JOURNAL OF MATERIALS SCIENCE 11 (1976) 1091–1098

# The influence of surface diffusion on topography development of an amorphous solid during sputtering

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The erosion and development of topography on an initially contoured amorphous solid due to the simultaneous action of ion beam sputtering and surface diffusion, is analysed. It is shown that derivation of formal expressions for the local rate of co-ordinate motion and radius of curvature is straight forward. Application of detailed prediction of the time varying behaviour of the surface profile is analytically difficult, however, and computational methods are suggested. The case of radiation enhanced surface diffusion is also considered briefly.

## 1. Introduction

In a series of four papers [1–4] the author and his colleagues have examined, theoretically, the development of topography of amorphous solids sputtered by ion beams. Other theoretical or computational studies of this problem have also been published [5–9] together with experimental observations [8, 10, 11] which, generally speaking, confirm theoretical predictions. In the theoretical work one major assumption has been that the amorphous solid is sputtered with a uniform flux of ions and that topography results only from the variation of the sputtering coefficient (number of sputtered solid atoms per incident ion) with direction of ion incidence to a surface element. Two exceptions to this assumption are in the work of Bayly [12] who allowed for a variable ion flux due to the contribution of sputtered atoms and ions reflected at small angles from a surface element on to another surface element, and in the studies of Sigmund [13] who considered the local variations in sputtering coefficient due to non-uniform energy deposition by an ion when surface features are of a size comparable to the dimensions of the zone near the solid surface where the ion deposits energy.

The assumption of the absence of other surface modifying processes which act during, or can be accelerated by, ion bombardment, such as surface and bulk diffusion and sublimation, has not yet been questioned, although recently several authors [14]–16] have found substantial changes in the topography developed upon polycrystalline Fe as a function of the temperature of the Fe. These changes have been tentatively ascribed to the increasing influence of surface diffusion with increasing temperature.

It is the purpose of this communication to present, as a preliminary step, the mathematical formalism which describes topography development when both sputtering and surface diffusion can occur during bombardment. It will be shown that although the formulation is straightforward, detailed predictions of the stages of topographic development are difficult and that computational methods may be required.

## 2. Theory

As in the author's previous studies, an amorphous solid is assumed so that there are no local perturbations in the assumed smoothly varying sputtering coefficient as a function of direction of ion incidence. This assumption is also important in the description of the surface diffusion process. Surface diffusion occurs both by atomic and defect migration over the macroscopic surface and by atomic and defect interchange between the surface and the bulk, the former process generally occurring at lower temperatures [17]. Without considering the detailed atomistics of either process, it is possible to describe the material flux parallel or perpendicular to a surface element in terms of the driving force for the transport process. The origin of this driving force is the chemical potential $\mu$ which exists at the boundary between different media, and there is a linear relationship between the appropriate gradient of this potential and the material flux. Since surface migration will probably occur first as a function of increasing temperature, in the following analysis we assume this to be the only diffusion process operating. Under these conditions the flux $J$ of surface diffusing atoms crossing unit length of a surface is given by [18–20]

$$J = \frac{D}{kTA_0}\nabla\mu \tag{1}$$

where $D$ is the surface diffusion coefficient at a temperature $T$, $A_0$ is the surface area per atom, and $k$ is Boltzmann's constant. The chemical potential $\mu$, from thermodynamic arguments [19], is a function of the radius of curvature of a surface and may be written

$$\mu = \mu_0 + \left\{\frac{\gamma}{R_1} + \frac{\gamma}{R_2} + \frac{1}{R_1}\frac{\partial^2\gamma}{\partial\theta_1^2} + \frac{1}{R_2}\frac{\partial^2\gamma}{\partial\theta_2^2} - \sigma\right\}\Omega \tag{2}$$

where $\mu_0$ is the chemical potential at a flat interface, $R_1$ and $R_2$ are the principal radii of curvatures of a surface element, $\sigma$ the surface normal stress (if any), $\Omega$ the atomic volume, $\gamma$ the surface energy or tension and the derivatives of $\gamma$ with respect to $\theta_1$ and $\theta_2$ are along the planes of principal curvature.

