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The effect of microrelief evolution on the angular dependence of polycrystalline Cu sputtering yield

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Abstract

The dynamically steady-state surface relief of polycrystalline copper sputtered by high-dose (10^{18} – 10^{19} ion/cm²) 30 keV Ar⁺ ion bombardment at room temperature has been traced as a function of the ion incidence angle θ_i in a wide (0–80°) angular range. For each incidence angle the distribution of local slope angles of relief facets $f(\beta)$ in ion incidence plane and the corresponding distribution of the local angles of incidence $w(\theta)$ have been measured using the laser gonio-photometry (LGP) technique. It has been found that the distribution $f(\beta)$ has a narrow peak at grazing ion incidence, two wide peaks at oblique, and three peaks at normal ion incidence. The obtained results are discussed in terms of the theory of surface erosion under ion bombardment. A correction to the angular dependence of the sputtering yield was produced using the OKSANA computer code simulation for a smooth copper surface and the measured distributions $w(\theta)$. The experimental data and the calculation results agree well in the angular range $\theta_i = 0$ –60°. At $\theta_i > 60^\circ$ the experimental points are close to the simulated curve for a smooth surface.

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1. Introduction

Sputtering is an ubiquitous process in all work with ion beams and hence the phenomenon has a numerous applications in different fields of science

and technology. High-dose sputtering is a key problem for the ion-surface interaction relevant to nuclear fusion technology, thin film deposition, ion milling and polishing, and the creation of repetitive surface structures [1,2].

The high-dose irradiation of solids usually results in an alteration of the initial surface relief and development of dynamically steady-state surface topography. The ion-induced surface

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topography may change both the absolute values of the sputtering yield, and its dependence on the nominal angle of the ion incidence $Y(\theta_i)$, as well as the spatial distributions of sputtered particles [3–5]. It has been found, that the experimental dependence $Y(\theta_i)$ for polycrystalline Cu obtained at high-dose bombardment by 30 keV Ar^+ ions differs noticeably from the simulated curve for a smooth copper surface [6,7]. This discrepancy was explained by the effect of ion-induced surface topography, cf. [8,9].

In the present paper a correlation between the angular dependence of the sputtering yield for polycrystalline Cu and the characteristics of the microrelief developed under intense 30 keV Ar^+ ion bombardment is examined. For characterization of the microrelief the laser reflectometry technique was used [10,11]. Then, taking into account the obtained data, the correction to the angular dependence of the sputtering yield simulated for a smooth copper surface, using the computer code OKSANA [12,13], was produced. The corrected angular dependence of the sputtering yield was compared to the experimental curve.

2. Experimental

The ion irradiation was performed using the mass-monochromator of the Institute of Nuclear Physics, Moscow State University [14]. The irradiation conditions and the target samples were totally identical to the ones used in our preceding studies of sputtering the polycrystalline Cu by 30 keV Ar^+ ions [6,7]. The scheme of mounting the targets in the collision chamber was presented in Ref. [15]. The target holder allowed a variation of the angle of incidence from 0° to 89° with some steps 0.25° . The irradiation was carried out with a 30 keV Ar^+ ion beam of density $0.1\text{--}0.3\text{ mA/cm}^2$ and cross-section 0.35 cm^2 . The total ion fluence was $10^{18}\text{--}10^{19}\text{ ion/cm}^2$. The target temperature during irradiation was $\leq 60^\circ\text{C}$. Before irradiation the samples were mechanically polished, chemically etched, and washed in ethanol. As a result, the traces of both the mechanical treatment and the oxidization disappeared. A laser goniophotometry (LGP) technique was used to analyze both the

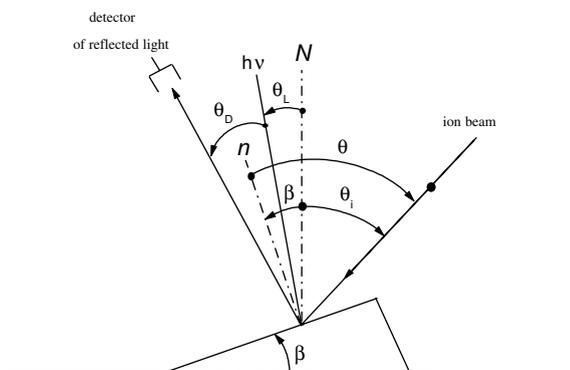


Fig. 1. Model of ridge-like structure and scheme of the experiment: $h\nu$ denotes the laser beam direction; θ_i is the nominal angle of ion incidence; θ is the local angle of ion incidence; θ_L is the laser beam angle of incidence relative to the nominal normal N ; θ_D is the detector location angle relative the laser beam direction; $\beta = (\theta_L + \theta_D)/2$ is the local angle of facet slope.

