The Equilibrium Topography of Sputtered Amorphous Solids

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A theory is developed for the sputtering of amorphous solid by an ion beam and the changes in surface topography to which this sputtering leads. It is shown that surfaces which have pinning regions, such as vertical steps and included impurities, reach an equilibrium in which cones are developed on the surface. The theory is compared with several experimental observations of such conical development.

1. Introduction

It is well known that solid surfaces exposed to an energetic ion beam erode and frequently acquire well-defined topography. Etch pits and valleys, hillock and conical protuberances have been observed and there is evidence that some of the microscopic effects are closely related to the type of bulk damage produced within the solid, whilst more macroscopic effects are related to initial surface topography, such as impurity inclusions at a surface and variations in the sputtering coefficient as a function of the angle of ion beam incidence to the surface. Wehner [3] has reported that iron and some other metal spheres exposed to sputtering by a uniform ion beam, achieve conical equilibrium shapes and has also indicated, in agreement with similar investigations by Stewart and Thompson [2] that Al, Si and Sn surfaces develop conical protrusions, a feature also observed with sputtered alkali halides by Marinkovic and Navinsek [1]. Stewart and Thompson [2] have used geometrical arguments to show that the micro-cones are generated as a result of non-uniformities of the sputtering coefficient over a surface because of the presence of surface contamination, and similar arguments can be used to predict the topographical changes of an initially curved surface. These considerations lead to the conclusion that cones or steps will develop on a surface, where the sputtering coefficient is a function of the angle between the direction of ion incidence to the surface and the normal to the surface, such that the semi-vertical angle of the cone is equal to the angle of the ions to the normal at which the sputtering coefficient is a maximum. It is the purpose of the present communication to investigate the erosion of a surface in a more quantitative fashion, assuming an amorphous solid from which correlated collision sequences are absent (which lead to large fluctuations in the sputtering coefficient as a function of incidence angle), and where atomic density is direction-independent.

2. Theoretical Discussion

In the following discussion, it will be assumed that surface changes occur only as a result of atomic ejection, so that surface relaxation due to diffusion will be ignored, as will the possibility of redeposition of sputtered material.

The relatively few measurements of sputtering coefficient as a function of ion incidence angle generally suggest that the form of $S(\theta)$ is as depicted in fig. 1, with a value of S_n at normal incidence ($\theta = 0°$), increasing to a maximum at $\theta = \theta_p$ and decreasing to zero as $\theta \to \pi/2$ (grazing incidence). The exact form of $S(\theta)$ and the value of θ_p depend, amongst other things, upon ion type and energy and target material but are undoubtedly determined by the form of the energy deposition function with depth of the ion in the target. $S(\theta)$ the sputtering coefficient is defined and measured as atoms removed from the target per incident ion, so that if a beam of ϕ ions/sec strikes unit area of a surface at an angle θ to the normal and the target is com-

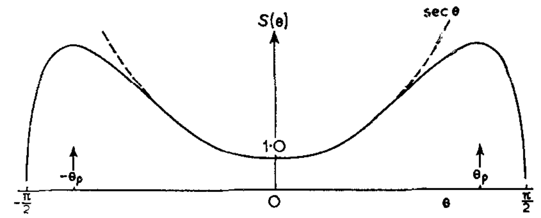

Figure 1 Typical representation of the variation of sputtering coefficient with angle of ion incidence.

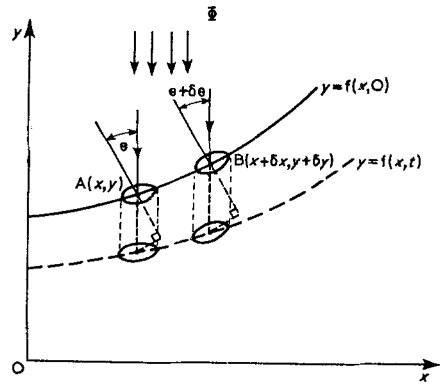

Figure 2 Geometry of erosion of a line contour by ion bombardment in the Oy direction.

pletely isotropic, then the rate of erosion of the surface *in the normal direction* is

$$\frac{S(\theta)}{n} \times \phi .$$

where n is the number of atoms per unit volume of the target.

