Dynamic scaling of ion-sputtered rotating surfaces

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Surfaces eroded by off-normal incidence ion bombardment often have a rippled topography, and this is undesirable in a number of applications. Sample rotation during sputtering inhibits or prevents surface roughening and is widely used in precision depth profiling. In this paper, I study the surface roughening of a sample that is simultaneously rotated and sputtered in the limit in which viscous flow can be neglected. I find that the structure of the surface depends on the sign of a parameter characterizing the curvature dependence of the sputter yield, $\mu_{\rm av}$. For $\mu_{\rm av}>0$, the asymptotic scaling behavior of the surface is in the same universality class as the Kardar-Parisi-Zhang equation in $2+1$ dimensions. At long times the surface is composed of a patchwork of paraboloids of revolution whose mean lateral dimension grows algebraically in time, a topography quite reminiscent of those observed experimentally. In contrast, the interface has a chaotic cellular structure described by the isotropic Kuramoto-Sivashinsky equation if $\mu_{\rm av}<0$. The mean cell width is constant in time in this case. [S1063-651X(96)08812-5]

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Off-normal incidence ion bombardment often produces periodic height modulations on the surface of an amorphous solid [1–4]. For incidence angles θ less than a critical angle θ_c from the normal, the wave vector of the modulations is parallel to the component of the ion beam in the surface plane. The wave vector is perpendicular to this component for incidence angles close to grazing. It is now well established that if the incident ions do not react chemically with the solid, these surface modulations form as a result of the curvature dependence of the sputter yield [1–3,5–7].

In practice, the ripples are not completely coherent because of the effects of shot noise. The topography is therefore disordered at long wavelengths. Moreover, as the amplitude of the ripples becomes appreciable, nonlinearities become increasingly important. Recently, considerable effort has been devoted to understanding the scaling behavior of the surface, which is influenced by both the noise and the nonlinearities [8–10]. The dynamics of the interface are governed by the noisy, anisotropic Kuramoto-Sivashinsky equation, which displays several different types of complex scaling behavior whose precise nature remains to be elucidated.

Formation of surface ripples is problematic in a variety of applications, including secondary ion mass spectroscopy (SIMS), Auger electron spectroscopy (AES), and ion milling. SIMS is one of the most widely used techniques for dopant profiling of semiconductors, while AES is an important tool in the structural characterization of multilayers. In a typical SIMS or AES apparatus, the primary ions are incident at an angle $\theta\neq 0$. Thus, as sputtering proceeds, ripples can be formed, and this leads to rapid degradation of the depth resolution. This is particularly problematic when SIMS or AES is used in conjunction with ion sputtering for depth profiling of modern thin film materials and devices [11].

Zalar first demonstrated that this problem can be overcome by rotating the sample about its surface normal as the depth profiling proceeds [12]. Zalar rotation has subsequently been used by many other groups, who found that in many cases, the surface remains remarkably smooth as the solid is eroded [13]. Moreover, Cirlin and co-workers have observed that if the sample is at first stationary and ripples are formed, subsequent rotation during erosion can lead to the production of a smooth surface [14]. Sample rotation does not always suppress surface roughening, however. In some instances, the sample roughens while it is simultaneously eroded and rotated, albeit at a slower rate than it does when it is eroded without being rotated [14,15].

Although Zalar rotation is widely used in sample analysis and is even applied in commercially available depth profiling systems, an understanding of its effectiveness has been lacking. Recently, Bradley and Cirlin advanced a theory that explains why sample rotation reduces or eliminates surface roughening during depth profiling [16]. When the sample is rotated, the smoothing effects of viscous flow and surface self-diffusion can prevail over the roughening effect of the curvature-dependent sputter yield and generate a smooth surface. The theory also accounts for the observations of Cirlin *et al.* [14] mentioned above.

