Scaling Theory for the Growth of Amorphous Films

C. Tang, S. Alexander,[a] and R. Bruinsma[b]
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106
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We present a scaling theory for the time evolution of the morphology of amorphous films, based on the Huygens-principle growth algorithm. During the coarsening stage of the growth, the time-dependent correlation length diverges with time as $\xi(t) \propto t^p$. We calculate p for a range of random and self-similar starting surfaces. When the effect of noise is taken into account, the exponent p reaches a universal value $\frac{1}{4}$, in good agreement with experiments.

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Amorphous films have a wealth of applications[1] in areas such as magnetic films for recording, conducting films for contacts, and amorphous-silicon films for photovoltaics. These films can be grown in a variety of ways. A frequently used method, with which we are concerned here, is sputter deposition.[2] During sputter deposition, energetic atoms move along ballistic trajectories with random incident angles and are deposited on a substrate. Sputter-grown films can exhibit fascinating large-scale structures which affect the physical properties of the film. At temperatures small compared to the melting temperature ("zone I"), the surface of the film is a collection of hill-like domed columns separated by deep grooves extending down to the substrate.

It is important to note that even when the film thickness h and the typical length scale of the surface morphology are macroscopic (of order cm and mm, respectively), the growth process is still in the "coarsening" stage: As the film thickness increases with time, the surface morphology retains its basic characteristic features except that the typical length scale of the columns, $\xi(t)$, increases with time.[3] The bigger columns swell while at the same time overwhelming smaller columns in a "survival of the fittest" competition. If one makes a time sequence of micrographs of the surface,[3] then it is possible to trace the evolution of the most striking surface features from early to late times. Similar coarsening phenomena are encountered for simple statistical-mechanics models, such as the Ising model, during the low-temperature annealing of a disordered initial state. As first discussed by Lifshitz and Slyozov,[4] coarsening obeys a scaling law for the characteristic length $\xi(t)$ of the form

$$\xi(t) \sim t^p . \tag{1}$$

Roy and Messier[5] (RM) found that Eq. (1) is obeyed for the growth of silicon carbide films under various conditions. At low temperatures they found that $p \approx 0.73$. For higher temperatures p depends on the film thickness h: For very thin films (~ 100 nm), p could be as small as 0.2. With increasing h, p did increase but it remained less than the low-temperature value. The aim of this Letter is to show that a well-known and simple growth mechanism, the Huygens principle (HP), can account for these scaling laws with values of p which are in agreement with experiment.

The low-temperature morphology of an amorphous film is mainly determined[2,6-8] by self-shadowing and surface diffusion. Shadowing magnifies the initial height irregularities, producing deep grooves between neighboring columns. Surface diffusion smoothes out surface irregularities on length scales less than the diffusion length $l_0 = (\mu \Omega^2 r \gamma / v)^{1/3}$. Here, μ is the surface mobility, Ω is the atomic volume, r is the area density of atoms, γ is the surface tension, and v is the growth velocity. In Fig. 1, the result is shown of a numerical calculation for small l_0 by Bales and Zangwill[7] with the initial surface a sine wave. The characteristic domed columns and grooves of zone I are apparent. Divide the surface of Fig. 1 into two parts: the smoothly varying regions around the broad tops of the columns and the narrow, deep grooves which have a width of order l_0. Shadowing from neighboring columns is important for the grooves (it stabilizes them) but *not* for the broad tops. Because the shadowing effect is not important, broad tops evolve (up to a uniform translation) as if they are growing with constant velocity along the surface normal.[9] This latter growth mechanism is nothing else but the HP of geometrical optics. The HP has been applied with success to the evolution of macroscopic surface features during growth and

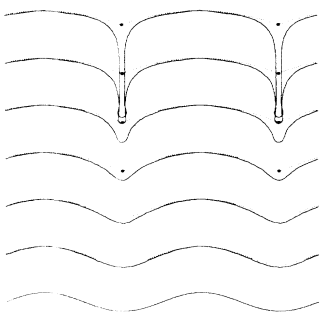

FIG. 1. Solid lines: Evolution of the surface by the shadow model (Ref. 7); dotted lines: Evolution of the same initial surface by the Huygens principle plus a translation, i.e., $v_n = v \times (1 + \cos\theta)/2$ (see Ref. 9). The solid circles indicate the cusp singularities.

erosion,[10] in particular for photolithography. We also show in Fig. 1 (dotted lines) the result of a HP construction which started from the same initial conditions. The HP construction reproduces, as claimed, the top parts of the column, while in the region of the grooves, the HP construction produces lines of cusplike mathematical singularities. This same relation between grooves and cusp singularities was also found for more complex initial conditions where we compare the HP construction with the numerical work of Ref. 8.