initial target surface and the ion-induced one. A He–Ne laser with $\lambda = 0.63\ \mu\text{m}$ was used as probing light source. The reflected light was detected by a semiconductor photodiode in the laser incidence plane, which usually was parallel to the ion incidence plane (see Fig. 1), where θ_D is the angle of reflected light detector location relative to the laser beam direction. In each measurement the series of reflected light indicatrix $I(\theta_D)$ for the fixed angles of the laser beam incidence θ_L and the reflectograms $I(\theta_L)$ for fixed θ_D were measured. The angle $\theta_L = 0^\circ, \pm 45^\circ$ and $\pm 70^\circ$ ($\theta_L > 0$ if located with θ_i at different sides from the normal N).

3. Simulation

The simulation of the argon interaction with copper was performed using the computer code OKSANA [12,13]. The code is based on the binary collision approximation and takes into account weak simultaneous collisions at large distances. The polycrystalline target is modelled by a single crystal of random orientation varying from impact to impact. The classic scattering in atomic collisions is described by the repulsive potentials, namely, the WHB (KrC) potential for Ar–Cu interaction and the MR potential for Cu–Cu

interaction. The WHB potential [16] allows to reproduce very well the Ar–Cu semichannel focusing energy found experimentally [17]. The MR potential refers to the Moliere potential with Robinson’s screening length $a = 0.075 \text{ \AA}$ [18] found to fit the experimental data on the threshold displacement energies in a row of metals including Cu. The inelastic energy losses were calculated using the Firsov formula. Allowance was made for the uncorrelated thermal vibrations in terms of the Debye model at 300 K. The surface barrier was planar and the heat of sublimation (3.53 eV) was taken as the surface binding energy. A flat surface was assumed. All other parameters were identical to the standard model [19,20].

4. Results and discussion

The reflected light features are often used to obtain the parameters of a rough surface [10,11]. The processing of the reflected light data depends on roughness degree and is different for mirror surfaces and for the surfaces, which reflect light diffusely [10]. Previously we studied the polycrystalline Cu topography developed under the same irradiation conditions using SEM [6,7]. It was found, that both at normal and oblique ion incidence the target surface is covered with cone populations, cone superpositions and also flattened out cones. The simplest model of such relief is the model of two-dimensional ridge-like structure, see Fig. 1.

At grazing ion incidence the smooth furrows parallel to the ion incidence plane were observed, see Fig. 5(a) in Ref. [7]. The typical sizes of the topographical elements in the ion-induced relief were several μm . Analysis of both the indicatris $I(\theta_D)$ and the reflectograms $I(\theta_L)$ measured in the present paper shows that the ion-induced Cu surface produces a diffuse light scattering typical for the rough surfaces with the unevenness dimensions comparable with the wavelength of the probing visible light. According to the diffuse reflection model [10,11] the indicatris $I(\theta_D)$ and the reflectograms $I(\theta_L)$ may be transformed into the reflected light intensity function $I(\beta)$ for the local slope angle $\beta = \theta_L + \theta_D/2$ and the distributions $f(\beta)$ (i.e. the distributions of the projections of

the local normals n on the target basal plane). In particular, as a first approximation $f(\beta) \propto I(\theta_L) \cdot \cos \theta_L$.

Typical distributions $f(\beta)$ are presented in Fig. 2. One can see that before irradiation the surface

