Microtopography of Surfaces Eroded by Ion-Bombardment

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Conical protrusions have been observed in the scanning electron microscope on surfaces that have been eroded by sputtering under 5 to 8 keV A$^+$ ion bombardment. It is shown why these have a half-angle corresponding to the principal peak in the graph of sputtering rate versus angle of ion incidence. These lead to some general conclusions on the microtopography of ion-eroded surfaces.

1. Introduction

It is well known that the bombardment of solids with energetic ions causes atoms to be ejected or sputtered with a consequent erosion of the surface. Interesting changes in surface topography have frequently been reported although the mechanisms responsible are imperfectly understood. The purpose of this paper is first to report some microscopic surface features that have been formed on several solids by A$^+$ ion-bombardment and observed in a scanning electron microscope. This will be followed by a discussion of the dependence of the sputtering rate on the angle of ion incidence and how this may be used to interpret the observations.

2. The Experiment

The 14 kV scanning electron beam struck the surface of the specimen at about 45°, and the resolution of the microscope was between 300 and 500 Å. The ion beam, which was formed by a 5 to 8 kV radio-frequency ion source [1] attached to the microscope, was perpendicular to the electron beam. The apparatus is described by Stewart [2].

The mean ion current density on the specimen was between 100 and 500 μA/cm². At the centre of the fixed ion beam the current density was greater than 5 mA/cm² and it was therefore possible by observing different parts of the specimen to see whether the type of structure was

a function of ion current density for densities less than 5 mA/cm².

The electron collection system was a plastic scintillator covered with approximately 1000 Å of aluminium, and coupled to a photomultiplier by a perspex light guide. The specimen was observed before and after each period of bombardment. It was possible to observe the specimen during bombardment as the collection system was relatively insensitive to ions, electrons, or light from the ion beam or liberated by the ion beam from the specimen. Simultaneous observation was not normally used and a shield was placed between the specimen and the final lens of the microscope to prevent sputtered material from entering the microscope column.

In all cases the specimen was near 25°C during bombardment and before mounting it was cleaned by etching in a suitable reagent solution.

3. Observations

Fig. 1 shows the surface of a tin crystal following a bombardment with 5 keV A$^+$ ions to a mean dose of 0.26 C/cm². From published data on sputtering rates [3] such a bombardment should have eroded the surface to an average depth of the order of 10 μm. The conical protrusions with their axes along the direction of ion incidence seen in fig. 1 were a common feature on all the tin surfaces observed. Cones were also seen on other materials and fig. 2 shows one formed on a
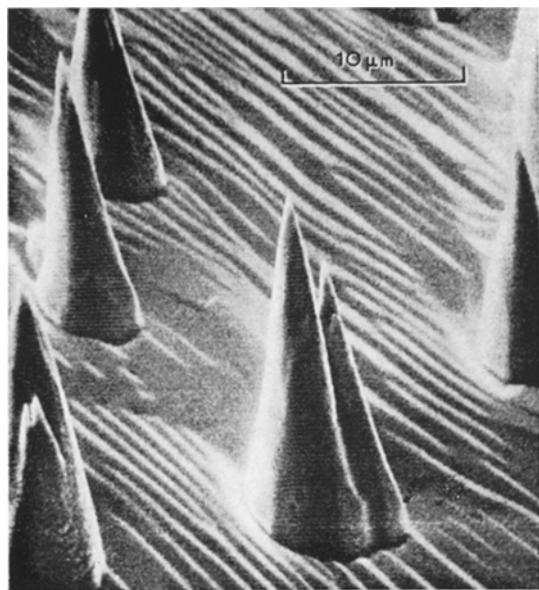
Figure 1 The surface of a tin crystal following a bombardment with 5 keV A⁺ ions to a dose of 0.26 C/cm².
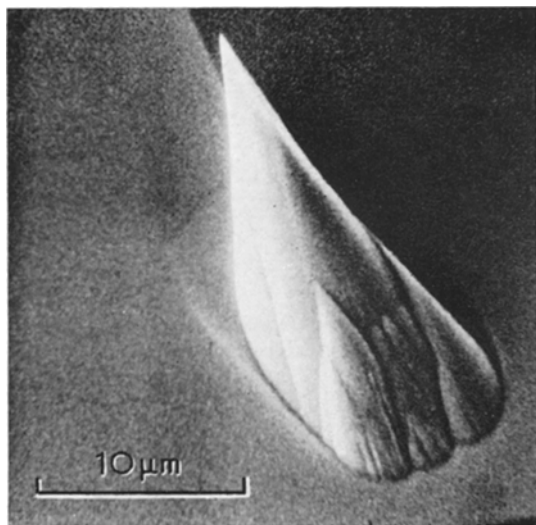

Figure 2 The surface of a silicon crystal after a similar bombardment to that in fig. 1.

silicon crystal by a similar bombardment.