For a growing thin film, the heights of the maxima of the columns are of course in general not the same (as they are in Fig. 1). However, as long as the typical height difference δh between column tops is small compared to the distance ξ between neighboring columns, shadowing should not play a dominant role for the evolution of the column tops. We will later show that indeed $\delta h/\xi \to 0$ at large times. Shadowing *is* important in the groove regions, as mentioned. However, for l_0 small compared to the width ξ of the columns, we may think of the grooves as sharp boundaries between the columns. The dynamical evolution of the grooves is then entirely determined by the growth or shrinkage of the columns — which themselves obey the HP.

We thus propose the HP construction as a model for the coarsening of amorphous films with the cusp singularities indicating the position of the grooves. This procedure may be compared with the use of macroscopic elasticity theory to describe the deformations of a solid containing structural defects. The mathematical singularities of elasticity theory also correspond to physical defects such as vacancies, dislocations, etc. Our model thus reduces the coarsening problem of a film to a study of the evolution of the mathematical singularities of the HP construction. To understand the evolution of the singularities of the HP construction, we first discuss a series of numerical HP constructions for a variety of initial surfaces and then compare these results with scaling laws derived from the HP construction.

We shall consider two classes of initial surfaces. We start with random surfaces with no spatial correlations in the height profile. In Fig. 2, we show the evolution of the cusp singularities on a one-dimensional ($d=1$) substrate for the case of Gaussian initial randomness. The

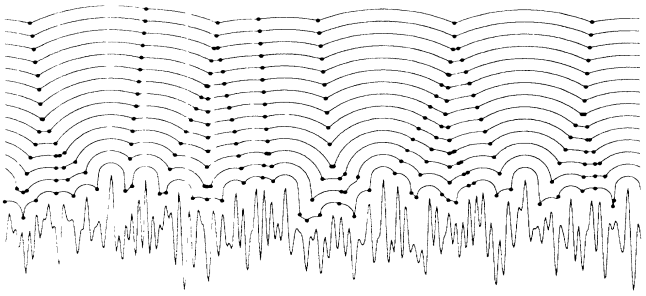

FIG. 2. The evolution of a random initial surface according to the Huygens principle. The solid circles indicate the cusp singularities.

singularities collide and their density decreases as the film height increases. The network of singularities shown in Fig. 2 bears a remarkably close resemblance to the subsurface groove networks found by Messier and Yehoda.[3] The characteristic length ξ for the surface features of Fig. 2 (which is simply the mean distance between singularities) indeed has a power-law dependence on time. In Table I, we list the exponents p found for uncorrelated initial surfaces with, respectively, uniform, Gaussian, and power-law height distributions for $d=1$ and 2. Next, we consider the case where the initial surface is correlated and in particular where it is *self-similar*. By this we mean that at $t=0$ the height-height correlation function obeys the relation

$$\langle |h(\mathbf{x})-h(\mathbf{x}')| \rangle_{t=0} \sim |\mathbf{x}-\mathbf{x}'|^{\zeta}, \tag{2}$$

with $0<\zeta<1$. Two cases are of particular interest. If the starting surface is thermally roughened, then the mean square of the Fourier transform of the height fluctuations, $\langle |h_{\mathbf{q}}|^2 \rangle$, has a spectrum proportional to $1/|\mathbf{q}|^2$, which implies $\zeta=\frac{1}{2}$ in $d=1$ (random walk) and $\zeta \to 0$ in $d=2$. If the starting surface is exposed to shot noise and annealed by surface diffusion, then we expect $\langle |h_{\mathbf{q}}|^2 \rangle \propto 1/|\mathbf{q}|^4$, in which case $\zeta=1$ in $d=2$. The exponents p for these two cases are also listed in Table I.

As is immediately evident from Table I, the exponent p is a sensitive function of both the initial height distribution function (for uncorrelated random surfaces) and the initial height correlation function (for self-similar

TABLE I. Analytic and numerical exponents for several initial surfaces.