For an isotropic, amorphous substrate $\gamma$ will be a constant, independent of surface orientation or curvature, whereas for crystalline solids it is known that $\gamma$ is a function of $\theta$ often exhibiting extrema along specific crystal orientations. Thus, in our present assumption of an amorphous solid, with no applied stresses, Equation 2 relaxes to

$$\mu = \mu_0 + \left\{\frac{\gamma}{R_1} + \frac{\gamma}{R_2}\right\}\Omega. \tag{3a}$$

If, as in previous treatments of topographic development we consider a two dimensional surface contour only (i.e. a planar section through the solid), then Equation 3a can be re-written

$$\mu = \mu_0 + \frac{\gamma}{R}\Omega, \tag{3b}$$

where $R$ refers to the radius of curvature of an element of the surface contour in the $x0y$ plane considered.

For surface diffusion parallel to the plane of curvature, $\nabla\mu$ becomes $\partial\mu/\partial z$, where $z$ is length measured along the surface contour and thus Equation 1 is written:

$$J = \frac{D\Omega}{kTA_0}\frac{\partial}{\partial z}\left(\frac{\gamma}{R}\right). \tag{4}$$

Since the radius of curvature will be co-ordinate dependent upon an initial arbitrary surface contour, then the atomic flux due to surface diffusion will also be co-ordinate dependent. By considering the atomic flux difference crossing boundaries at lengths $z$ and $z + \delta z$ on the surface, it is readily shown that there may be an accumulation or depletion of surface material at any point, thus leading to an advance or regression of a surface element in the direction of the surface normal. Mullins [17–18] has shown that this normal rate of surface motion is given, from Equation 4 by

$$\left|\frac{\partial n}{\partial t}\right|_D = \frac{DN_0\Omega^2}{kT}\gamma\frac{\partial^2}{\partial z^2}\left(\frac{1}{R}\right) \tag{5}$$

where $N_0$ is the surface atomic density.

The sign of $\partial n/\partial t$ in Equation 5 thus depends upon the second derivative of $1/R$ with respect to $z$. The convention has been adopted in surface diffusion studies [18] that the local radius of curvature $R$ between a solid and vacuum is a positive quantity towards the $0x$ axis for a convex bulge upon a solid and measured negative for a concave surface trough. If, as earlier [1–4], $\theta$ is the angle between the ion beam (the $0y$ direction) and the normal to the surface at a given point (equal to the angle between the tangent to the surface at that point and the $0x$ axis) then $R = -\mathrm{d}z/\mathrm{d}\theta$ in the above convention and Equation 5 is re-written

$$\left|\frac{\partial n}{\partial t}\right|_D = -\frac{DN_0\Omega^2\gamma}{kT}\frac{\partial^3\theta}{\partial z^3} \tag{6}$$

where $|\partial n/\partial t|_D$ represents the rate of growth of a surface element away from its centre of curvature. Thus, since for a convex bulge $\partial^3\theta/\partial z^3$ is everywhere positive, Equation 6 indicates that a surface element of a convex bulge erodes towards its centre of curvature at a rate $DN_0\Omega^2\gamma/kT\ \partial^3\theta/\partial z^3$. For a concave trough the behaviour is reversed and there is a net accretion of material. Thus in the presence of surface diffusion alone an initially contoured infinite surface would relax by erosion of bulges and filling in of troughs towards a flat surface as shown by Mullins [17].