The object of the present paper is to elucidate the nature of the surface roughening that sometimes occurs even when the sample is rotated. According to Bradley and Cirlin's theory, the surface roughens during concurrent ion sputtering and sample rotation if the viscosity of the sample is too high. We will study the surface roughening in this high-viscosity regime. For simplicity, the viscosity will be taken to be large enough that the effects of viscous flow can be neglected altogether. I find that the structure of the surface depends on the sign of a parameter characterizing the curvature dependence of the sputter yield, $\mu_{\rm av}$. If $\mu_{\rm av}<0$, the interface has a chaotic cellular structure which is described by the much-studied *isotropic* Kuramoto-Sivashinsky (KS) equation [17,18]. We can therefore simply refer to the existing literature to learn a great deal about the nature of the surface roughening in this case—for example, the mean cell width is constant in time. When $\mu_{\rm av}$ is positive, on the other hand, the long-time, long-wavelength scaling behavior of the surface is the same as that of the Kardar-Parisi-Zhang (KPZ) equation in $2+1$ dimensions [19,20]. Once we have arrived at this

conclusion, the vast literature on the KPZ equation allows us to describe the scaling properties of the interface for $\mu_{\rm av}>0$ quite satisfactorily. At long times, the surface is composed of a patchwork of paraboloids of revolution whose mean lateral dimensions grow as $t^{1/z}$ when t is large. The dynamic critical exponent z has an asymptotic value of roughly 1.7. However, if the surface is very rough initially, there is a long-lived transient regime in which $z=2$. This prediction is in qualitative agreement with the experimental results of Barna, Barna, and Zalar [21].

Let us begin by considering a flat surface of an amorphous solid that is subjected to off-normal incidence ion bombardment. We choose stationary coordinate axes (x,y,z) with the unit vector $\hat{z}$ normal to the surface. Let the unit vector along the ion beam direction of incidence be $-\hat{e}$, and let the polar and azimuthal angles of $\hat{e}$ be θ and ϕ, respectively. The angle of incidence θ is nonzero and, to begin, we set $\phi=0$. Finally, let $h(x,y,t)$ denote the height of the interface above the point (x,y) in the x-y plane at time t. Naturally, $h(x,y,t)=h_0-v_0t$, where h_0 is the initial height of the interface and $v_0=v_0(\theta)$ is the speed of the surface while the solid is being eroded.

A real surface is not perfectly flat initially, and this has a profound effect on the time evolution of the surface. Let $u(x,y,t)\equiv h(x,y,t)-h_0+v_0t$ be the deviation of the surface height away from the perfectly flat surface. We assume that the effects of viscous flow are negligibly small and for the moment omit the effect of shot noise. The equation of motion for u is then

$$u_t=v_0'u_x+\mu_1u_{xx}+\mu_2u_{yy}-B\nabla^2\nabla^2u+\frac{\lambda_1}{2}\,u_x^2+\frac{\lambda_2}{2}\,u_y^2, \tag{1}$$

where $u_t\equiv\partial u/\partial t$, $u_x\equiv\partial u/\partial x$, and so forth [1,8]. Here $\nabla^2=\partial^2/\partial x^2+\partial^2/\partial y^2$, $v_0'\equiv(dv_0/d\theta)|_{\theta=\theta_0}$, μ_1, μ_2, λ_1 and λ_2 are constants, and B is the surface self-diffusivity. In Eq. (1), the first term on the right-hand side accounts for the angular dependence of the sputter yield, the second and third terms arise because the sputter yield depends on the surface's curvature [1], and the fourth term accounts for the effect of surface self-diffusion [22]. The final two terms make Eq. (1) nonlinear and have an important effect once the surface width has become sufficiently large. Equation (1) is the anisotropic Kuramoto-Sivashinsky equation [9,23].