Now consider the situation depicted in fig. 2, in which a section through a surface is considered at a time t in one plane only ($x0y$) and which is exposed to a uniform flux of ions in the $0y$ direction of magnitude ϕ ions/sec/unit length in the $0x$ direction. If the surface generator in the $x0y$ plane is described by a function $y = f(x, t)$ then, from consideration of the changes in the spatial location of two points A and B (with co-ordinates (x, y), $(x + \delta x, y + \delta y)$) on the generator with time, it is possible to express formally the time variation $f(x, t)$ and to determine the equilibrium condition precisely. If the angle between the ion incidence direction and the surface normal at A is θ (and at B, $\theta + \delta\theta$), the rate of bombardment per unit length of the generator at A is $\phi \cos\theta$ and the normal rate of recession of the surface at A is

$$\frac{\phi}{n} S(\theta) \cos\theta \cdot$$

The effective rate of recession in the $0y$ direction $-\partial y/\partial t$ is thus

$$\frac{\phi}{n} S(\theta) \frac{\cos\theta}{\cos\theta} = \frac{\phi}{n} S(\theta) .$$

$$\text{i.e. } -\frac{\partial y}{\partial t} = \frac{\phi}{n} S(\theta) . \tag{1}$$

At B, the rate of recession in tbe $0y$ direction is

$$-\frac{\partial}{\partial t}(y + \mathrm{d}y) = -\frac{\partial}{\partial t}\left(y + \delta x \frac{\partial y}{\partial x}\right)$$

and is given by

$$\frac{\phi}{n} S(\theta + \delta\theta) .$$

$$\text{i.e.} \quad -\frac{\partial}{\partial t}\left(y + \delta x \frac{\partial y}{\partial x}\right) = \frac{\phi}{n} S(\theta + \delta\theta) .$$

Expanding the right-hand side of this equation gives,

$$-\frac{\partial}{\partial t}\left(y + \delta x \frac{\partial y}{\partial x}\right) = \frac{\phi}{n}\left[S(\theta) + \delta\theta \frac{\mathrm{d}S(\theta)}{\mathrm{d}\theta}\right] \tag{2}$$

Subtracting 1 and 2 and proceeding to the limit leads to the result,

$$\frac{\partial}{\partial t}\left(\frac{\partial y}{\partial x}\right) + \frac{1}{\delta x}\frac{\partial y}{\partial x}\frac{\partial}{\partial t}(\delta x) = -\frac{\phi}{n}\frac{\mathrm{d}S(\theta)}{\mathrm{d}\theta}\frac{\delta\theta}{\delta x} .$$

For recession in y direction *only* where $\partial/\partial t$ $(\delta x) = 0$ then,

$$\frac{\partial}{\partial t}\left(\frac{\partial y}{\partial x}\right) = -\frac{\phi}{n}\frac{\mathrm{d}S(\theta)}{\mathrm{d}\theta}\frac{\mathrm{d}\theta}{\mathrm{d}x} = -\frac{\phi}{n}\frac{\mathrm{d}S(\theta)}{\mathrm{d}x} \cdot \tag{3}$$

Such a situation will occur when the bombarded surface is prevented from moving sideways (in the $0x$ direction). This can occur when a part of the curve is parallel to the beam ($0y$) direction such that sputtering is zero at that region. The vertically inclined region, therefore, has the character of a pinning region and will be important, therefore, in the sputtering of a completely spherical surface. This equation relates the time rate of change of the slope of the generator $y = f(x, t)$ to the contour variation of the sputtering coefficient $S(\theta)$ and, as will be indicated subsequently, allows formal specification of the approach of the curve to an equilibrium configuration. The equilibrium state itself will be reached when the time rate of change of the surface is zero, i.e. $\partial/\partial t(\partial y/\partial x) = 0$ for all x.

Thus, in equilibrium,

$$\frac{\phi}{n}\frac{\mathrm{d}S(\theta)}{\mathrm{d}x}=0\,. \qquad (4)$$

Hence,

$$\frac{\phi}{n}\frac{\mathrm{d}S(\theta)}{\mathrm{d}\theta}\frac{\mathrm{d}\theta}{\mathrm{d}x}=0 \qquad (5)$$

One solution of this equation is that $\mathrm{d}\theta/\mathrm{d}x=0$ or $\theta=$ constant for all values of x. Hence, if the curve $f(x, 0)$ is initially a straight line, $\partial\theta/\partial x=0$ initially and remains so. A continuous line of initial slope θ_i will merely recede in the negative y direction at a rate $\phi/n\ S(\theta_i)$ and will maintain its slope constant.

