Thin-Film Growth and the Shadow Instability

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We propose a growth model for deposition of thin amorphous films by the sputtering technique. For small values of the diffusion constant, the film develops a self-similar mountain landscape. As the diffusion constant is increased a regime is reached where growth of compact flat films is possible up to a critical height. Further deposition leads to surface roughening.

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Thin films are commonly grown by the deposition of atoms from a vapor onto a substrate. Ideally, this process will result in a smooth film. All too frequently, however, growth via vapor deposition leads to surfaces with a significant amount of roughness due to a growth instability in which the increase in surface area associated with a protuberance leads to a locally enhanced growth rate.[1]

The way in which various mechanisms, such as dynamics of atoms in the vapor, affect the development of a growth instability depends on the physical conditions that characterize the specific growth process. For both sputtering[2] and molecular-beam epitaxy, the incoming atoms have a long mean free path and can thus be assumed to move ballistically. Molecular-beam-epitaxy atoms are, in addition, collimated in the direction normal to the substrate; the rate at which a portion of the film surface grows depends entirely on the local environment.

By contrast, the vapor atoms that are deposited via sputtering are incoming from all directions. The local growth rate is roughly proportional to the exposure angle of a site. The shadowing of valleys by peaks enhances the instability leading to roughness. The growth rate at a point on the surface is strongly influenced by the topology of both nearby and not-so-nearby regions. As a competing mechanism, surface diffusion[3] erodes peaks and fills in the valleys. Diffusion in all cases tends to smooth out roughness in a growing surface.

A class of growth models has been proposed for growth under ballistic conditions, as exemplified by the popular Eden model,[4] where the specific conditions of the deposition process are assumed to be unimportant. In the case of the Eden model the growth rate depends only on the *local* curvature and normal to the surface. It has been found that this model yields a width $w(L)$ for the surface of a $d=1$ sample which diverges with L as L^{χ}. The exponent $\chi \cong \frac{1}{2}$. According to Kardar, Parisi, and Zhang,[4] this exponent is universal, i.e., independent of specific details. If, on the other hand, the growth rate is limited by diffusion, as in the case of the diffusion-limited aggregation (DLA) model, the rate is a *nonlocal* functional of the interface morphology just as for the shadowing effect. Sputtering deposition thus appears to share features of both the Eden and DLA models and it is reasonable to ask whether it lies in either universality class and whether the dynamics of the vapor is a crucial control parameter of surface morphology.

To answer this question and to gain further insight into the consequences of shadowing on growth processes, we have considered the simplest model that incorporates the basic physics of sputtering deposition. We assume a "solid-on-solid" model where the film height $h(x,t)$ grows according to the following rate equation:

$$\frac{\partial h}{\partial t} = -D\frac{\partial^4 h}{\partial x^4} + R\theta(x,\{h\}) + \eta(x,t)\,. \qquad (1)$$

The first term on the right-hand side is the divergence of the surface diffusion current[5] with D proportional to the surface diffusion constant. In Eq. (1) we assumed low vapor pressure. If reevaporation and subsequent adsorption is a significant annealing mechanism, then a "surface-tension"-type term $\gamma(\partial^2 h/\partial x^2)$ should be used instead. The second term is the deposition rate R with $\theta(x,\{h\})$ the exposure angle (Fig. 1) and the last term is the combined effect of shot noise and thermal noise of the substrate. We can also use Eq. (1) as a model for the dynamics of sputter cleaning of rough surfaces.[6] This would simply mean choosing a negative value of R. The erosion rate of a site of a rough surface is then as-

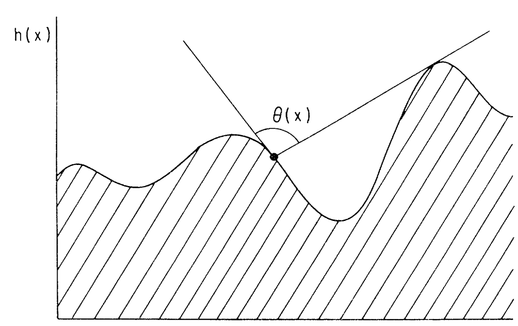

FIG. 1. The shadow effect. A site in a valley grows with a rate proportional to the exposure angle θ.

sumed to be proportional to the exposure angle. Since Eq. (1) does not include any effects of crystallinity (faceting), it can only apply to the growth of amorphous films. The nonlinearity in Eq. (1) lies in the functional dependence of θ on the height profile $h(x,t)$. Equation (1) ignores any local nonlinearity, not because those effects are insignificant, but because we are here interested in the asymptotic properties of a surface roughened by shadowing alone.