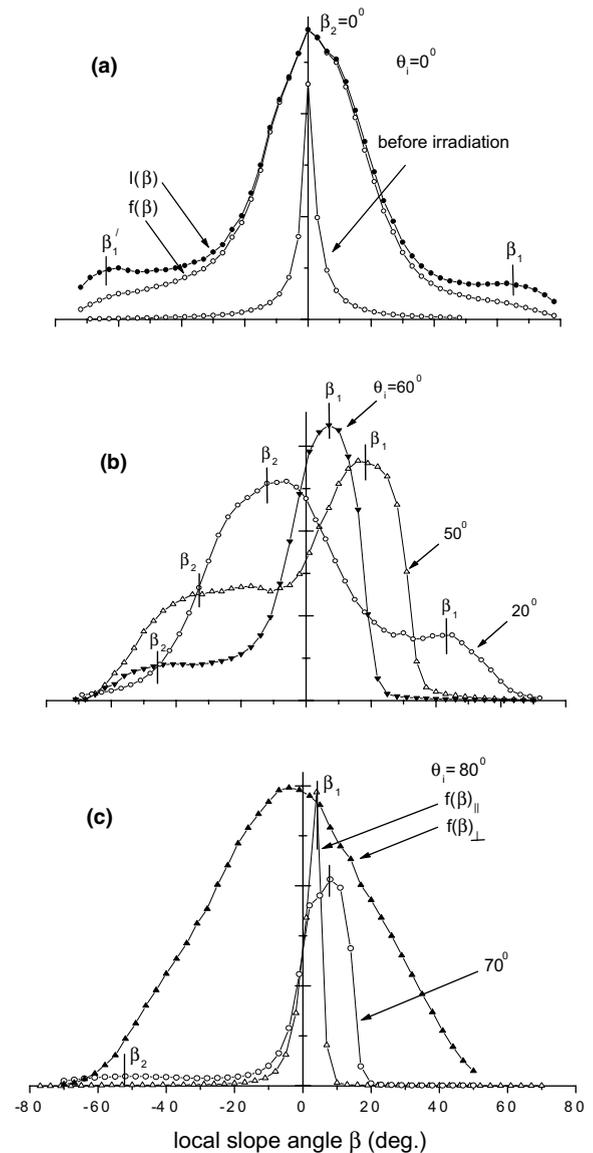


Fig. 2. The non-normalized distributions of local slope angle of relief facet $f(\beta)$ for polycrystalline Cu bombarded by 30 keV Ar^+ ions at different nominal angles of ion incidence: (a) normal incidence $\theta_i = 0^\circ$, the $I(\beta)$ dependence is also presented; (b) $\theta_i = 20^\circ, 50^\circ, 60^\circ$; (c) $\theta_i = 70^\circ, 80^\circ$.

relief is characterized by a narrow symmetrical distribution $f(\beta)$ with the half-width $\approx 6^\circ$. Ion irradiation essentially alters the surface microtopography, and at similar conditions, the modification is determined by the nominal angle of ion incidence. At normal ion incidence ($\theta_i = 0$) the developed relief results in a wide cupola-shaped distribution of local angles β : the main maximum is observed at $\beta = \beta_2 \approx 0$ and two less pronounced maxima – at $-\beta'_1 \approx \beta_1 = 68^\circ$ (see Fig. 2(a)). The maxima of $f(\beta)$ correlate with the certain topographical elements of the ion-induced relief and the main maximum positions characterize the average slope of the facets. So, at normal ion incidence the maximum at $\beta_2 \approx 0$ corresponds to the smooth microareas with basal surface orientation. The maxima at $\beta = \pm 68^\circ$ reflect the average slope of the ridge-like elements of the surface topography. According to our previous SEM studies, in the case of normal ion incidence some cone populations covered the copper surface. The slope angle of lateral cone facets relative the basal surface was $\approx 77^\circ$ [6].

At oblique ion incidence ($\theta_i = 10\text{--}60^\circ$) the distributions $f(\beta)$ are wide and have two maxima (see Fig. 2(b)) located nonsymmetrically relative to $\beta = 0$, i.e. the topographic surface elements with small local angles β cease to dominate in the ion-induced relief to the contrary of the normal incidence case. At grazing ion incidence ($\theta_i = 70^\circ, 80^\circ$) a sharp narrowing of the $f(\beta)$ -distributions was observed, see Fig. 2(c). As θ_i increases, the β_1 -maximum approaches $\beta = 0$, and at $\theta_i = 80^\circ$ the facets with $\beta_1 \approx 4^\circ$ dominate. The study of cone structures using three-dimensional reconstruction technique by computer processing of the SEM-stereopairs, allows to obtain a map of relief isolines and profilograms for chosen directions, that gives a possibility to determine the parameters of the topographical elements [6,7]. In particular, in the case of ion irradiation of Cu at $\theta_i = 82^\circ$ the analysis of the profilograms, parallel to the ion incidence plane and passing through flatted out cone top, has shown that the “upper” side of cone makes with the basal target surface an angle up to $\approx 5^\circ$. This is close to the result obtained in the present work using the LGP (4° at $\theta_i = 80^\circ$). As it was noted above, at grazing ion incidence the furrows are virtually parallel to the ion