A second solution of equation 5 is that $\mathrm{d}S(\theta)/\mathrm{d}\theta=0$, which means that S is independent of θ and a curve of any initial slope will remain unchanged except for a constant recession in the negative y direction. Wehner [3] suggests this latter situation where $S(\theta)$ is independent of θ applies to several metals (Ag, Ni, Pt) under low energy Hg^{+} bombardment.

If $S(\theta)$ is a function of θ however, there will be a change in the surface topography during sputtering, except in the case of a plane. In addition, for any element of a curve for which $\mathrm{d}S/\mathrm{d}\theta=0$,

$$\frac{n}{\Phi}\frac{\partial}{\partial t}\left(\frac{\partial y}{\partial x}\right)=-\frac{\mathrm{d}S}{\mathrm{d}\theta}\frac{\mathrm{d}\theta}{\mathrm{d}x}=0\,,$$

the slope will remain unaltered. Fig. 1 shows that $\mathrm{d}S/\mathrm{d}\theta=0$ for $\theta=0$ and $\pm\,\theta_\rho$, so that regions of curves initially at these slopes, will remain unaltered. Let us consider how the slopes of various general curves with θ not equal to one of these turning values will change.

Consider a concave downward surface bombarded in the $0y$ direction as in fig. 3, in which in the portion to the left of the point A, θ is initially always greater than θ_ρ, and to the right of A, θ is less than θ_ρ. For the left-hand portion $\mathrm{d}\theta/\mathrm{d}x$ is negative and since $\theta>\theta_\rho$, $\mathrm{d}S/\mathrm{d}\theta$ is negative. Thus, from equation 3, $\mathrm{d}S/\mathrm{d}\theta\ \partial/\partial t(\partial y/\partial x)$ is negative and as the slope decreases, the curve tends towards a line at angle θ_ρ to $0x$, since as has been shown above, once $\theta=\theta_\rho$ equilibrium is attained. For the right-hand portion $\mathrm{d}\theta/\mathrm{d}x$ is negative, $\mathrm{d}S/\mathrm{d}\theta$ is positive for $\theta<\theta_\rho$, so that $\partial/\partial t(\partial y/\partial x)$ is positive, and the slope increases. Hence the curve tends to a line with an angle to the axis of θ_ρ in the equilibrium condition. Thus, the initial concave downwards curve becomes a slope with the angle to the $0x$ axis $=\theta_\rho$, a

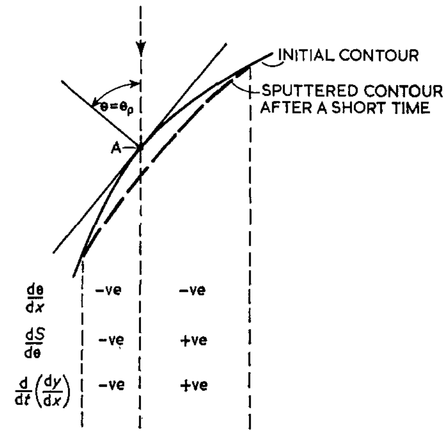

Figure 3 Conditions for equilibrium topography for sputtering of a particular curved surface.

conclusion reached by Stewart and Thompson [2]. If the curve of fig. 3 had been totally concave upwards but with θ always less than θ_ρ, then both $\mathrm{d}S/\mathrm{d}\theta$ and $\mathrm{d}\theta/\mathrm{d}x$ are positive and the equilibrium situation would result when the curved portion had relaxed to a horizontal line, i.e. $\theta\rightarrow 0$. On the other hand, if the contour had again been always concave upwards but such that $\theta>\theta_\rho$ then a vertical line would be the equilibrium configuration, since although $\mathrm{d}\theta/\mathrm{d}x$ is positive, $\mathrm{d}S/\mathrm{d}\theta$ is negative and $\partial/\partial t(\partial y/\partial x)$ is positive, so that $\theta\rightarrow\pi/2$. One would, therefore, expect a continuous curve, with θ ranging from $0\rightarrow\pi/2$ initially and concave upwards, to develop into a vertical step.