If η is a Gaussian white-noise source and if shadowing is neglected, we simply replace θ by its average $\langle\theta\rangle$, then the resulting surface has a mean height $R\langle\theta\rangle t$ and a width w proportional to[7] $(t^3/D)^{1/3}\langle\eta^2\rangle^{1/2}$. As our initial consideration of the shadowing effect, we perform a linear stability analysis around a flat surface. If, at $t=0$, the surface has a small periodic modulation $h(x,0)=h_k^0\cos kx$, then the exposure angle $\theta\cong\langle\theta\rangle+\alpha k h_k^0 \cos kx$ with $\alpha\cong 0.59$, neglecting higher harmonics. The perturbation grows exponentially $h_k(t)=h_k^0 e^{\omega_k t}$ with a rate constant

$$\omega_k=\alpha R k - D k^4 . \tag{2}$$

Note that for $k\rightarrow 0$, we can write ω_k as Uk with U proportional to the steady-state growth rate R. Short-distance details of the surface erode quickly ($\omega_k<0$), while the amplitude of a long-wavelength mode grows ($\omega_k>0$). The mode with the highest growth rate has a wave vector $k^*=(\alpha R/4D)^{1/3}$ and a growth rate $\omega^*=\frac{3}{4}\alpha R(\alpha R/4D)^{1/3}$.

What is the surface morphology if shadowing is the dominant effect? Divide the x axis into cells of width a (large compared to a lattice constant) and let $h(i)$ be the mean height of the cell centered at $x=ai$. The continuous surface is then represented by a set of N columns. Assume that the nearest neighbors of column i have a slightly greater height. This means that the exposure angle $\theta(i)$ is

$$\theta(i)=\pi-\arctan\left[\frac{h_{i+1}-h_i}{a}\right]-\arctan\left[\frac{h_{i-1}-h_i}{a}\right]. \tag{3}$$

At $t=0$ the surface is nearly flat and columns have heights with relative height differences of order a. From Eqs. (1) and (3), it follows that after a time τ of order a/R, $\theta(i)$ has reduced so much that our column stops growing. This happens to roughly $\frac{1}{2}$ of all columns. The remaining columns continue to grow. Again, consider one of these columns which is flanked by two slightly higher columns at a distance $2a$. The new exposure angle is

$$\theta(i)=\pi-\arctan\left[\frac{h_{i+1}-h_i}{2a}\right]-\arctan\left[\frac{h_{i-1}-h_i}{2a}\right]. \tag{4}$$

This column stops growing at a height of order $2a$ after a time 2τ. Iteration of this argument results at late time into $N/2$ columns of height a, $N/4$ columns of height $2a$, ... $N/2^p$ columns of height $2^{p-1}a$. We must stop when $2^p=N$ so the highest column is roughly $L/2$. If we define the coverage $c(h)$ as the number of atoms per unit area between h and $h+a$ then, by summing all columns whose height exceed h, one finds that

$$c(h)\propto\frac{1}{h^p}, \tag{5}$$

with $p=1$. The exponent p and χ are related by the scaling relation[8] $\chi=(2-p)/2$ so $\chi=\frac{1}{2}$. The resulting column distribution is self-similar and it is very similar to the Cantor bar.[9]

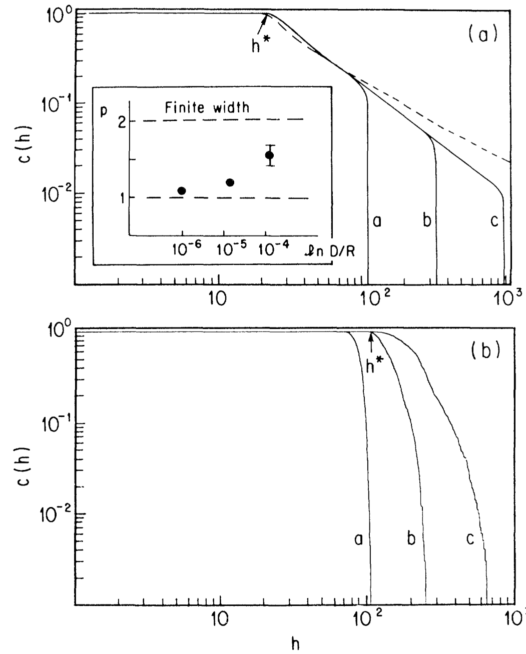

FIG. 2. (a) Log-log plot of coverage $c(h)$ vs h for $D=10^{-5}$ at $t=111,333,1000$ (curves a,b,c, respectively) time steps. The dashed curve shows the $D=0$ limit $c(h)\propto 1/h$. Inset: The dependence of the exponent p on D. (b) Same plot for $D=10^{-1}$. h^* is the critical height until which the growing surface is smooth.