As the electron and ion beams were perpendicular it was possible to obtain a reasonably true view of the profile of the spike without moving the specimen, and it was possible to take stereo pair micrographs when accurate measurements of size were required.

There appeared to be a correlation between the number of cones and the presence of foreign inclusion in the specimen. The tin crystal was grown in graphite mould and was therefore expected to contain graphite and oxide particles. In the early stages of erosion many cones terminated in a small blob sometimes less than 1 μm in diameter, which shrank and eventually disappeared as bombardment proceeded. It is thought that these blobs were foreign particles, initially present as inclusions in the crystal and uncovered during the erosion of the surface. Apparently, by shielding the surface immediately beneath them they were able to cause the formation of a conical protrusion.

In experiments with high-purity, zone-refined, aluminium such cones were rarely seen unless fine dust particles were introduced on to the surface. Protrusions with conical sides then developed beneath the dust particles.

The shape of the spikes did not seem to be a function of current density over the range of current densities available. At very low current densities the results will not be meaningful as the rate of arrival of the ions at the specimen was less than the rate of arrival of gas molecules from the surrounding vacuum. (The specimen was surrounded by a baffle cooled by liquid nitrogen, and the pressure measured outside this baffle by an ion gauge was between 10^{-5} and 10^{-6} torr.)

4. Discussion of the Sputtering Mechanism

The ejection of atoms from a crystal surface by sputtering is due to atomic collisions in a surface layer of the crystal induced by the bombarding ion. The effective depth of this layer, R, depends on the penetration of the ion and on the efficiency of kinetic energy transfer through the crystal and may involve mechanisms such as channelling and the focused collision sequence [4]. In the cases under discussion we may expect R to be of the order of 10 Å. The discussion will be limited to ions of sufficiently high energy that they penetrate deep into the crystal, far beyond the depth R when incident normal to the surface.

We define the sputtering ratio $S(\theta)$ as the number of atoms ejected by each ion incident at an angle θ to the surface normal. This may safely be assumed to be proportional to the energy deposited in the surface layer of depth R. If dE_1/dx is the rate at which the ion loses its energy E_1 in collision with the atoms of the

crystal, the energy lost in the surface layer, in which the path length is $R\sec\theta$, is

$$\int_0^{R\sec\theta} \frac{dE_1}{dx} \, dx. \qquad (1)$$

Provided that E_1 is less than about M_1 keV, where M_1 is the atomic mass number of the ion, energy loss to electron excitation may be neglected [5].

When one assumes that the potential energy of interaction between ion and atom is of the inverse-square form,

$$V(r) = (2E_R/e\,(Z_1Z_2)^{5/6}\,(a_0/r)$$

it can be shown that dE_1/dx can be approximately written:

$$dE_1/dx = (\pi^2/4)\,a^2nE_a \qquad (2)$$

valid for $E_1 \lesssim E_a$. Here E_R is the Rydberg energy 13.6 eV, e is 2.718, Z_1 and Z_2 are the atomic numbers of ion and atom, M_1 and M_2 the corresponding mass numbers, a_0 is the Bohr radius 0.53 Å, n the density of atoms per unit volume and E_a the value of E_1 that allows the ion and atom to approach to a distance $a = a_0/(Z_1Z_2)^{1/6}$ in a head-on collision, i.e.

$$E_a = 2E_R(Z_1Z_2)^{7/6}\,(M_1 + M_2)/M_2e \qquad (3)$$

(The derivation of these formulae will be found in a book by Thompson [5].)

Expression 2 shows that dE_1/dx is independent of energy, thus the integral in equation 1 is trivial and we can write for the sputtering ratio

$$S\,(\theta) = (\pi^2/4)\,sa^2nE_aR\sec\theta \qquad (4)$$

valid for $E_1 \sim E_a$ where s is a crystal constant giving the number of atoms ejected per unit energy deposited in the surface layer.

For polycrystalline solids, where one averages over many orientations, such a dependence on $\sec\theta$ is roughly true for angles θ up to about $60°$ at energies of the order of E_a (1 to 100 keV, depending on Z_1 and Z_2). With single crystals, anisotropies occur owing to effects such as channelling [3, 6-8]. These will be neglected in the first approximation.