In the presence of sputtering, however, it has already been shown [1, 7] that the rate of erosion, for both convex bulges and concave troughs, of a surface element along the normal direction is given by

$$\left|\frac{\partial n}{\partial t}\right|_S = -\frac{\phi}{N} S \cos\theta \qquad (7)$$

where $\phi$ is the sputtering ion beam flux and $N$ the solid atomic density. Thus, if sputtering and surface diffusion are uncorrelated processes the total rate of normal erosion may be written as the sum of the contributions of Equations 6 and 7.

$$\left|\frac{\partial n}{\partial t}\right|_T = -\left\{\frac{\phi}{N} S \cos\theta + \frac{DN_0\Omega^2\gamma}{kT}\frac{\partial^3\theta}{\partial z^3}\right\}. \qquad (8)$$

If, however, the sputtering ion beam influences the surface diffusion process by, for example, production of excess defects, then the contributions will be correlated. A simple first order correlation will be discussed later.

Since $S$ is the sputtering coefficient due to ion beam erosion, then one can define, from Equation 8 an effective sputtering coefficient, $S_e$, which accommodates the effects of both ion erosion and surface diffusion. Thus we define

$$\frac{\phi}{N} S_e \cos\theta = \frac{\phi}{N} S \cos\theta + \frac{DN_0\Omega^2\gamma}{kT}\frac{\partial^3\theta}{\partial z^3}$$

or

$$S_e = S + \frac{N}{\phi}\frac{DN_0\Omega^2\gamma}{kT} \sec\theta \frac{\partial^3\theta}{\partial z^3}. \qquad (9)$$

With this definition it is now a straightforward matter to determine the velocities and directions of motion of points on a surface as derived in [4] and [6].

Thus, if one considers the behaviour of surface points which maintain a constant orientation relative to the ion beam direction and study the so called constant orientation trajectories, then the components of velocity of motion of a point of constant orientation $\theta$ are given by

$$\left.\frac{\partial x}{\partial t}\right|_\theta = \frac{\phi}{N}\frac{dS_e}{d\theta}\cos^2\theta$$

parallel to the $0x$ direction

and

$$\left.\frac{\partial y}{\partial t}\right|_\theta = \frac{\phi}{N}\left\{\frac{dS_e}{d\theta}\sin\theta\cos\theta - S_e\right\}$$

parallel to the $0y$ direction. (10)

The direction of motion of such a point is thus along an orientation $\phi$ with respect to the ion beam given by

$$\tan\phi = \frac{\dfrac{dS_e}{d\theta}\sin\theta\cos\theta - S_e}{\dfrac{dS_e}{d\theta}\cdot\cos^2\theta}\ . \qquad (11)$$

Substitution of Equation 9 into Equations 10 and 11 then leads to expressions for the velocity and direction of motion of a point of constant orientation in terms of $S$, $\theta$ and derivatives of $\theta$ with respect to $z$.

The rate of change of radius of curvature of a surface point may also be readily determined from the studies of Ducommun *et al.* [7] or by the following simple argument.

Since

$$R = \left|\frac{dz}{d\theta}\right|, \text{ then } \left.\frac{\partial R}{\partial t}\right|_\theta = \left.\frac{\partial}{\partial t}\left(\frac{\partial z}{\partial\theta}\right)\right|_\theta$$

Thus

$$\left.\frac{\partial R}{\partial t}\right|_\theta = \frac{\partial}{\partial\theta}\left(\frac{\partial x}{\partial t}\right)_\theta \left|\frac{\partial z}{\partial x}\right|_\theta = \frac{\partial}{\partial\theta}\left(\frac{\partial y}{\partial t}\right)_\theta \left|\frac{\partial z}{\partial y}\right|_\theta$$

and

$$\left.\frac{\partial R}{\partial t}\right|_\theta = \frac{\phi}{N}\frac{1}{\cos\theta}\left\{\frac{d}{d\theta}\left(\frac{dS_e}{d\theta}\cos^2\theta\right)\right\}$$
$$= \frac{\phi}{N}\frac{1}{\sin\theta}\left\{\frac{d}{d\theta}\left(\frac{dS_e}{d\theta}\sin\theta\cos\theta - S_e\right)\right\}$$

and finally

$$\left.\frac{\partial R}{\partial t}\right|_\theta = \frac{\phi}{n}\left\{\frac{d^2S_e}{d\theta^2}\cos\theta - 2\frac{dS_e}{d\theta}\sin\theta\right\} \qquad (12)$$

