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Dependence of the Sputtering Yield on the Angle of Ion Incidence on the Target Surface

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We investigated the behavior of sputtering yield for oblique angles of ion incidence on the target surface. The range of incidence angles varied from 0° (normal incidence) to almost 90° (limiting grazing incidence). Computer simulation was performed by the program PAOLA. Theoretical analysis included numerical solution of the Chandrasekhar integral equation. The results obtained are compared with the results of other authors

Keywords: sputtering under ion bombardment, oblique ion incidence, sputtering yield, computer simulation, theoretical analysis.

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Introduction

Sputtering of hards under ion bombardment plays an important role in design of fusion reactors, thin film production, development of surface analysis methods, and ion implantation. Recently a significant array of experimental data has been accumulated for various sputtering parameters [1,2]. One important characteristic is sputtering yield, i.e., the ratio of sputtered atoms to number of ions incident on a target. Dependence of sputtering yield on incidence angle for heavy ions is non-monotone. First, sputtering yield Y grows from its value at normal incidence $Y(0°)$ to the maximum value, and then decreases to the value $Y(90°)$ at sliding incidence. Calculations using code SRIM-2013 [3] provide finite values $Y(90°)$, but in paper [4] the calculations using OKSANA code provided zero values of $Y(90°)$ when modelling silicon sputtering by ions of noble gases.

Measurement of sputtering yield under sliding incidence of ions on a target surface is a complex experimental problem. In the literature [2] there are known measurement results for incidence angles only that do not exceed 85°. Therefore, additional research of the phenomenon is necessary and very desirable. In this paper the problem of sputtering yield dependence on ion incidence angle is considered by methods of computer modelling and theoretically. We generalize our previous results for normal incidence [5] for the case of oblique ion incidence.

1. Computer simulation

Computer simulation was performed using PAOLA code based on the binary collision approximation and the screened Coulomb potential [5]. PAOLA contains considerably fewer adjustable parameters than SRIM-2013 and is capable of simulating various limit modes, such as the case of particle interaction under the law of hard spheres.

Before each elastic collision, three random numbers R_1, R_2, R_3 are generated in the range from 0 to 1. These numbers define particle path between collisions $\lambda = \lambda_0 \ln(1/R_1)$, polar angle of scattering ω in the system of center of mass

$$\cos\omega = \frac{2(1+\varepsilon)R_2 - 1}{1 + 2R_2\varepsilon} \qquad (1)$$

and azimuthal angle of scattering $\varphi = 2\pi R_3$, where λ_0 means average length of free path, and ε — reduced energy. Angles of scattering of oncoming particle Ω_1 and knocked-on atom Ω_2 in the laboratory system of coordinates are produced from equations

$$\cos\Omega_1 = \frac{A + \cos\omega}{\sqrt{1 + 2A\cos\omega + A^2}}, \quad A = \frac{M_1}{M_2}, \qquad (2)$$

$$\cos\Omega_2 = \sqrt{\frac{1-\cos\omega}{2}}, \qquad (3)$$

where M_1 and M_2 — masses of ion and atom of the target, respectively. If oncoming particle energy before collision is E, then after collision this particle and the knocked-on atom will have energies of

$$E_1 = E(1 - \gamma\cos^2\Omega_2), \qquad (4)$$

$$E_2 = \gamma E\cos^2\Omega_2, \qquad (5)$$

where $\gamma = 4A/(1+A)^2$.

It should be noted that equations (1)–(5) are universal and are valid for both light ions ($A \le 1$) and heavy ions ($A \ge 1$).

Statistics of cascade atoms is accounted for as follows. Each knocked-on atom is characterized by two integers: number of a generation in a cascade and an order number within that generation. Cascade registration involves sequential consideration of primary knocked-on atoms, secondary knocked-on atoms, and further generations of the atomic cascade. The cascade stops when the particle leaves the target, or its energy is reduced to cutoff energy $E_{\min}$ insufficient to knock the target atom out of its equilibrium position.

2. Theoretical analysis

Theoretical review of cascading particle multiplication within sputtering phenomenon is usually conducted under some form of simplifying assumptions. In theory [6] isotropic angular distribution approximation of sputtered atoms is used. In theories [7–9] angular distribution is represented as a sum of several spherical functions. However, delta-type surface boundary condition describing ion entry into a target may not be accurately enough described by two or three spherical functions. In theory [10], the method of discrete streams has solved the boundary condition problem, but three discrete streams are sufficient only to consider normal ion incidence. In case of inclined incidence, the number of discrete streams must be increased, and the theoretical solution is transformed from analytical to numerical.