At large angles θ the sputtering ratio increases less slowly than $\sec\theta$, passes through a maximum between 70 and $80°$ and falls towards zero at $\theta = 90°$, as shown in fig. 3 [6, 7]. It is suggested that this occurs because, as θ increases, the incident ion has an increasing probability of being reflected without traversing the effective surface

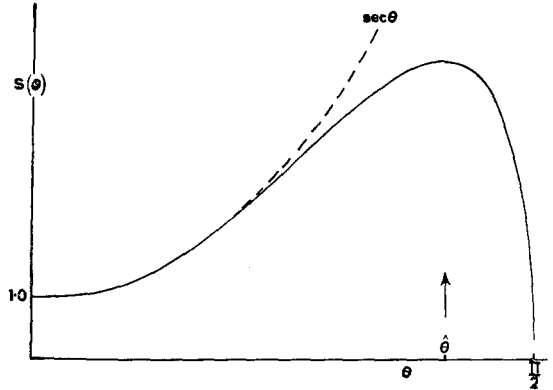layer. Hence the energy deposited eventually falls with increasing θ.

Figure 3 Schematic graph of sputtering ratio versus angle of ion incidence.

A simple physical criterion may be applied to determine $\hat{\theta}$, the angle at which $S(\theta)$ reaches its maximum value: when $\theta = \hat{\theta}$, reflection of the ion from the potential barrier associated with the surface plane of atoms prevents penetration [9]. Lindhard [10] shows that the critical angle for such reflection is given by

$$\frac{\pi}{2} - \hat{\theta} = \sqrt{\frac{5\pi a_0^2\,n^{2/3}Z_1Z_2E_R}{(Z_1^{2/3} + Z_2^{2/3})\,E_1}}. \qquad (5)$$

Thus when $\theta > \hat{\theta}$, penetration is not possible.

Equation 5 implies that $\hat{\theta}$ increases with ion energy, which is reasonable since the ion can more easily penetrate the surface potential barrier at higher energies. The effect of increasing either Z_1 or Z_2 is to decrease $\hat{\theta}$, since both parameters increase the surface potential. If values appropriate to 5 keV A^+ ions on Sn are inserted in equation 5 one obtains $\hat{\theta} \approx 80°$, which seems reasonable as a rough estimate.

5. Interpretation of Surface Features

Referring to fig. 4, consider a surface consisting of two inclined planes A and B on which bombarding ions are incident at angles α and β respectively. During erosion the planes move to positions A' and B' and their line of intersection moves from O to O'. The angle δ is made between the direction of ion incidence and the path OO' followed by the intersection. The distances advanced by the planes are a and b, that advanced by the intersection is c and the sideways movement of the intersection, transverse to the direction of incidence, is d. The flux of ions crossing unit area of surface normal to their
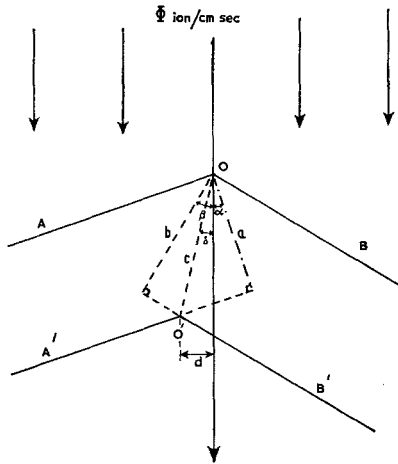
Figure 4 The motion of the intersection between two plane surfaces during erosion.

direction is Φ in the course of the erosion. On unit areas of the two planes the respective fluxes are $\Phi \cos \alpha$ and $\Phi \cos \beta$ and the distances advanced are

$$a = \frac{1}{n} \Phi \cos \alpha \, S(\alpha) .$$

$$b = \frac{1}{n} \Phi \cos \beta \, S(\beta) .$$

From the geometry of the system,
$$a = c \cos (\alpha + \delta)$$
$$b = c \cos (\beta - \delta).$$
We can use these equations to calculate the ratio d/a which relates the sideways movement of the intersection to the distance by which plane A advances:

$$\frac{d}{a} = \frac{S(\beta) - S(\alpha)}{S(\alpha) \cos \alpha \, (\tan \alpha + \tan \beta)} . \quad (6)$$

This equation provides the means of deciding whether the intersection line O moves towards A or towards B during erosion (i.e. whether d is positive or negative).
(i) If $S(\alpha) = S(\beta)$, $d = 0$ and O does not move laterally;
(ii) if $S(\alpha) > S(\beta)$, $d < 0$ and O moves into B;
(iii) if $S(\alpha) < S(\beta)$, $d > 0$ and O moves into A.

If the angle between the planes had been acute, viewed from above, rather than obtuse, the directions of motion would have been reversed.

Thus the crest of a ridge will move towards the side for which S(θ) *is least. The foot of a valley will move towards the side for which* S(θ) *is greatest.*

Now the significance of our $S(\theta)$ function becomes apparent.