In the case of sputtering only, Equation 12 is important, since it reveals, as shown by Ducommun *et al.* [7], where discontinuities, or edges, first develop in an initially continuous curve. In this case $\partial R/\partial t|_\theta$ can take positive or negative values, depending only upon the form of the $S/\theta$ curve, and an edge first forms in finite time when the radius of curvature of a surface segment is reduced to zero. Edge formation also depends upon the initial surface contour and thus for any arbitrary initial contour and arbitrary $S/\theta$ function it is not possible to predict theoretically the erosion controlled development of the surface. After edges form it is equally difficult to predict the continuing development except when it can be recognized that edges will intersect and combine to form new edges. Successful predictions of contour development and end forms are thus carried out using geometrical construction from the Barber *et al.* [6] erosion slowness curve or computer simulations. In the present case, where co-ordinate and radial motion, as defined by Equations 10–12, are contour dependent, not only through the $S-\theta$ variation (which is time independent), but also through the time dependent local contour variations defined by the derivatives of $\theta$ with $z$, analytical prediction of contour development is even more difficult.

Thus, for example, in the Barber *et al.* [6] treatment where a unique erosion slowness curve can be defined by the angular variation of $1/(S\cos\theta)$, independent of the instantaneous contour geometry, for the case of sputtering alone, when one attempts to define an erosion slowness curve when surface diffusion is also operative, it is immediately noted that this is *not* instantaneous contour geometry independent. This is readily perceived since

$$\frac{1}{S_e\cos\theta}=\frac{1}{S\cos\theta+K\gamma\dfrac{\partial^3\theta}{\partial z^3}}$$

where $$K=\frac{DN_0\Omega^2}{kT}\frac{N}{\phi}.$$

Consequently, when surface diffusion is also operative a unique erosion slowness curve cannot be defined, but a family of such curves exists according to the allowed values of $\partial^3\theta/\partial z^3$. For surface convex bulges this parameter varies from $0\rightarrow\infty$ and so the appropriate family of erosion slowness curve is contained between a point at the origin ($\partial^3\theta/\partial z^3=\infty$) and the "sputtering only" erosion slowness curve ($\partial^3\theta/\partial z^3=0$). For surface concave troughs the diffusion parameter varies from 0 through $-S\cos\theta/K\gamma$ to $-\infty$ and the family of erosion slowness curves is contained between the "sputtering only" curve in the negative half plane (i.e. $1/(S\cos\theta)$ measured negatively), $-\infty$ (when $\partial^3\theta/\partial z^3=-S\cos\theta/K\gamma$) and in all of the positive half plane between $1/(S\cos\theta)=\infty$ and the origin. These variations of the "effective" erosion slowness are shown in Fig. 1.

The physical meaning of this behaviour of the erosion slowness curves is that convex bulges are always eroded by sputtering and surface diffusion, whereas surface troughs may either erode or accrete material, depending upon the relative influences of sputtering and surface diffusion.

The difficulty in analytic prediction of contour development now becomes apparent, since for every point on a surface contour, there will be different erosion slowness curves, depending upon the local value of $\partial^3\theta/\partial z^3$, and thus the appropriate erosion slowness curves to use will be both co-ordinate and time dependent. It, therefore, appears, at the present, that the most suitable method of following contour development when surface diffusion is operative will be by computer simulation, using Equations 10–12, with the value of $S_e$ determined from Equation 9 and using $S\phi kT/NN_0D\Omega^2\gamma$ as a variable parameter, for a variety of initial contours. Such a programme of study has been initiated by the author and results will be presented at a later date.