	$d=1$		$d=2$	
Initial surface	Analytic	Numerical	Analytic	Numerical
Gaussian	$\frac{1}{2}$	0.48	$\frac{1}{2}$	0.47
Uniform	$\frac{1}{3}$	0.333	$\frac{1}{4}$	0.22
Power law	($\tau=3$)		($\tau=4$)	
	$\frac{2}{3}$	0.67	$\frac{3}{4}$	0.73
Self-similar ($\langle |h_{\mathbf{q}}|^2 \rangle \sim 1/q^2$)	$\frac{2}{3}$	0.63	$\frac{1}{2}$	0.44
Self-similar ($\langle |h_{\mathbf{q}}|^2 \rangle \sim 1/q^4$)			1	0.9

surfaces). How can we understand this wide range of coarsening exponents? We proceed by asking the following question: What is the chance for a peak, say $h(\mathbf{0})$ at $\mathbf{x}=\mathbf{0}$, to survive after a time t? We can estimate the survival probability for uncorrelated initial surfaces in the following way. As indicated in Fig. 3, a peak $h(\mathbf{x}_0)$ at $\mathbf{x}_0$ cannot "screen" $h(\mathbf{0})$ if $h(\mathbf{x}_0)<h(\mathbf{0})+x_0^2/2vt$, with v the growth velocity and with $|\mathbf{x}_0|\ll vt$. The survival probability for $h(\mathbf{0})$ is then

$$p_s(t)\propto\prod_{\mathbf{x}\neq\mathbf{0}}\text{Prob}\{h(\mathbf{x})<h(\mathbf{0})+x^2/2vt\}. \quad (3)$$

Averaging over $h(\mathbf{0})$ with respect to the initial height distribution gives

$$n(t)=\int_0^\infty p_s(t)P(h(\mathbf{0}))dh(\mathbf{0})$$

$$\propto\int_0^\infty dh\,P(h)\exp\left\{\sum_{\mathbf{x}\neq\mathbf{0}}\ln\int_0^{h+x^2/2vt}dh'P(h')\right\}. \quad (4)$$

Here, $n(t)$ is the surface density of hills and $P(h)$ is the height distribution function of the initial surface. It is straightforward to calculate the exponent p from Eq. (4) and the fact that $n(t)=1/\xi^d(t)$. For uniform $[P(h)=\text{const},\ 0\le h\le h_{\max}]$, power-law $[P(h)\propto h^{-\tau},\ h_{\min}\le h<\infty]$, and Gaussian distributions of the initial height, we get, respectively, $p=1/(d+2)$, $p=(\tau-1)/[2(\tau-1)-d]$ $(\tau\ge d+1)$, and $p=\frac{1}{2}$. For the power-law distribution with $2<\tau<d+1$, our simple argument fails. Numerical results indicate that $p=1$ in that case.

To predict the coarsening exponent for self-similar initial surfaces, we use a heuristic scaling argument. Note that we can define the correlation length $\xi(t)$ by the condition that a column a distance $\xi(t)$ from the origin first starts to screen the column at the origin at a time t. From the preceding argument (see Fig. 3), this leads to the requirement that $\langle|h(\xi)-h(0)|\rangle_{t=0}\approx\xi^2(t)/vt$. If we combine this result with Eq. (2), we find[11] $p=1/(2-\zeta)$. This result should be valid in any dimension. The above analytically calculated exponents compare very well with the numerical results in Table I.

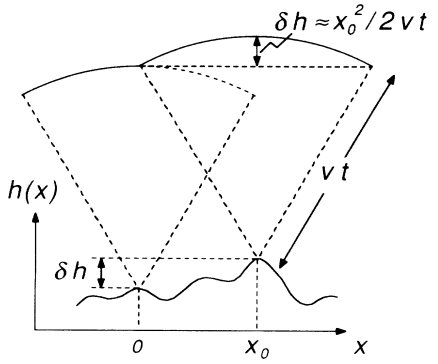

FIG. 3. A peak at x_0 which is just starting to screen a peak at the origin. The initial height difference between the peaks δh must equal $vt-[(vt)^2-x_0^2]^{1/2}\approx x_0^2/2vt$ in order for this condition to be satisfied.

The continuum growth theory discussed so far leads, however, to a physical inconsistency for $p<\frac{1}{2}$. As we saw from Fig. 3, the typical height difference between columns, $\delta h(t)$, is of order ξ^2/vt. For large t and $p<\frac{1}{2}$, $\delta h(t)$ goes to zero as t^{2p-1}. Clearly, for $\delta h(t)$ of order of an atomic size, continuum theory would be invalid. The most important discreteness effect neglected in our continuum theory is the noise in the deposition current. This noise is a combination of shot noise and instrumental noise. These statistical fluctuations in the deposition current in turn lead to column-height fluctuations. We will see that for $p<\frac{3}{4}$, these noise-induced height fluctuations eventually dominate the evolution.