In the general case, the sputtering theory considers a system of two transport equations: one equation for ions, the other — for cascade atoms. To illustrate the method, we will consider a case of self-sputtering, where ions and atoms of the target have equal masses and are indistinguishable from each other. The distribution function of self-sputtered atoms remove the word $f(x, \mu, u)$ depends on normalized depth of the target x, cosine of angle θ between the velocity of atom and internal normal to the surface, $\mu = \cos\theta$, and relative energy $u = E/E_0$, where E_0 — energy of bombarding ions.

Mellin transform by energy variable

$$F(x, \mu, s) = \int_0^1 u^{s-1} f(x, \mu, u) du \tag{6}$$

reduces transport equation to one-velocity equation of transfer

$$\mu \frac{\partial F(x, \mu, s)}{\partial x} + F(x, \mu, s) = \int_{-1}^{1} \sigma(\mu, \mu') F(x, \mu', s) d\mu'. \tag{7}$$

Delta-type boundary condition for equation (7) takes the form $F(0, \mu) = \delta(\mu - \mu_0)$ and means that ion incidence angle is equal to θ_0, $\mu_0 = \cos\theta_0$. Function $\sigma(\mu, \mu')$ means weighted cross section of scattering averaged over represents angle

$$\sigma(\mu, \mu') = \frac{2}{\pi} \int_0^{\pi} \sigma(\cos\Omega)(\cos^{2s}\Omega + \sin^{2s}\Omega) d\varphi, \tag{8}$$

where Ω — angle of scattering in the laboratory system of coordinates,

$$\cos\Omega = \mu\mu' + \sqrt{1-\mu^2}\sqrt{1-\mu'^2}\cos\varphi. \tag{9}$$

The collision integrals (8) summed in brackets mean that an elastic collision produces two atoms moving in two mutually perpendicular directions. If we substitute $s = 1$, the equation in brackets turns to one in remove the word agreement with the energy conservation law.

The atomic potential choice is an important problem for any theoretical study. The ZBL [1] potential used in SRIM-2013 code contains too many parameters and cannot be used for analytical investigation. Paper [11] proposed potential based on calculation of atomic state density (potential DFT), but authors specify no analytical formulae. In this paper we use screened Coulomb potential, for which differential cross section of scattering in the laboratory system of coordinates may be recorded in analytical form [12]

$$\sigma(\cos\Omega) = \begin{cases} \frac{2\eta(1+\eta)\cos\Omega}{(1+\eta-\cos^2\Omega)^2} & for\ \ \cos \geq 0 \\ 0 & for\ \ \cos\Omega \leq 0 \end{cases}, \tag{10}$$

through $\eta = [4\varepsilon(1+\varepsilon)]^{-1}$ specifies the screening parameter. At low energies ($\eta \gg 1$) the scattering cross section (10) complies with scattering on a potential of hard spheres, at high energies ($\eta \ll 1$) — Rutherford scattering on a Coulomb potential.

One of the possible methods to solve the equation of transfer (7) is to divide the integration interval into N equal parts and to calculate values of unknown distribution function in $N + 1$ discrete point. To find these values, the solution is decomposed into series of functions exponentially reducing in depth, and eigenvalues and eigenvectors of square matrix of size $N \times N$ are defined to describe angular distribution. Decomposition constants are determined from boundary condition after solving the system of N algebraic equations. The disadvantage to the method consists in the fact that the solution contains additional information that we do not need: we receive the values of the distribution function in all depths of the target, while we are interested only in the values of the function on the surface.

The alternative method consists in a solution for a nonlinear integral equation obtained by Chandrasekar after applying the principle of invariance to equation (7) [13,14]. This method contains no distribution of atoms in depth of the target and reviews only their angular distribution on the surface. In Chandrasekar method the reflection function $R(\mu_0, \mu_1, s)$ depends only on two variables — ion incidence angle θ_0 and angle of sputtered atom exit from the target θ_1,

$\mu_1 = \cos\theta_1$. Integration over all exit angles gives sputtering yield as function of parameter s:

$$Y(\mu_0, s) = \int_0^1 \mu_1 R(\mu_0, \mu_1, s) d\mu_1, \qquad (11)$$

and inverse transformation results in energy distribution of sputtered atoms. Integration of energy distribution provides sputtering yield.