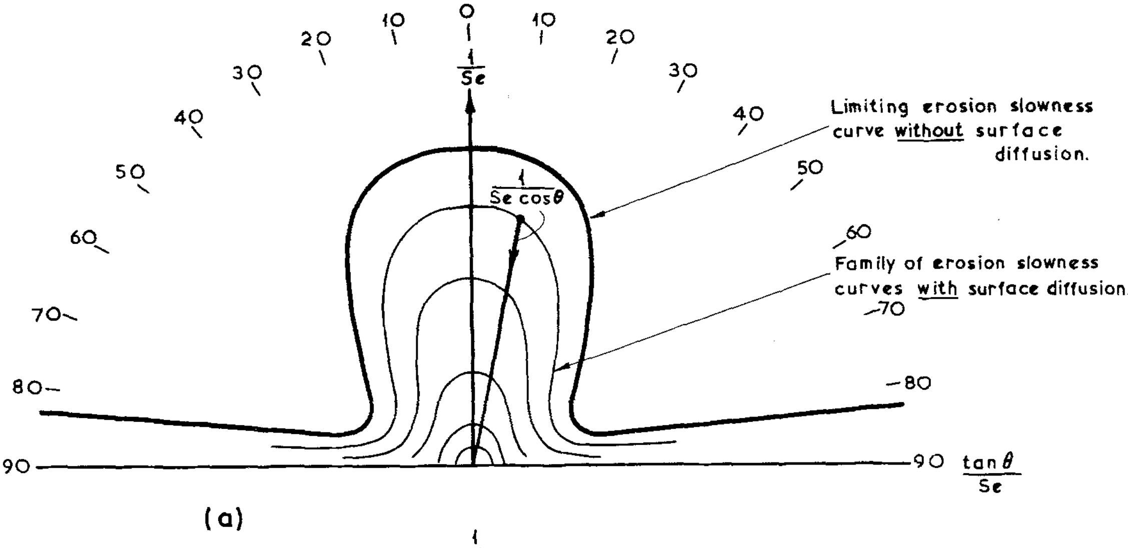
Despite the difficulty of predicting detailed contour development and the progress towards end forms ($t\rightarrow\infty$) at this stage, some interesting observations can be made.

If Equation 13 is re-written in full, using Equation 9, one obtains

$$\left.\frac{\partial R}{\partial t}\right|_\theta=\frac{\phi}{N}\left\{\frac{d^2S}{d\theta^2}\cos\theta-2\frac{dS}{d\theta}\sin\theta\right\}+\frac{\phi K\gamma}{N}\left(\frac{\partial\theta}{\partial z}\right)^{-3}\left\{\frac{\partial^5\theta}{\partial z^5}\cdot\frac{\partial\theta}{\partial z}-\frac{\partial^4\theta}{\partial z^2}\cdot\frac{\partial^2\theta}{\partial z^2}+\frac{\partial^3\theta}{\partial z^3}\cdot\left(\frac{\partial\theta}{\partial z}\right)^3\right\}\quad(13)$$

where the first term on the right hand side of Equation 13 describes the effects of sputtering and the second describes the influence of surface diffusion.

The first term is sign convention independent (i.e. whether one assumes positive or negative radius of curvature for a surface bulge or trough), but the second term is sign convention dependent. The first term has been investigated by Ducommun *et al.* [7] and it is shown that for the experimentally observed form of the $S-\theta$ relation, this term can possess positive and negative values, depending on the value of $\theta$. Thus radii of curvature in both bulges and troughs can increase or decrease with time and indeed the local radius of curvature can



