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FAST TRACK COMMUNICATION

Angular dependence of sputtering yield of amorphous and polycrystalline materials

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Abstract

An analytical formula is developed for the evolution of angular dependence of sputtering yields by extending the theory of sputtering yield proposed by Sigmund. We demonstrate that the peak of sputtering yield at oblique incidence can be attributed to a balance between the increased energy deposited on the surface by incident ion which enhances the sputtering yield and the decreased depth travelled by recoil atom which reduces the sputtering yield. The predicted dependence of sputtering yield on the incident angle is in good agreement with experimental observations.

(Some figures in this article are in colour only in the electronic version)

Ion-induced sputtering is a subject of constant research by many scientists over the last few decades due to its wide application in the semiconductor industry, in surface analysis and deposition. The understanding of this phenomenon lies in the framework of Sigmund's theory [1]. This theory was derived on the basis of the linear Boltzmann transport equation under the assumption of random slowing down in an infinite medium. For amorphous and polycrystalline targets, Sigmund revealed that the sputtering yield is proportional to the energy accumulated by ions on the surface. It was shown that this theory can be used successfully to predict energy-dependent sputtering yields for a wide range of energies and a variety of ion–target combinations [2–6]. Many surface features induced by ion bombardment, including ripple and nanodot formation are based on this theory [7–15]. However, one challenging problem associated with this process is the angle-dependent sputtering yield. According to Sigmund's theory, the evolution of sputtering yield with ion energy E and incident angle θ measured from the surface normal is given by

$$Y(E, \eta) = \Lambda F(E, \eta), \tag{1}$$

where $\eta = \cos\theta$, $\Lambda = 0.042/(NU_0)$, N is atomic density, U_0 is surface binding energy, $F(E, \eta)$ is energy distribution. This equation can be understood as the production of sputtered atom density (in unit of atoms per length) per bombarding ion and depth from which sputtered atoms come [1, 16]. By solving the linear Boltzmann's equation under the assumption of an infinite medium using Thomas–Fermi cross section $d\sigma = C_m E^{-m} T^{-1-m}\, dT$ with $m = 0$ and $C_0 = \frac{1}{2}\pi\lambda_0 a^2$, where $\lambda_0 = 24$ and $a = 0.219$, Sigmund obtained incidence dependent sputtered atom density $F(E, \eta)/(\pi^2 U_0)$ and incidence independent depth $3/(4NC_0)$ [1, 16]. The production of these two terms determines the sputtering yield (equation (1)). Assuming a Gaussian distribution of deposited energy distribution $F(E, \eta)$, from equation (1) the normalized sputtering yield can be approximated as

$$\frac{Y(E, \eta)}{Y(E, \eta = 1)} = (\cos\theta)^{-f_s}, \tag{2}$$

where the exponent $f_s \approx 1 \sim 2$, depending on the mass of ion and atom [1,5]. This means that sputtering yield increases with the incidence angle and goes to infinity for grazing incidence.

It is well known from experiment that the sputtering yield reaches a maximum at an oblique incidence of about 70° and then approaches zero at $\theta = 90°$. Sigmund pointed out that this maximum sputtering yield at a certain glancing angle cannot be explained on the basis of the assumption of an infinite medium [1]. Although this subject is mostly of applied interest and has been intensively investigated over several decades [2, 17–20], angular dependence of sputtering yield is still not well understood.

In this letter, starting with the recoil atom density [1, 16, 21], we show that the sputtered atom depth is proportional to the cosine of incident angle. The peak of sputtering yield can be attributed to a balance between two competitive effects: one is the deposited energy $F(E, \eta)$, which increases with the incident angle and thus enhances the sputtering yield, and another is the sputtered atom depth, which decreases with the incident angle and thus reduces the sputtering yield.

According to Sigmund's theory [16, 21], the average number of recoil atoms passing through the surface plane with energy $(E_1, \mathrm{d}E_1)$ in the solid angle $(\mathbf{\Omega_1}, \mathrm{d}\Omega_1)$ per incident ion is given by [21]

$$Y = \iint J(E_1, \mathbf{\Omega}_1)\, \mathrm{d}E_1\, \mathrm{d}^2\Omega_1, \tag{3}$$

where $J(E_1, \mathbf{\Omega_1})$ is the number of recoil atoms per unit energy and unit solid angle. Equation (3) gives the sputtering yield if we integrate over $E_1 \cos^2\theta_1 > U$, where θ_1 is the angle between $\mathbf{\Omega_1}$ and the outward surface normal, $U/\cos^2\theta_1$ is the surface binding energy. Following the approach suggested by Falcone and Sigmund [21], using power cross section with $m = 0$, $J(E_1, \mathbf{\Omega_1})$ is given by