To apply Chandrasekar method, let us divide differential cross section of scattering (8) into two parts

$$p(\mu, \mu') = \sigma(\mu, \mu') \quad \text{for} \quad \mu\mu' < 0,$$

$$q(\mu, \mu') = \sigma(\mu, \mu') \quad \text{for} \quad \mu\mu' > 0. \qquad (12)$$

Scattering cross section $p(\mu, \mu')$ relates to atoms, speed of which as a result of collision changed its direction from inside the target to outside, and vice versa. Scattering cross section $q(\mu, \mu')$ relates to atoms, whose speed after collision preserved its direction either inside the target or outside. We divided integration interval [0,1] into N equal parts and substituted the integral with finite sums. If we specify the reflection function and both cross sections of scattering by two indices only R_{01}, p_{01}, q_{01}, then integral equation of Chandrasekar may be written in tensor form

$$(\mu_0 + \mu_1)R_{01} = p_{01} + \mu_0 R_{02} q_{21} + q_{02} R_{21} \mu_1 + \mu_0 \mu_1 R_{02} p_{23} R_{31}, \qquad (13)$$

where it is suggested to sum up using matching indices. The integral equation (13) was solved by the method of successive iterations.

Inverse Mellin transformation may be performed by two methods. The first method consists in reviewing only real values of parameter s in equation (6) and adjustment of the produced numerical values for one of the known table functions [15]. Accuracy of such method is unpredictable and requires additional justification. The other method consists in review of complex values of parameter $s = 1 + i\omega$ and integration on a complex plane. Besides, this complicates solution to integral equation (13), which shall be recorded both for real and imaginary parts of the reflection function. But as a result the inverse Mellin transformation results in calculation of the only integral, and the accuracy of the solution depends only on the number of divisions N in integration interval. Convergence of the solution was monitored by increasing number N, the maximum value of which made $N = 500$. Validity of the method was checked using some test problems, which had analytical solution, and also using PAOLA code.

3. Results and discussion

Fig. 1 and 2 show the simulation results using PAOLA program presented in the form of smoothened curves. Theoretical results produced by solving an integral equation differ from the simulation results by not more than 2%.

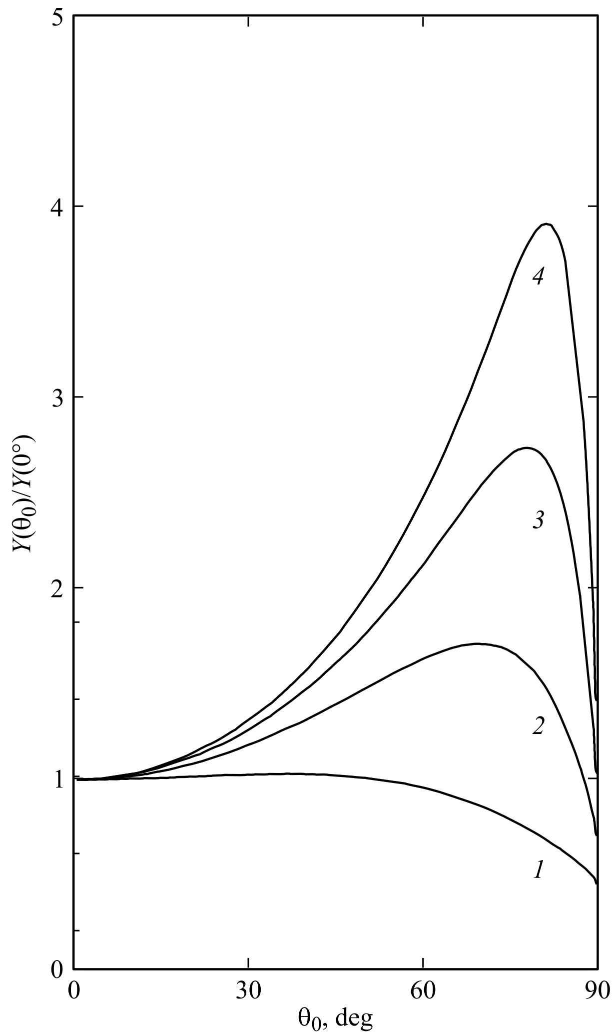

Figure 1. Dependence of sputtering yield Y on ion incidence angle θ_0 for various mass ratios $M_1/M_2 = 1$ (curve *1*), 5 (curve *2*), 10 (curve *3*), 15 (curve *4*) at $E_0/E_{\min} = 1000$.