If the azimuthal angle ϕ is nonzero, we consider a rotated coordinate system (X,Y,Z) in which $\hat{e}$ lies in the X-Z plane. Specifically,

$$X=x\,\cos\phi+y\,\sin\phi, \tag{2a}$$

$$Y=-x\,\sin\phi+y\,\cos\phi, \tag{2b}$$

$$Z=z. \tag{2c}$$

Equation (1) then continues to hold, but with x, y, and z replaced by X, Y, and Z. Transforming this equation to the original frame of reference (x,y,z), we obtain

$$\begin{aligned}u_t=&\mu_{\rm av}\nabla^2u-B\nabla^2\nabla^2u+\tfrac{1}{2}\lambda_{\rm av}(\nabla u)^2\\&+v_0'(u_x\,\cos\,\phi+u_y\,\sin\,\phi)+\tfrac{1}{2}\Delta\mu\,\cos(2\phi)(u_{xx}-u_{yy})\\&+\Delta\mu\,\sin(2\phi)u_{xy}+\tfrac{1}{4}\Delta\lambda\,\cos(2\phi)(u_x^2-u_y^2)\\&+\tfrac{1}{2}\Delta\lambda\,\sin(2\phi)u_xu_y,\end{aligned} \tag{3}$$

where $\mu_{\rm av}\equiv(\mu_1+\mu_2)$, $\lambda_{\rm av}\equiv\frac{1}{2}(\lambda_1+\lambda_2)$, $\Delta\mu\equiv\mu_1-\mu_2$, and $\Delta\lambda\equiv\lambda_1-\lambda_2$.

We now turn to the effect of rotating the sample about the z axis with a constant angular velocity ω during ion bombardment. This is equivalent to rotating the ion beam and holding the sample fixed. We shall adopt the latter viewpoint because it turns out to be simpler. When ω is small, ripples begin to form after a time. The orientation of these ripples depends on the azimuthal angle ϕ. As the beam rotates, the ripple wave vector rotates along with it. This is the only effect of rotating the beam when ω is small.

Now consider the opposite limit in which the ion beam is rotated rapidly. When ω is large, it is as if the solid were simultaneously bombarded from all azimuthal angles ϕ. Thus we can obtain an approximate equation of motion by averaging Eq. (3) over the range $0\leqslant\phi\leqslant2\pi$. This gives

$$u_t=\mu_{\rm av}\nabla^2u-B\nabla^2\nabla^2u+\tfrac{1}{2}\lambda_{\rm av}(\nabla u)^2. \tag{4}$$

When is ω large, and when is it small? When the beam is not rotated, the time needed for the ripple amplitude to become appreciable is proportional to $B/\mu_{\rm av}^2$ [1]. Thus, when $\omega\ll\mu_{\rm av}^2/B$, the orientation of the ripples will slowly rotate. Conversely, Eq. (4) applies when $\omega\gg\mu_{\rm av}^2/B$. Ripple topographies are never observed at the rotation rates used in SIMS or AES and so we will assume that $\omega\gg\mu_{\rm av}^2/B$ for the remainder of the paper.

The time evolution of the interface is qualitatively different for positive and negative $\mu_{\rm av}$. Consider first the case in which $\mu_{\rm av}$ is negative. Equation (4) is then the isotropic Kuramoto-Sivashinsky equation [17,18], which we will refer to as simply the KS equation. The KS equation has previously been used to model diffusive instabilities of chemical waves [17] and wrinkled flame fronts [18]. The linearized KS equation has a band of unstable modes with wave vector smaller than a threshold value. The most remarkable feature of Eq. (4) is that the nonlinear term prevents the runaway growth of the unstable modes, and, as a result, the surface gradient remains bounded. The interplay between the linear instability and the nonlinear coupling between the unstable modes leads to a state of spatiotemporal chaos [24].

When fully developed, the surface topography has a cellular structure. If $\lambda_{\rm av}$ is positive, each cell is a rounded protrusion on the surface, while the cells are basins if $\lambda_{\rm av}<0$ (see, for example, Ref. [25]). Note that the cellular structure of the surface is neither periodic in space nor in time—the cells appear and disappear chaotically as time passes.