$$J(E_1, \mathbf{\Omega}_1) = \frac{3F(E,\eta)}{2\pi^3} \times \int_0^\infty \frac{\mathrm{d}E_0}{E_0^2} \int_0^\infty \mathrm{d}x\, \delta(E_1 - f(E_0, x, \mathbf{\Omega_1})), \tag{4}$$

where $F(E, \eta)$ is the deposited energy density on the surface, E_0 is the initial energy of recoil energy, δ is the Dirac delta function, $f(E_0, x, \mathbf{\Omega_1})$ is the energy of the recoil atom with initial energy E_0 after travelling from x to the surface in the direction $\mathbf{\Omega_1}$. In order to integrate equation (4), we need to know the relationship between energy E_1 and initial energy E_0 at depth x for recoil atoms.

We assume in general that energy loss for both ion and recoil atom has the form [21]

$$\frac{\mathrm{d}E}{\mathrm{d}R} = -CE^\gamma, \tag{5}$$

where R is the travelled path length and C and γ are constants. For power approximation of cross section, $\gamma = 1$–$2\,$m. If we assume $m = 0$, according to Sigmund's assumption [1], then $C = NC_0$, where N is target atomic density.

For an incident ion with initial energy E_i and incidence θ, the energy E_0 at depth x from the surface is given by (integrating equation (5) under $m = 0$ for incident ion)

$$E_0 = E_i \exp\left(-\frac{Cx}{\eta}\right). \tag{6}$$

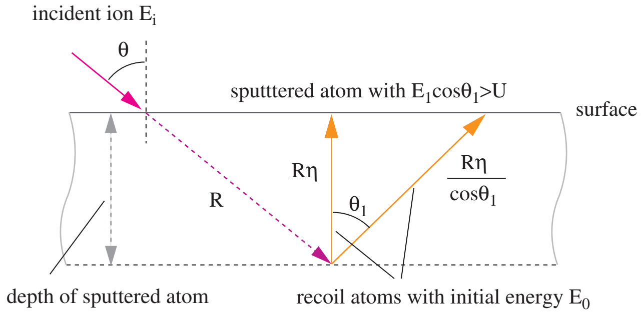

Figure 1. Schematic illustration of the variation of sputtered atom depth with incident angle. Recoil atom with initial energy E_0, after travelling depth $R\eta/\cos\theta_1$, reached the surface with energy E_1. Sputtered atoms satisfy $E_1\cos\theta_1 > U$ (surface binding is given by $U/\cos^2\theta_1$).

This equation shows that the depth of an incident ion that has energy E_0 under off-normal bombardment is equal to the cosine of the incident angle times the depth of the incident ion with the same energy under normal bombardment (figure 1). Because this energy will be transferred to the recoil atom, for a given energy E_0, the depth of the recoil atom has the same relationship between normal and off-normal bombardment. Thus the energy E_1 of a recoil atom with initial energy E_0 at depth x from the surface is given by (integrating equation (5) under $m = 0$ for the recoil atom)

$$E_1 = E_0 \exp\left(-\frac{Cx}{\eta\cos\theta_0}\right), \tag{7}$$

where θ_0 is the angle between $\mathbf{\Omega_0}$ and the outward surface normal. This equation is different from that derived by Falcone and Sigmund [21] by a parameter of η on the right-hand side of equation (7). This reduced depth at off-normal bombardment shows that more recoil atoms can easily escape from the surface without inducing further recoil atoms, and then lead to the decrease in sputtering yield. The schematic explanation of this difference is shown in figure 1. Substituting equations (7) and (4) into equation (3) yields

$$Y(E, \eta) = \eta\Lambda F(E, \eta), \tag{8}$$

where power approximation of the Thomas–Fermi cross section with $m = 0$ is used. At normal bombardment, this equation reduces to Sigmund's result. At off-normal bombardment with increasing incident angle, η decreases and $F(E, \eta)$ increases. When the incident angle is equal to 90°, because the depth of sputtered atoms is zero, the sputtering yield reaches zero. Deposited energy distribution $F(E, \eta)$ on the surface can be approximated as a Gaussian distribution set up in terms of the moments [1]

$$F(E, \eta) = \frac{E_i}{(2\pi)^{1/2} A} \exp\left(-\frac{\eta^2 a^2}{2A^2}\right), \tag{9}$$

where a is the projected energy range, $A^2 = \eta^2\alpha^2 + \eta'^2\beta^2$, α and β are the energy range stragglings along the longitudinal and lateral directions, respectively, $\eta' = \sqrt{1-\eta^2}$. With increasing incident angle, deposited energy increases through