The argument obviously is heuristic. To check it, and in order to study the effect of surface diffusion, we have solved the discretized version of Eq. (1) numerically. As a sidelight, we note that this growth lends itself readily to simulation studies, in contrast to the most widely studied models of nonlocal growth (e.g., DLA) that require the solution of nontrivial transport equations in the vapor. The noise term was not included and the surface at $t=0$ had a small amount of disorder. In Fig. 2(a) we show $c(h)$ for the case $D=0$, $t=1000$ and $D=10^{-5}$, $t=111,333,1000$ in units of $a=1$ with $R=1$, $N=1000$. At later times, $c(h)$ indeed assumes a power-law dependence. However, for $D\neq 0$, the exponent p is greater than 1. In the inset of Fig. 2(a) we show the dependence

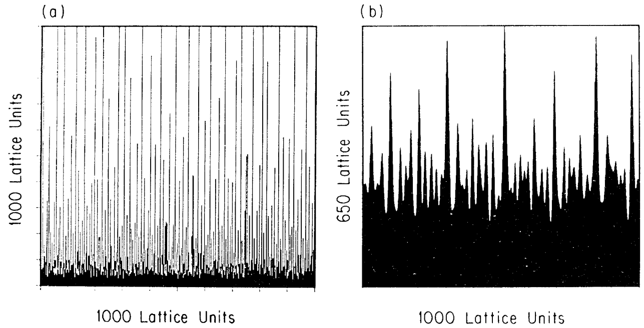

FIG. 3. Surface profiles for (a) $D/R=0$; and (b) $D/R=10^{-1}$.

of p on D. As D goes to zero, p approaches 1 as expected from our argument. In Fig. 3(a) we show the resulting surface. With increasing D, p also increases. For $p=2$, $\chi=0$ so the surface roughness would be finite in the limit $L\to\infty$. We expect $p=2$ to be the maximum-allowed value for p. However, already around $D\cong10^{-4}$, power-law behavior is observed only for a *finite* range of h values.

In Fig. 2(b), we show the growth scenario for $D\gg10^{-4}$, in this case $D=10^{-1}$. For earlier times we have a compact film ($c=1$) whose thickness increases with time. The film reaches a maximum height h^* such that at later times $c(h)=1$ for $h<h^*$ and $c(h)<1$ for $h>h^*$. The surface is very flat until it reaches h^* and for $h>h^*$ it grows a mountain structure [see Fig. 3(b)]. The height profile $c(h)$ falls off more rapidly than a power law and it has a width which increases with time. This result has very interesting implications for thin-film growth. It is possible, in our model, to grow flat films if $D/R>0.1$. However, growth should be terminated, or rather R must be reduced, once h reaches the critical height h^* because from then on the film only roughens. Can we compute h^*? If the amplitude of the fastest growing mode is proportional to $\exp^{\omega^* t}$, then roughening becomes important if $\omega^* t$ is of the order 1. The corresponding height $h^*\sim R/\omega^*\sim(D/R)^{1/3}$. The actual critical height h^* was found to depend on D roughly as $D^{0.3}$.

As a function of the surface diffusion constant D, we found a power-law dependence for $c(h)$ if D is small ($D/R<10^{-4}$) with the exponent χ dependent on the value of D. For large D ($D/R>10^{-4}$), the surface has a more compact profile. This is reminiscent of the phase transition of the two-dimensional X-Y model, but more accurate numerical work will be necessary to establish whether or not there is indeed a phase transition at a critical value of D. If we compare these results with the Eden model, then we note that in our case $\chi=\frac{1}{2}$ for $D=0$ and no noise, which also happens to be the χ value for the Eden model *with* noise. Without noise, the Eden model does not lead to rough surfaces. The growth instability of our model is thus intrinsic and not noise driven. The inclusion of noise in Eq. (1) will very likely increase χ beyond $\frac{1}{2}$ in our model. If, in the DLA model, we demand the solid-on-solid condition, then the resulting exponent[10] will be $p\cong0.7$–0.8, which would indicate that neither are we in the universality class of DLA. Surfaces produced by sputtering are, in general, expected to be more rough and convoluted than those produced by molecular-beam epitaxy. In some respects, the growth instability discussed in this Letter is closer to the dendritic growth instability. In that case one also finds a mode dispersion $\omega\sim Uk$ with U the steady-state growth rate.[11]

The most relevant local nonlinearity we neglected is the Eden-model term $(\partial h/\partial x)^2$. We included this term and found that for larger D the mountain landscape of Fig. 3 transforms into a columnar structure. It is possible that this model provides an explanation of the columnar growth mode[12] frequently observed during sputtering.

A serious limitation of the present study is the use of the solid-on-solid condition. Because at late times growth is dominated by a few high peaks, the total growth rate diminishes in time while in actuality it must be a constant. The missing flux goes into side growth of the peaks. Numerical studies[13] of Eq. (1) which go beyond the solid-on-solid condition have shown that due to side growth, most of the mountains coalesce. The growing surface has a finite width and leaves an intricate trail of holes below the surface.[13]

Finally, it should be noted that the restriction to $d=1$ may have exaggerated the shadow effect. Numerical studies in $d=2$ are beyond our current resources.

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[7]This result is derived by noting that for constant θ, Eq. (1) is of a form similar to a Langevin equation allowing the use of the Fokker-Planck method.

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