In the case of a surface contaminated with material of low sputtering coefficient (for example the surfaces studied by Stewart and Thompson [2]) a hillock will develop at the site of each contaminant atom or atom cluster. The surface is then analogous to our concave downward example of fig. 3, only we now have to consider a three-dimensional pattern. Clearly the line of uniform slope $\theta=\theta_\rho$ will again develop and is the generator of a cone of semi-vertical angle $\pi/2-\theta_\rho$.

The treatment of a contoured surface where there are no points of pinning is not amenable to the type of analysis considered here (which essentially applies to a surface constrained to move on the $0y$ direction only), and will be reported at a later date.

The relaxation to a $\theta=\theta_\rho$ contour however, is seen to depend upon the initial existence of concave downwards curvature, where $\mathrm{d}\theta/\mathrm{d}x$ is negative and where θ values both greater and less than θ_ρ are presented to the beam. Thus, in the bombardment of spheres, one expects to achieve conical equilibrium, as observed with iron and tantalum by Wehner [3]. For iron, the semi-vertical cone angle is $\sim 40°$ for 400 eV Hg bombardment and this compares very favourably with the θ_ρ value of $50 \pm 2°$, which may be deduced from Wehner's data. Wehner has, in fact, deduced a sputtering coefficient $S'(\theta)$ as a function of θ from the initial rate of erosion of spheres in a direction parallel to the ion beam, (see fig. 4). According to our earlier definition of $S(\theta)$, it is clear that $S'(\theta) = S(\theta)/\cos\theta$ and if Wehner's $S(\theta)$ values are multiplied by the appropriate $\cos\theta$ values, $\theta_\rho = 40 \pm 5°$ is obtained, as shown in fig. 4. It is clear from Wehner's discussion that the author expects an equilibrium conical form when $\theta = \theta_\rho$, i.e. when the attack angle is such that the sputtering coefficient is maximised; however, the reason for this correlation is not suggested.

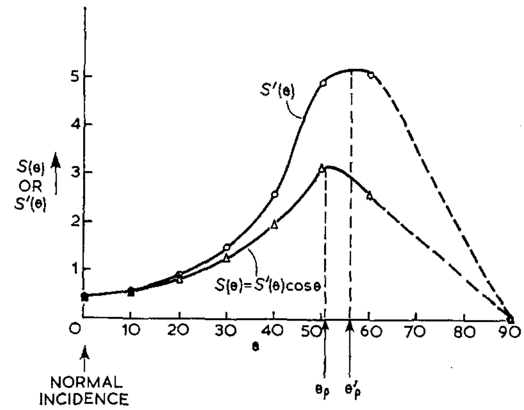

Figure 4 Variation of sputtering coefficient as a function of ion incidence for iron as measured by Wehner [3], and re-evaluated in the present work.

Conical production is expected when there is a symmetry of bombardment in the $x0y$ plane and a plane perpendicular to this, as in the case of a spherical surface or a plane surface with local contamination. It should be remarked however, that when micro-cones develop because of surface contamination, the development will only continue until the contaminant itself is eventually sputtered away. At that stage, the situation will become unstable, since the conical protrusion will then erode in the direction of the ion beam at a rate faster than the surrounding plane, so that the only true equilibrium will result when the cone is finally planed down, a fact noted by Wehner during continued bombardment of initially contaminated targets.

Finally, it should be noted that, if the ion flux is incident at some angle α to the $0y$ direction, the mathematical arguments are unchanged if the co-ordinate system is rotated through α. Thus, a plane surface with local irregularities when bombarded with ions at an angle α to the normal will develop cones having axes aligned with the beam direction and a semi-vertical angle $(\pi/2 - \theta_\rho)$. The stability of such angled cones will then be considerably influenced by the elastic properties of the target and some may snap off and overlay the surface, as observed by Marinkovic, and Navinsek [1] with alkali halides and these may form the basis of a tangled filigree structure, often observed with ion bombarded alkali halides.

More detailed considerations of the topographical changes in a contoured surface at non-normal incidence require evaluation of the effects of shadowing of adjacent features, a problem which is now occupying our attention.

References

1. V. MARINKOVIC and B. NAVINSEK, Proc. 3rd European Regional Conf. on Electron Microscopy, Prague, 1964. pp. 311-12.
2. A. D. G. STEWART and M. W. THOMPSON, *J. Materials Sci.* **4** (1969) 56.
3. G. K. WEHNER, *J. Appl. Phys.* **30** (1959) 1762.