We can readily determine the typical dimensions of these cellular structures by introducing the dimensionless variables

$$\tilde{x}=\left(\frac{\mu_{\rm av}}{B}\right)^{1/2}x,$$

$$\tilde{y}=\left(\frac{\mu_{\rm av}}{B}\right)^{1/2} y,$$

$$\tilde{u}=\left(\frac{\lambda_{\rm av}}{\mu_{\rm av}}\right) u,$$

$$\tilde{t}=\left(\frac{\mu_{\rm av}^2}{B}\right) t,$$

Equation (4) now becomes

$$\tilde{u}_{\tilde{t}}=\tilde{\nabla}^2\tilde{u}-\tilde{\nabla}^2\tilde{\nabla}^2\tilde{u}+\tfrac{1}{2}(\tilde{\nabla}\tilde{u})^2. \tag{5}$$

We conclude that the mean width of the cells $\langle w\rangle$ is proportional to $(B/\mu_{\rm av})^{1/2}$, and that their average height scales as $\mu_{\rm av}/\lambda_{\rm av}$.

Both $\mu_{\rm av}$ and $\lambda_{\rm av}$ are proportional to the ion flux f and are independent of the sample temperature T. Therefore, the mean cell height does not depend on f or T. The variation of the mean cell width $\langle w\rangle$ with temperature and flux is more complex. $\langle w\rangle$ is proportional to the ripple wavelength λ observed in the absence of sample rotation [1,26]. Thus, when the ion flux f is relatively low and the temperature T is relatively high, the mean cell width varies as $\langle w\rangle \sim (fT)^{-1/2}\exp(-\Delta E/2k_BT)$, where ΔE is the activation energy for surface self-diffusion and k_B is Boltzmann's constant [1]. In the opposite limit of high fluxes and low temperatures, radiation-induced surface self-diffusion becomes important [27]. The surface self-diffusivity B includes the effects of both thermally activated and ion-induced surface self-diffusion. B grows linearly with f, since the frequency of atomic jumps induced by the impinging ions is proportional to f. Thus, $B_{\rm eff}=B_0+af$, where B_0 is the thermally activated surface self-diffusivity and a is a positive constant. Since $\mu_{\rm av}$ is proportional to f, when the flux is high and the temperature is low, both $\langle w\rangle$ and λ are independent of f. Also, note that since the energy of the ions is typically much larger than k_BT, the temperature dependence of the coefficient a should be negligible. Thus, in the high-flux, low-temperature regime, $\langle w\rangle$ and λ should be nearly independent of temperature as well.

Partial corroboration for this picture comes from experimental results on the wavelength λ in the absence of sample rotation. In their experiments on the off-normal incidence erosion of GaAs, MacLaren *et al.* found that if the temperature is reduced while the ion flux is held fixed, λ crosses over from its high-temperature Arrhenius behavior to a roughly constant value at low temperatures [3].

We now turn our attention from the structure of the surface at short length scales to its long-wavelength scaling properties. It is now well established that in $1+1$ dimensions the scaling behavior of the KS equation is the same as that of the KPZ equation [28–31]. There is evidence that KS and KPZ equations are in the same universality class in $2+1$ dimensions as well [25], but there is not yet a consensus on this issue [32]. In reality, Eq. (4) is not quite complete as it stands—a shot noise term ought to appear on the right-hand side of this equation. Cuerno and Lauritsen have recently argued that in $2+1$ dimensions the noisy KS equation is in the same universality class as the KPZ equation, but they concede that their argument is not conclusive [33]. The scaling behavior of the surface when $\mu_{\rm av}<0$ is therefore still an open question.

