=0.16$.

The function (23) is the exact Laplace transform to

$$\frac{dR}{dt}=\frac{1}{t}e^{-at}\cdot I_1(at) \tag{24}$$

(Ref. 21), where $I_1(x)$ is the modified Bessel function of the first kind.[22] If now we return to the variable u in the accordance with Eq. (10), we obtain the energy distribution of backscattered ions integrated over all exit angles:

$$\frac{dR}{du}=\frac{(1-p)\exp[-a(1-u^{1-p})/h]}{u^p(1-u^{1-p})}\cdot I_1\left(\frac{a}{h}(1-u^{1-p})\right). \tag{25}$$

Equation (25) represents the central result of the work. It shows that the shape of energy distribution depends upon two parameters: the power p in the electronic stopping law (1), and the ratio $b=a/h$, where the function $a(\eta)$ corresponds to the reflection coefficient by a single collision and the parameter h describes the number of collision events.

Behavior of the energy distribution (25) at low energies and at the energies close to E_0 can be obtained by using asymptotic forms of the function $I_1(x)$ at $x\gg 1$ and $x\ll 1$, correspondingly. In the case of $x\gg 1$ we have the energy distribution of ions which participated in a large number of collisions:

$$\frac{dR}{du}=\left(\frac{b}{2\pi}\right)^{1/2}\cdot\frac{1-p}{u^p(1-u^{1-p})^{3/2}}\quad \text{for}\ \ b(1-u^{1-p})\gg 1. \tag{26}$$

In the case of $x\ll 1$ we obtain the energy distribution of backscattered ions which participated in a single collision:

$$\frac{dR}{du}=\frac{(1-p)b}{2u^p}\quad \text{for}\ \ b(1-u^{1-p})\ll 1. \tag{27}$$

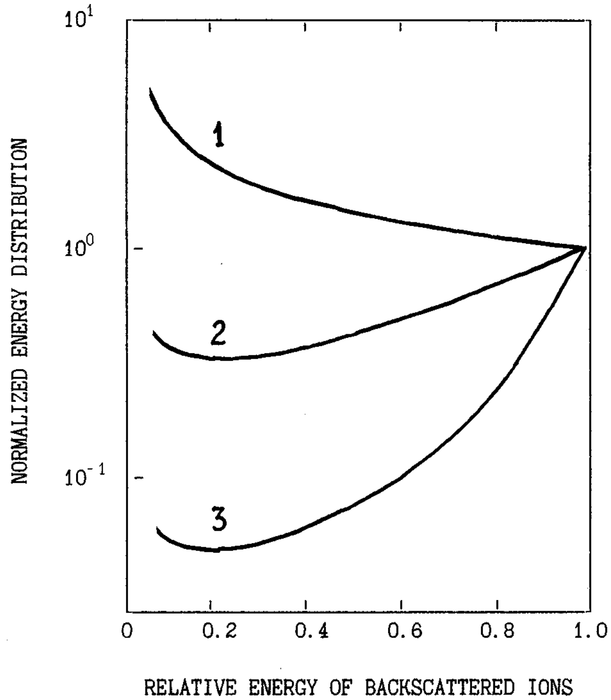

FIG. 1. Energy distribution of backscattered ions (25) as a function of their relative energy $u=E/E_0$ for $b \ll 1$ (curve 1), $b=5$ (2), and $b=20$ (3). The distribution is normalized to its value at $u=1$.

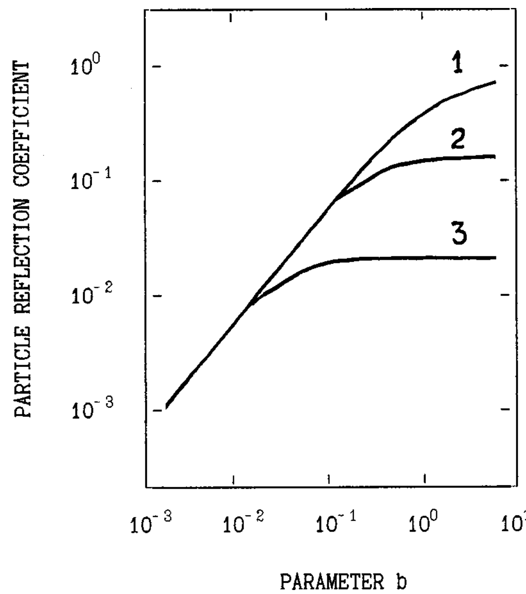

FIG. 2. The particle reflection coefficient R_N (curve 1) and the single-collision reflection coefficient R_1 for $\eta \gg 1$ (2) and $\eta=0.1$ (3).

The result (27) coincides with the solution obtained in the single-collision approximation and it also can be deduced by inverse Laplace transformation of the expression (17).

IV. THE ENERGY DISTRIBUTION

Figure 1 presents the energy distribution (25) for different values of the parameter b. Here and everywhere in this section we assume that inelastic energy losses are proportional to the ion velocity and consider Eq. (25) for the particular case of $p=\frac{1}{2}$. At $b \ll 1$ the distribution is a monotonically decreasing function of energy and it is similar to the energy distribution of the ions after the single collision in the case of $h \gg 1$ when the probability of a scattering event is small. When the parameter b increases, the low-energy part of the distribution falls, and for $b>1$ there appears a maximum at $u=1$. In the limiting case of $b \gg 1$, when inelastic energy losses may be disregarded, the distribution takes the delta-type shape and contains only ions with relative energies $u \approx 1$.