incidence plane. For this reason at $\theta_i = 80^\circ$ the $f(\beta)$ -distributions were measured in two planes: in the ion incidence plane ($f(\beta)_\parallel$) and in the perpendicular one ($f(\beta)_\perp$), see Fig. 2(c). One can see that the furrows have a wide range of the local angles β_\perp that results in strong probing light scattering. The spatial distribution of reflected light looks like a narrow bright band in a direction oriented perpendicular to the ion incidence plane. The half-width of the $f(\beta)_\perp$ is $\approx 58^\circ$ which correlates with the data obtained in Refs. [6,7].

So, the maxima of the $f(\beta)$ -distributions characterize the certain topographical elements of the ion-induced surface relief. One may conclude that the β_2 -maximum corresponds to the facets that were irradiated virtually at normal incidence, while the β_1 -maximum is due to grazing ion incidence. The Fig. 3 presents the dependencies of the β_1 and β_2 -maxima positions on nominal angle of incidence θ_i . One can see that as θ_i rises, β_1 decreases, while the module of β_2 increases (β_1 is assumed positive, β_2 -negative, see the scheme at the top of Fig. 3).

According to the theory of surface erosion, the relief evolution and the formation of steady-state surface topography under ion irradiation are determined by the sputtering yield dependence on local angle of incidence $Y(\theta)$. In particular, the extremes of $Y(\theta)$ are the characteristic points and determine a steady-state relief at prolonged ion irradiation [1]. In the studied case, the function $Y(\theta)$ calculated using the computer code OKSANA has two extremes: a minimum at $\theta = 0$ and a maximum at $\theta_{\max} = 78^\circ$. In the Fig. 3 the values of the characteristic local angles were presented as straight lines $\beta = 78^\circ - \theta_i$ and $\beta = -\theta_i$. One can see that these lines at $\theta_i \leq 60^\circ$ describe reasonably the dependencies of average facet slopes local angles $\beta_1(\theta_i)$ and $\beta_2(\theta_i)$. The dotted line in the Fig. 4 corresponds to the limiting angle of ion incidence $\theta = 90^\circ$ when the bombarding ions move along the facet and virtually do not sputter it. The density of the distribution of the facets with $\beta > 90^\circ - \theta_i$ at given θ_i is negligible, see Fig. 3. In other words, at prolonged ion irradiation all surface areas are subjected to sputtering at θ raising from 0° to 90° , cf. Ref. [8].

When the calculations or simulations of the rough surface sputtering were produced, two main

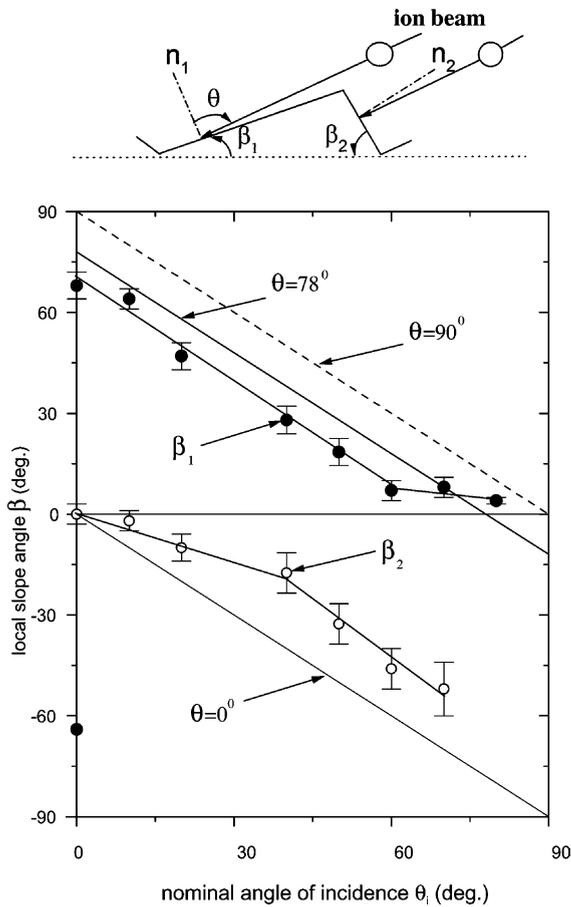


Fig. 3. The dependencies of maxima locations of local slope angle of relief facet β_1 and β_2 on nominal angles of ion incidence θ_i . Scheme of ridge-like surface structure is also presented.