Now let us consider the case in which $\mu_{\rm av}$ is positive. In this case, the surface is stable against small perturbations away from a perfectly flat state, and as a result, the effect of surface self-diffusion can be neglected. (In contrast, the surface is unstable when $\mu_{\rm av}<0$, and the effect of surface self-diffusion is important in determining the length scale of the short-range structure.) We therefore set B to zero in Eq. (4). We will also take the effect of shot noise into account by adding a noise term $\eta(x,y,t)$ to the right-hand side of Eq. (4). Explicitly,

$$u_t=\mu_{\rm av}\nabla^2 u+\tfrac{1}{2}\lambda_{\rm av}(\nabla u)^2+\eta, \tag{6}$$

where $\eta(x,y,t)$ has a Gaussian distribution with $\langle\eta(x,y,t)\rangle=0$ and

$$\langle\eta(x,y,t)\,\eta(x',y',t')\rangle=2D\,\delta(x-x')\,\delta(y-y')\,\delta(t-t').$$

Here D is a constant.

Equation (6) is the KPZ equation in $2+1$ dimensions [19,20]. It therefore appears that the scaling behavior of the sample surface is in the same universality class as the KPZ equation in $2+1$ dimensions. Thus the interface width w of a sample of linear dimension L follows the scaling form

$$w(L,t)=L^{\alpha}f\left(\frac{t}{L^z}\right), \tag{7}$$

where $f(x)\sim x^{\beta}$ for $x\ll 1$, $f(x)$ approaches a constant $x\to\infty$, and $z=\alpha/\beta$. The dynamic critical exponents α and z satisfy the scaling relation $\alpha+z=2$. The exact value of z is not known in $2+1$ dimensions, but numerical estimates typically yield values in the neighborhood of 1.7 [20].

Let us now suppose that the sample surface is very rough initially. If this is the case, then the effects of shot noise may be neglected for all but the longest times. Thus, we shall drop the noise term from the KPZ equation (6), yielding the deterministic KPZ equation. The scaling behavior of the surface is again governed by Eq. (7), although the values of α and z are different. Let us suppose that the initial width of the surface scales as L^{α_0}. Using scaling arguments, Krug and Spohn found that

$$z=\min(2,2-\alpha_0) \tag{8}$$

and that $\alpha=2-z$ [34]. These predictions agree well with numerical integrations of the deterministic KPZ equation in $1+1$ dimensions [35], but have not yet been tested in $2+1$ dimensions.

The short-wavelength structure of the surfaces generated by the deterministic KPZ equation have also been studied [19,34]. For sufficiently long times, the solution is composed of a patchwork of paraboloids of revolution of the form $h_n(\vec{r},t)=a_n-|\vec{r}-\vec{r}_n|^2/(2\lambda_{\rm av}t)$ joined together by discontinuities in ∇h. The lateral dimensions of these paraboloids grow as $t^{1/z}$.

It is interesting to compare these predictions with the experimental results of Barna, Barna, and Zalar [21], who subjected multilayer Ni-Cr films to off-normal incidence ion

bombardment during sample rotation. The surfaces of their samples were quite rough initially. After sputtering had proceeded for some time, they found that the surfaces of their samples were composed of a patchwork of bulges, and that the lateral dimensions of these bulges grew in the course of time. It therefore seems likely that to a good approximation the time evolution of the sample surface in their experiments was governed by the deterministic KPZ equation. The value of α_0 for a conventionally prepared sample surface is of course zero. Equation (8) then gives $z=2$, and so the theory predicts that the bulge width grows as $t^{1/2}$. As a result, the total number of cells on the surface N should decay as t^{-1}. Barna, Barna, and Zalar provide a sequence of photographs of the sample surface at several different times, and N is clearly decreasing with time. However, the resolution of these photographs do not permit a quantitative check of the prediction $N \sim t^{-1}$.

Topographies similar to those of Barna, Barna, and Zalar were first observed more than three decades ago in experiments on the sputtering of a rotating glass sample [36] and yet systematic experimental work has not yet been done. Hopefully, quantitative experiments will be done in the near future which will permit a detailed test of the theory. It would also be quite interesting to see whether cellular topographies of the Kuromoto-Sivashinsky type occur in some instances, as suggested by the theory. If so, a detailed experimental study of rotating, sputtered surfaces might resolve the controversy over the scaling behavior of the Kuramoto-Sivashinsky equation in $2+1$ dimensions.

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