We should like to emphasize that both energy distribution (25) and all analytical energy distributions obtained in the single-collision approximation[3–8] are finite at the energy $E=E_0$ $(u=1)$. The infinity of the energy distribution calculated in Ref. 11 at $E=E_0$ is only the consequence of the approximate method of solution but not of the physical background of the problem.

Figure 2 shows the dependence of the particle reflection coefficient R_N and the single-collision reflection coefficient R_1 on the parameter b. The value of R_N was obtained by numerical integration of the function (25) over energy variable u in the limits (0, 1) and the value R_1 was calculated from Eq. (19). From Fig. 2 we may find the region of validity of the single-collision approximation in terms of the reflection coefficient R_N: at low ion energies and hard-sphere interaction $(\eta \gg 1)$ the single-collision model can be applied for $R_N<0.1$, and at higher energies and screened Coulomb interaction $(\eta=0.1)$ for $R_N<0.01$.

Measurements of the energy distributions at normal ion incidence mainly contain the data for fixed exit angles θ. Inasmuch as we could not find in the literature experimental distributions integrated over all θ, we used the data of computer simulation.[23]

Figure 3 gives the energy distribution of backscattered ions at primary energy $E_0=300$ eV for the combination D-W (the target atom to ion mass ratio $M_2/M_1=92$). The theoretical curve is obtained from Eq. (25) for the value $b=5$ which corresponds to the reflection coefficient $R_N=0.65$ in Fig. 2, the former being deduced by interpolation

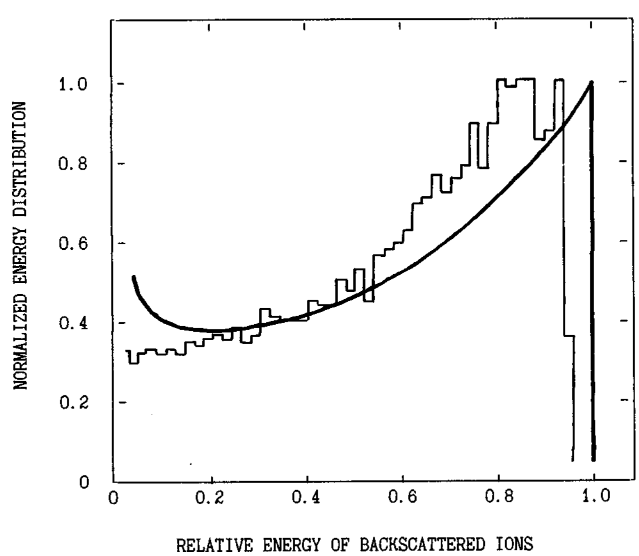

FIG. 3. Combination D-W at $E_0=300$ eV. Energy distribution of backscattered ions normalized to the maximum value. Solid line: Eq. (25), histogram: computer simulation (Ref. 23).

of experimental[24] and computer data.[2] Agreement of the theoretical and computer distributions is good with the exception of energies close to the bombarding energy. Equation (25) cannot explain the shift of the maximum of the distribution from $E/E_0=1$ to $E/E_0\approx0.9$. This shift can be the consequence of either elastic energy losses due to the finite mass ratio M_2/M_1 or the fluctuations of inelastic energy losses, but it is difficult to predict in advance which of these two effects dominates.

V. RESULTS AND DISCUSSION

Development of the theory of ion backscattering from a solid represents an extremely complicated problem of taking into consideration and mathematical interpretation a number of separate effects: (i) the effect of half infinite target, (ii) the effect of delta-type boundary condition, (iii) elastic scattering of the ions by the target atoms, (iv) the inelastic energy losses, and (v) the elastic energy losses. In the case of moderate and low ion energies, when the multiple scattering is dominating, all the existing theories are based on the solution of the Boltzmann equation by one or another approximate method.

At present the most complete theory of ion backscattering is the theory of Vicanek and Urbassek[11] which allows for all the effects mentioned above. However, the theory[11] contains two serious defects. First, the elastic energy losses are considered within the limits of the diffusional approximation. That neglects the correlation between the energy loss and the scattering angle in a collision event and distorts the energy distribution at least at energies close to the primary energy. And second, in Ref. 11 the angular dependence of the ion flux is represented in a form of two first Legendre polynomials. As it was pointed out in Ref. 25, that leads to the problem of negative ion density. Indeed, a closer investigation of intermediate calculations in Ref. 11 shows that at definite target depths the number of ions with definite energies becomes negative.

In the theories of Refs. 12 and 13 the problem of negative ion density was avoided by applying another method of approximate solution of the Boltzmann equation—the method of discrete streams. Unfortunately, the authors did not succeed in simultaneous interpretation both elastic and inelastic energy losses, and in Refs. 12 and 13 electronic stopping was disregarded.

In the present work, on the contrary, the inelastic energy losses are included in the theory, but the elastic losses are neglected, which holds for the cases of light ions (H, D, and He) and heavy targets. The approximate solution is constructed as an extrapolation of two analytical solutions known in the literature—the solution for isotropic scattering and the solution for a single collision event. The final energy distribution (25) is valid for arbitrary values of the parameter p in the electronic stopping law (1) that makes the result valid for a rather wide range of ion energies. Agreement between theoretical and computer simulation distributions is good with the exception of ion energies near the bombarding energy. One can expect that the divergence will be removed by taking into consideration the effects of elastic energy losses and fluctuations in the electronic stopping.

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