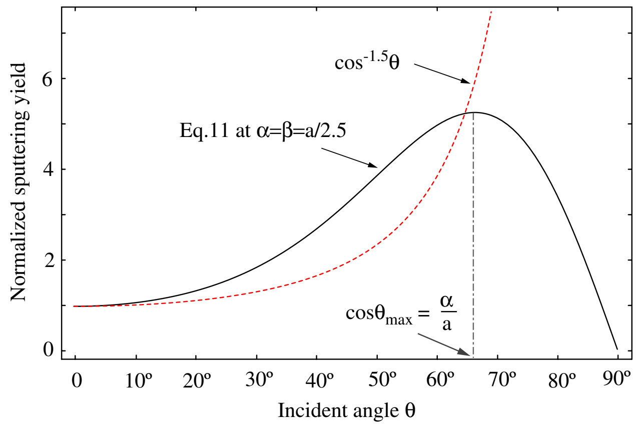

Figure 2. Normalized sputtering yield as a function of incident angle from our model. Sigmund's theory (----) is given for comparison.

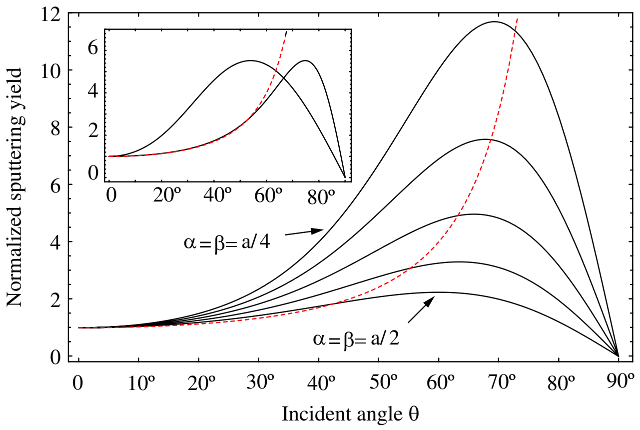

Figure 3. Angular dependence of sputtering yield given by equation (11) with different ratios of energy range to straggling: from top to bottom (symmetry case $\alpha = \beta$), $a/\alpha = 4, 3.5, 3, 2.5, 2$, inset showing the sputtering yield for the asymmetry case: from left to right $a = 2.5\alpha = 1.5\beta$, $a = 2.5\alpha = 4\beta$. The dashed curve shows the Sigmund theory given by $\cos^{-2}\theta$.