Fig. 1 shows the dependence of the sputtering yield on the ion incidence angle with fixed energy of ions and various ratios of masses of target ion and atom $A = M_1/M_2$. All coefficients are normalized to values at normal incidence. If $A = 1$, sputtering yield monotonously decreases with increase of ion incidence angle. If ion mass ($A > 1$) increases, a maximum appears in the angular curve in the region of incidence angles from 70° to 80°, the maximum height increases with growth of ratio A. In the limit case of incidence angles close to 90°, all distributions take finite, and not zero values.

Fig. 2 shows the dependence of sputtering yield on the ion incidence angle with fixed mass and various energies of ions. We can see that height of distribution maximum decreases as energy increases. In case of limit grazing incidence angles the sputtering yield still takes finite values.

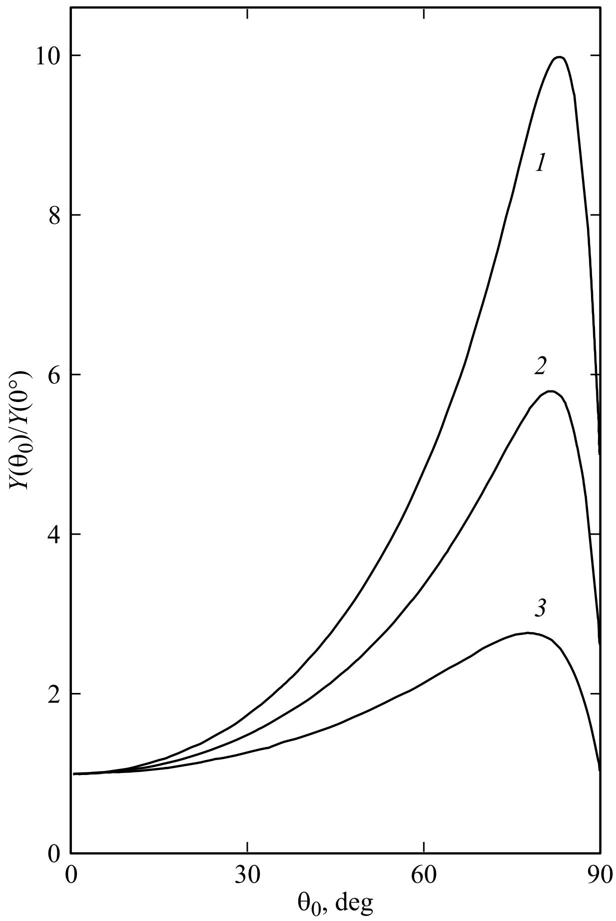

Figure 2. Dependence of sputtering yield Y on ion incidence angle θ_0 for different ion energies $E_0/E_{\min} = 100$ (curve *1*), 200 (curve *2*), 1000 (curve *3*) at $M_1/M_2 = 10$.

The produced results are agreeable with the results of calculations from SRIM program, but are opposite to calculations using OKSANA program. In paper [4] it is specified that difference in results is related to different interpretation of the first ion collision after entry into the target. In SRIM-2013 code the first collision occurs at the depth equal to the length of free path λ_0. In OKSANA code the first collision occurs in the depth equal to the radius of atomic potential exposure d. In case of a silicon target, we have $d < \lambda_0$, which results in reduction of sputtering yield. In PAOLA code the first collision occurs at depth $\lambda = \lambda_0 \ln(1/R_1) \cos\theta_0$, which may be less or more than depth λ_0. This prevents from conclusions in favor of this or that approach, but quite definitely indicates finite values of sputtering yield with grazing ion incidence angles.

4. Conclusion

With ion incidence angles close to 90°, sputtering yield takes finite values for any ion-target combinations and any ion energies. This result is produced by two independent methods — by simulation in PAOLA program and using numerical solution of the Chandrasekar integral equation. Conclusion on zero value of sputtering yield for grazing ion incidence may be related to inaccurate interpretation of the first collision of ion entering the target.

Conflict of interest

The authors declare that they have no conflict of interest.

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