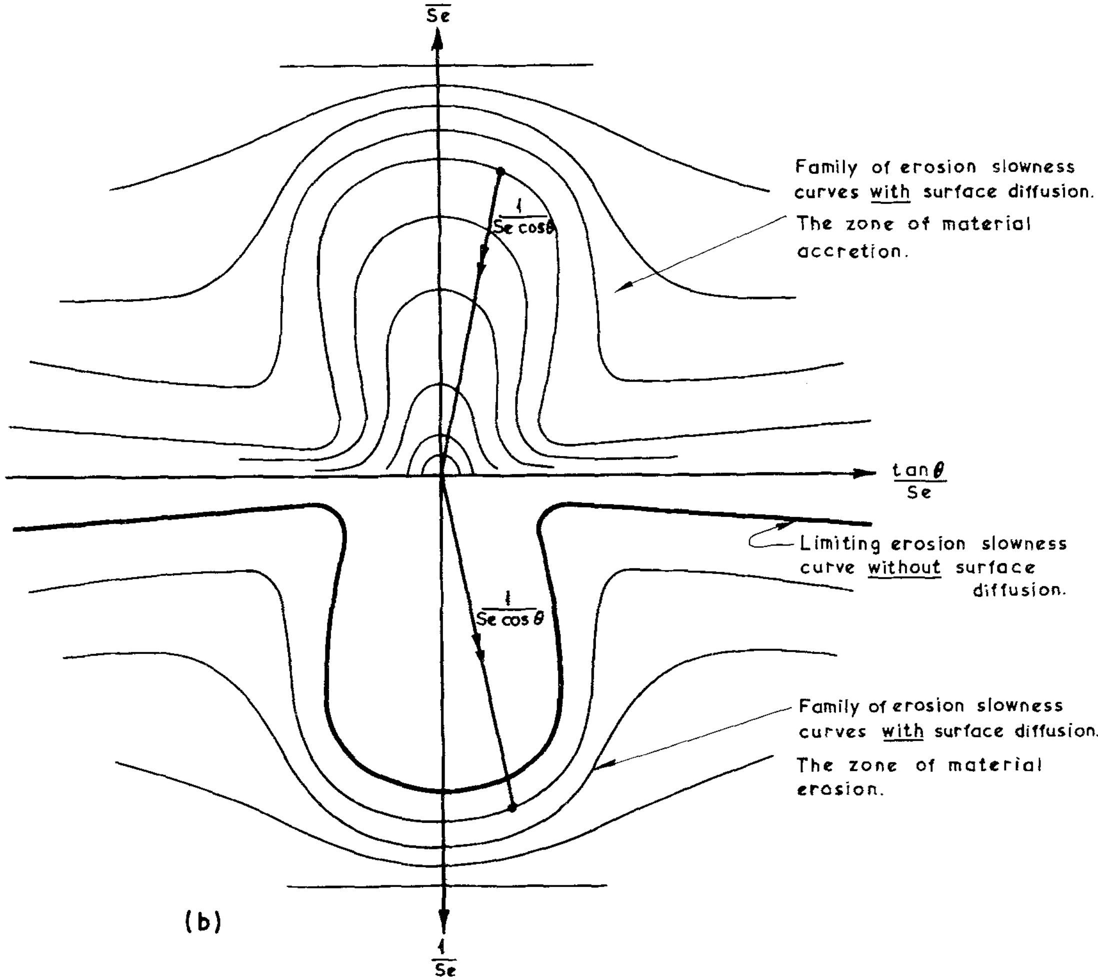




*Figure 1* Schematic representation of the behaviour of the "effective erosion slowness' ($1/S_e \cos\theta$) as a function of direction of ion incidence $\theta$ to a surface element, for various values of $\partial^3\theta/\partial z^3$ for (a) convex surface bulge, (b) concave surface trough.

reach zero in finite time. This condition indicates edge formation.

Considering the second term in Equation 13 alone, if one adopts the sign convention of positive radius of curvature for a surface bulge, then apparently the local radius of curvature could increase or decrease with time, depending upon the relative magnitudes of the first through to the fifth derivatives of $\theta$ with respect to $z$. In the case of time decreasing radius of curvature, however, then as the radius approaches zero, the final term in $\partial^3\theta/\partial z^3$ is dominant and with the convention of positive radius for a convex bulge, $\partial^3\theta/\partial z^3 \to +\infty$, which indicates that although the first sputtering term may be tending to decrease $R$, the second surface diffusion term becomes dominant and increases $R$. The inverse argument applies to the case of a concave surface trough. Thus $R = 0$ cannot occur. The implication of this result is that edges cannot form, even with the smallest contribution from surface diffusion. This finding is completely compatible with the situation of surface diffusion alone, where it is found [21], for example, in field emitter tips, the blunting occurs due to surface diffusion. Indeed the anology of the present study with the behaviour of field emitter tips is strong since in the present case, sputtering is the cause of edge formation, whilst in the field emitter case an applied electrostatic field exerts a similar influence, with both opposed by the effects of surface diffusion.