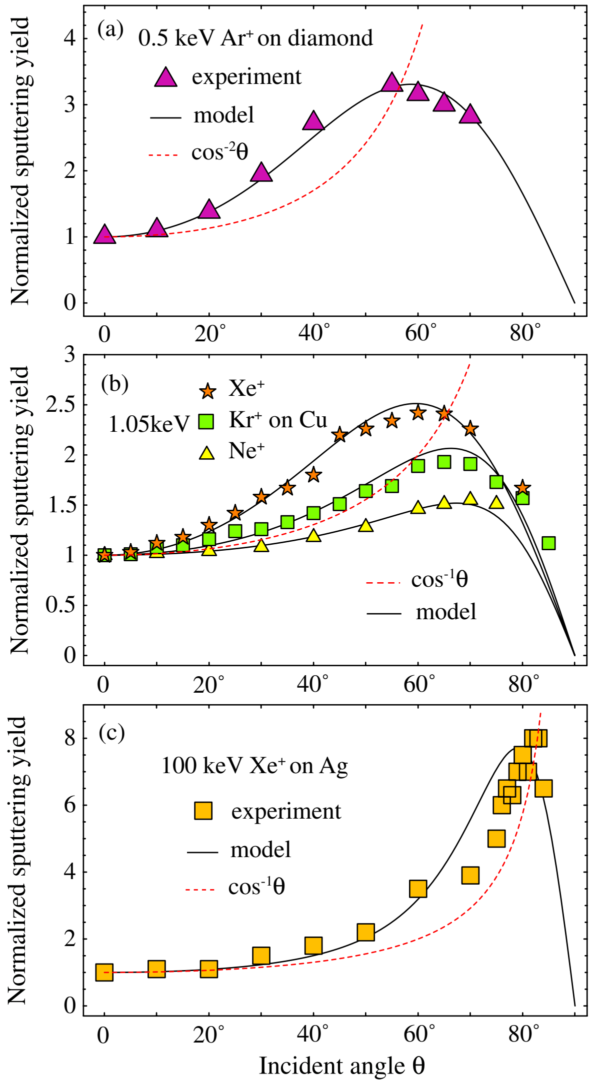

Figure 4. Comparison of angular dependence of sputtering yield predicted by the model with experimental results. *(a)* 0.5 keV Ar ion on diamond [22], $a = 18$ Å, $\alpha = 8$ Å, $\beta = 10$ Å. *(b)* 1.05 keV Xe, Kr and Ne ions on Cu [23], for Xe on Cu, $a = 17$ Å, $\alpha = 8$ Å, $\beta = 9$ Å; for Kr on Cu, $a = 11$ Å, $\alpha = 6$ Å, $\beta = 4$ Å; for Ne on Cu, $a = 16$ Å, $\alpha = 9$ Å, $\beta = 6$ Å. *(c)* 100 keV Xe on Ag [24], $a = 156$ Å, $\alpha = 59$ Å, $\beta = 35$ Å.

the exponential term while the corresponding depth of the sputtered atoms decreases through the cosine term. A balance between these two terms gives rise to the peak position of sputtering yield (figure 2).

Substituting equation (9) into equation (8) and letting the derivative of equation (8) in terms of η be zero, we have the incident angle $\theta_{\max}$ in which the sputtering yield achieves its maximum value. For simplicity, we assume the symmetric case $\alpha = \beta$.

$$\cos\theta_{\max} = \frac{\alpha}{a}, \tag{10}$$

which means the maximum sputtering yield depends only on the energy range and straggling (deposited energy distribution). For deposited energy, if we assume $a \approx 2.5\alpha$, equation (10) shows the maximum sputtering yield will appear at $\theta_{\max} = 66^\circ$ (figure 2). This is in good agreement with the experimental observation showing the maximum sputtering yield takes place around $\theta = 70^\circ$. From equation (8), under the assumption of Gaussian distribution and symmetry case (equation (9)), the normalized sputtering yield is

$$\frac{Y(E,\theta)}{Y(E,\theta=0)} = \cos\theta \exp\left(\frac{a^2\sin^2\theta}{2\alpha^2}\right), \tag{11}$$

where we replace η by $\cos\theta$.

The variation of sputtering yield with energy range and straggling is shown in figure 3. With increasing ratio of projected range to straggling, the maximum value of the sputtering yield moves to larger incident angle. For higher energy and lighter ion, a/α becomes larger and the peak tends to move to higher incidence. This prediction is consistent with the experimental results: the higher the ion energy or the lighter the ion, the larger the incident angle for the maximum sputtering yield.

Figure 4 shows the comparison of the angle-dependent sputtering yield predicted by equation (11) with the experimental results for different energies and different ion–target systems. The quantitative values of the coefficients a, α, β for energy distribution can be found using the theory of Winterbon *et al* [25] from the corresponding values for the ion distribution using the Monte Carlo simulation code SRIM [26]. We can observe that the theoretical predictions in equation (11) compare fairly well with the experimental data.

It is well known that the average projected energy range is given by $\langle x_\theta \rangle = \eta \langle x_0 \rangle$ [25], where $\langle x_\theta \rangle$ and $\langle x_0 \rangle$ are the average damage depths at off-normal and normal bombardment, respectively. This relationship was derived from the linear Boltzmann transport equation under the same assumptions as those in Sigmund's theory. For approximation with $m = 0$ in the Thomas–Fermi cross section, the average projected energy range is energy independent [1, 21, 25]. This means the recoil atoms with different energies, including sputtered atoms at the surface which have energy larger than surface bonding, satisfy the same equation describing the relationship for the range between normal and off-normal bombardment. Therefore, we can assume the average depth of recoil atoms is equal to the average depth of sputtered atoms. This can be confirmed by equation (7). The average depth of recoil atoms is given by $2\eta/(\pi\lambda_0 N a^2)$, where $2/(\pi\lambda_0 N a^2)$ is the average depth at normal bombardment which agrees very well with the estimate of the sputtered atom depth $(3/4)\cdot 2/(\pi\lambda_0 N a^2)$ given by Sigmund [1].

In summary, we have derived an expression for interpreting the evolution of sputtering yield as a function of incident angle based on Sigmund's theory. We showed that the peak of angular dependence of sputtering yield results from two competitive effects: increased energy deposited on the surface by the incident ion and decreased depth travelled by the sputtered atom. The results predicted by this model are in good agreement with experimental observations.

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