In the special case where $\alpha = 0$ and the ions are incident normally on plane A, equation 6 becomes

$$\frac{d}{a} = \frac{S(\beta) - S(0)}{S(0) \tan \beta} \quad (6a)$$

and if $\beta \sim \theta$, consideration of fig. 3 shows that this is of order unity; consequently such a ridge should move sideways at about the same rate as it moves downwards.
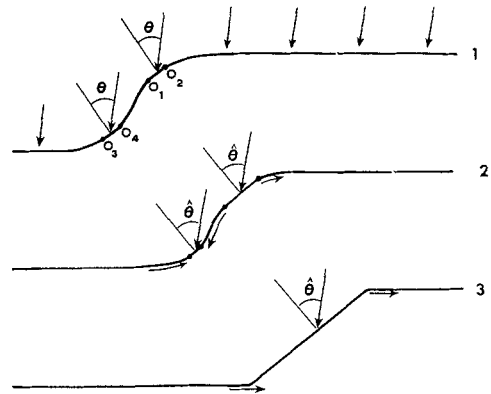

Figure 5 The erosion of a surface step.

Consider a step on a surface undergoing erosion by ion bombardment, as shown in profile 1 of fig. 5. This can be treated as though it were an assembly of planar facets, such as O_1O_2 on the convex part of the step and O_3O_4 on the concave part of the surface. Suppose the function $S(\theta)$ has the form shown in fig. 3, then, by applying the above criterion to the corners at O_1, O_2, O_3 and O_4, we deduce:

(i)   if $\theta < \hat{\theta}$ $\begin{cases} O_1 \text{ and } O_2 \text{ move to the right} \\ O_3 \text{ and } O_4 \text{ move to the right}; \end{cases}$

(ii)   if $\theta > \hat{\theta}$ $\begin{cases} O_1 \text{ and } O_2 \text{ move to the left} \\ O_3 \text{ and } O_4 \text{ move to the left}; \end{cases}$

(iii) if $\theta = \hat{\theta}$ $\begin{cases} O_1 \text{ moves to the left} \\ O_2 \text{ moves to the right} \end{cases}$
         and the facet grows

$\begin{cases} O_3 \text{ moves to the right} \\ O_4 \text{ moves to the left} \end{cases}$
         and the facet shrinks.

The effect on the convex part of the step is to form a facet with angle of incidence $\hat{\theta}$, whilst the concave part develops a smaller radius of curvature as the corners for which $\theta < \hat{\theta}$ run towards
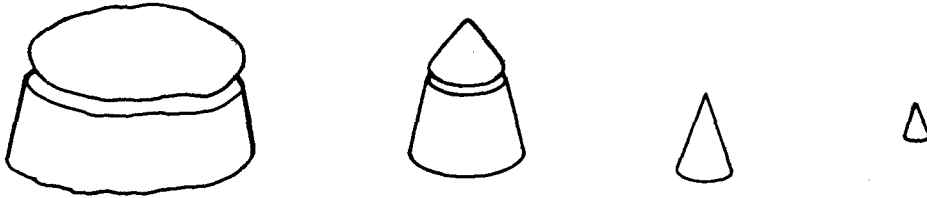
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Figure 6 Stages in the formation of a cone.

those for which $\theta > \hat{\theta}$, eventually making a single concave corner. Finally one obtains the profile 3 in which a single facet making an angle of incidence $\hat{\theta}$ moves across the surface.

Note that if $S(\theta)$ has more than one peak, owing to crystallographic effects, facets will grow at first corresponding to each peak. But when these meet, the one for which $S(\theta)$ is greatest will always win and we will generally expect this to be at $\theta = \hat{\theta}$.

Now suppose a step forms under the edge of a foreign particle resting on the surface. As the particle itself shrinks the step will be inclined at an angle $\hat{\theta}$ and eventually when the particle is removed the step contracts inwards until it becomes a cone of angle $(\pi - 2\hat{\theta})$ with its axis along the direction of ion incidence (see fig. 6).

Equation 5 thus shows how to estimate the cone angle and the values obtained in the region of $20°$ seem in good agreement with the observations.

The observation by Wehner [6] that single-crystal spheres bombarded by 400 eV ions become conical with a half angle corresponding to the maximum in the sputtering yield curve can also be understood.

We thus have a reasonable explanation of the cone formation but must point out that the effect of surface diffusion processes, enhanced by the ion-bombardment, have been entirely neglected. Cases undoubtedly occur when this would not be

justified and if diffusion were the dominant mechanism, surface-faceting of crystal planes with low free energy might then result. In such cases one should expect dependence on ion current density, which is absent here.

A unique feature of these experiments is their ability to determine the position of $\hat{\theta}$ on a microscopically defined surface. Sputtering yield measurements are generally made only as a function of the angle of incidence to the *macroscopic* surface.

The sweeping of steps across a surface could form a basis for understanding etching and polishing by ion beams and other erosion phenomena of practical importance.

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