Equilibrium, or end form conditions, can be determined by demanding that $\partial x/\partial t|_\theta$ and $\partial y/\partial t|_\theta$ are simultaneously independent of $\theta$ in Equation 10 or equivalently that $\partial R/\partial t|_\theta$ in Equation 13 is zero. A general contour, satisfying Equation 13 = 0, can be determined, but it is also interesting to note that the condition $R = \infty$, $\partial\theta/\partial z = 0$ also satisfies this condition. Thus, a special end form is certainly a straight line (or plane in three dimensions). This is the form expected from considerations of sputtering alone [4–6] or self diffusion alone [18] on an infinite surface and is, therefore, not surprising. Without detailed consideration of the dynamic of contour development, it is not possible, analytically, to determine whether the smoothing is faster when both contouring processes operate simultaneously rather than in isolation, but heuristically one might expect such to be the case. Indeed, the recent experiments of Vasiliu *et al.* [16] suggest that as temperature is increased, sputtering induced topography growth becomes less apparent and bombarded surfaces adopt a smoother habit. If, as proposed by Vasiliu *et al.*, surface diffusion is a major cause of this effect, then this would be expected to increase with temperature, since the surface diffusion coefficient increase rapidly with temperature $T$ via a relation of the form $D = D_0 \exp(-E/kT)$, where $E$ is the activation energy for the process.

Indeed, if in Equation 9 the second right hand term is identified as an "effective sputtering coefficient" due to surface diffusion, this is seen to increase with both $D$ and $\gamma$. Thus, since progress towards equilibrium is undoubtedly faster the larger the sputtering coefficient (e.g. all erosion velocities scale with $S$), then it is expected that increased values of $D$ and the surface tension $\gamma$ would lead to more rapid attainment of equilibrium – both physically realistic conclusions.

At this point, comment will be made upon the influence of bombardment enhanced surface diffusion. Vasiliu *et al.* [16] observed that, at elevated substrate temperatures, surface features were minimized within the bombarded area of an Fe sample, but that features, atypical of thermal facetting, were observed outside the irradiated area. Hermanne has also reported (private communication) that surface feature growth during ion bombardment of Cu can be inhibited at 350 to 400° C, whereas features produced by room temperature irradiation can only be annealed at temperatures $\geqslant 400°$ C. This evidence suggests that bombardment may enhance surface diffusion, just as it is known to enhance volume diffusion in a number of circumstances [22]. The process of radiation damage enhanced diffusion is usually associated with the presence of a vacancy density in excess of the thermal equilibrium density. In the case of our hypothetical amorphous solid, such a process is less meaningful, but since it is believed that the present results can be taken over into the crystalline solid case (with some care since other topography induction effects also exist), the same assumption of the cause of diffusion enhancement will be made.

When an ion slows down to rest in a solid it creates a displaced atom cascade [23], forming a depth distribution of point and extended defects. Some of these defects will rapidly anneal, due to both thermal and athermal processes, but a fraction will survive, potentially assisting the diffusion process. If the surface defect density is not too large (i.e. not approaching complete disorder, where this is a curious concept in a random amorphous solid), then it is reasonable to assume that each bombarding ion will produce a certain number $n$/unit area surviving defects, where $n$ will be a function of ion type and energy, target material, orientation and temperature. At high levels of $n$ it would be more appropriate to write $n$ as the solution of $(dn/dt) = \phi A(1 - an)$, where $A$ is a constant describing the number of surviving defects produced per ion and $a$ is the area of each surface defect.