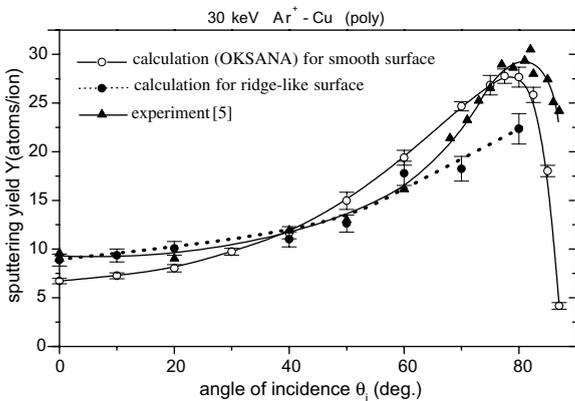


Fig. 4. The angular dependencies of the sputtering yield of polycrystalline Cu bombarded by 30 keV Ar^+ ions.

effects were distinguished which determine the differences in sputtering of the smooth and rough surfaces. Namely, the distribution $w(\theta)$ of the local angles of incidence θ on a rough surface and the shadowing by the topographical element facets of the sputtered particles [8,9]. In terms of ridge-like surface model, a normalized distribution

$$w(\theta) = W(\theta) / \int_{-\pi/2}^{\pi/2} W(\theta) d\theta \quad (1)$$

may be connected to the distribution of local slope angles $f(\beta)$ by the ratio $W(\theta) = f(\beta) \cdot \cos(\beta + \theta_i) / \cos \beta$. If $Y(\theta)$ is the dependence of the sputtering yield on the local angle of ion incidence on a smooth surface, the average yield value

$$\langle Y \rangle = \int_{-\pi/2}^{\pi/2} Y(\theta) \cdot w(\theta) d\theta \quad (2)$$

corresponds to a ridge-like surface sputtering at nominal angle of incidence θ_i . A comparison of the dependencies $w(\theta)$ obtained by the LGP, and the dependence of $Y(\theta)$ calculated by the code OKSANA for a smooth Cu surface shows clearly an influence of ion induced topography on surface sputtering. In particular, the wide distributions $w(\theta)$ at the nominal angles near normal ion incidence result into essential increase of $\langle Y \rangle$ for a rough surface compared to a smooth one. Fig. 4 presents the experimental angular dependence of the sputtering yield $Y(\theta_i)$ obtained earlier for the case of high-dose bombardment by 30 keV Ar^+ ions of polycrystalline Cu [6]. The dependence $Y(\theta_i)$ was simulated using the code OKSANA for a smooth Cu surface. The dependence $\langle Y \rangle(\theta_i)$ was obtained by the correction of simulated $Y(\theta_i)$ dependence taking into account the distributions of local angle of incidence $w(\theta)$. One can see that in the angular range between 0° and $\approx 60^\circ$ the surface roughness correction leads to a better agreement of the simulation results with the experimental data. However, at grazing ion incidence the experimental dependence $Y(\theta_i)$ is closer to the simulated curve for a smooth surface. Perhaps, it is connected with the change of the ion-induced topography from ridge-like type to furrow-like one for the studied combination Ar–Cu.

5. Conclusion

Using the laser goniophotometry (LGP) technique the ion-induced surface microtopography of polycrystalline Cu under high-dose 30 keV Ar⁺ ion bombardment has been studied in the range of nominal angle of incidence θ_i between 0° and 80°.

It has been shown that the measured distributions of local slope angles of relief facets $f(\beta)$ have some maxima, the positions of which depend on the nominal angle of ion incidence and reflect the topographical element geometry. The positions of maxima have been found to be close to the predictions of erosion theory for the dynamically steady-state conditions under high-dose bombardment.

The computer code OKSANA has been used to calculate the sputtering yield angular dependence for a smooth Cu surface, and the one with ion-induced surface topography created under 30 keV argon bombardment. The surface roughness correction leads to a better agreement between experiment and calculations in the angular range from $\theta_i = 0^\circ$ to $\approx 60^\circ$. At grazing incidence angles, the experimental data are in reasonable agreement with the simulation for a smooth copper surface. This may be connected with the change of topographical features from a ridge-like type to the smooth furrows.

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