At the lower levels of $n$, we will assume, as a first order approximation, that the competing processes of defect production and annealing reach rapid equilibrium, so that the excess defect density is a linear function of the bombarding ion flux. This assumption also embraces the concept that surface atomic diffusion does not change this excess defect concentration (i.e. a dilute solution approximation). In these circumstances, we can write $n = B\phi$, where $B$ is a constant. In the dilute solution approximation we can write [24] the diffusion coefficient $D_E$ in the presence of excess defects as

$$D_E = D + nD_V \tag{14}$$

where $D_V$ is the diffusion coefficient of defects (presumably vacancies) at the substrate temperature during irradiation. For a surface element inclined at angle $\theta$ to the 0$x$ direction, the ion flux is $\phi \cos\theta$, thus, for this element

$$n = B\phi\cos\theta. \tag{15}$$

Thus, in the presence of the ion flux

$$D_E = D + B\phi\cos\theta\, D_V, \tag{16}$$

and substituting this expresion into Equation 9 yields an "effective sputtering coefficient" in the presence of thermal and radiation enhanced surface diffusion as

$$S_e = S + \frac{N}{\phi}\frac{N_0\Omega^2\gamma}{kT}\sec\theta\,\frac{\partial^3\theta}{\partial z^3}(D + B\phi\cos\theta\, D_V) \tag{17}$$

In this case, therefore, there are three terms describing the effective sputtering, two of which are beam flux dependent and one which is not. The extra term in Equation 17 when compared with Equation 9 is the correlation term of bombardment and diffusion. Detailed prediction of growth contours clearly requires computer simulation in this case. If the beam flux is relatively large, the effect of thermal surface diffusion could be much smaller than that due to enhanced diffusion, and this difference will become increasingly more important with increasing temperature since $D_V/D = \exp(E - E_V/kT)$. Thus, for high beam fluxes and temperatures where $(E - E_V/kT)$ is reduced, bombardment enhanced diffusion will dominate and be (in a first approximation) flux independent. Thus, one can perceive three temperature stages for topographic development. The first at low temperatures where both thermal and radiation enhanced surface diffusion are absent and sputtering will dominate with flux independence. At intermediate temperatures where defect mobility is relatively low, but diffusion may be dramatically enhanced due to a large ion bombardment induced defect density, both sputtering and surface diffusion will modify the topography and be beam flux independent at high beam flux, but flux dependent at low flux. Finally, at high temperatures, thermal surface diffusion will dominate and topography development will be beam flux independent.

Although this treatment is very superficial, it does indicate the complexity of events, even for the assumed amorphous solid. In the case of a crystalline solid, treatment is even more difficult since not only will the influence of subsurface extended defects become important, but also the anisotropy of $\gamma$ as a function of crystalline orientation and $S$ as a more complex function of orientation will perturb the situation discussed here. Thus one might expect, in the case of crystalline solids, at low temperatures where sputtering is dominant, the development of non-equilibrium surface facets of high sputtering yield, and at higher temperatures the development of thermal facets corresponding to $\gamma - \theta$ discontinuities. At intermediate temperatures, in the absence of experimental data, it is hazardous to guess the effects.

## 3. Conclusions

A simplified analytic treatment of the development of surface topography of a solid influenced by ion bombardment induced erosion, together with surface diffusion, both thermal and radiation enhanced, results in intractable mathematical descriptions of the temporal topographic growth. Computer simulation is required for detailed prediction, but some general conclusions as to the influence of diffusion can be drawn. Thus, the net effect of diffusion is to prohibit angular discontinuity formation and a more rapid attainment of equilibrium end forms. Diffusion assists the erosion of bulges on surfaces, but can impede the erosion of troughs.

## Acknowledgement
The author is indebted to M. J. Nobes for many fruitful discussions, particularly concerning edge development and the derivation and consequences of Equation 12.

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Received 27 October and accepted 17 November 1975.