To understand how noise plays a role in the growth process, let us first examine the effect of noise on a single column. Imagine that there are some noise-induced local height fluctuations around the column maximum. Surface diffusion will quickly average these fluctuations over an area l_0^2 (in $d=2$), typically forming a small bump which has a width of order l_0. This bump will then spread along the paraboloid top of the column due to the lateral growth. Since the column size ξ increases in time as t^p, with $p<1$, and the speed of the lateral growth is proportional to vt, such a fluctuation can always spread over the whole column top.[12] Thus, during the growth process up to a time t, the column top retains, on average, its paraboloid shape, while its height will deviate from the average height vt by an amount proportional to $t^{1/2}/l_0$. Next, we notice that, because of the deep grooves, mass transfer between columns is unlikely. If we restrict ourselves to shot noise, then at time t the typical height difference δh_N of the columns, induced by noise, is (in $d=2$)

$$\delta h_n\approx(a^{3/2}/l_0)(vt)^{1/2}, \quad (5)$$

where a is an atomic size. For other uncorrelated noise sources or for d other than 2, δh_N will still be proportional to $t^{1/2}$ but the coefficient in Eq. (5) would be different. As long as $\delta h_N\ll\delta h\approx\xi^2/vt$, we can neglect noise. From Eq (5), it follows that for large t, this requires $p>\frac{3}{4}$. What happens if $p<\frac{3}{4}$ for our starting surface? Apparently, the noise-induced height difference δh_N will then eventually control the growth, i.e., $\delta h_N\approx\xi^2/vt$ at large times. We will call this regime "late-stage coarsening." The associated correlation length ξ_N is, using Eq. (5),

$$\xi_N\approx(a^{3/4}/l_0^{1/2})(vt)^{3/4}. \quad (6)$$

The coarsening exponent p for late-stage growth is thus $p=\frac{3}{4}$.

Crossover from "early-stage coarsening" (with $\delta h_N\ll\xi^2/vt$) to late-stage coarsening takes place when $\xi(t)=\xi_N(t)$. To estimate the crossover height h^*, we will take the case of Gaussian initial randomness. Then, during early-stage growth $\xi(t)\approx(w_0vt)^{1/2}$, with w_0 the width of the starting surface. Equating $\xi(t)$ with $\xi_N(t)$ gives $h^*\approx(w_0l_0)^2/a^3$. To estimate h^*, we note that l_0

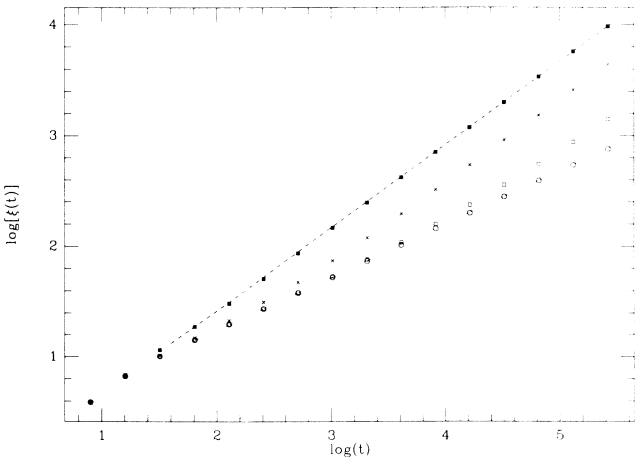

FIG. 4. The average column size ξ as function of t calculated from the HP construction in $d=1$ for a substrate of size $L=300000$. The initial random surface has Gaussian distribution. The noise amplitude is 0.5 (solid squares), 0.1 (crosses), 0.01 (open squares), and 0 (circles) (arbitrary units). The dashed and dotted straight lines have slopes 0.75 and 0.48, respectively.

is strongly temperature dependent through the surface mobility μ. At very low temperatures, l_0 will be of order a so $h^* \approx w_0^2/a$. In this regime, we are then *always* in the late-stage growth regime and $p=\frac{3}{4}$. However, for elevated temperatures and low deposition rates l_0 will exceed 1000 Å. In that case, h^* will exceed 1 mm for $w_0=10$ Å and we expect early-stage behavior or, at best, crossover behavior. Note from Eqs. (5) and (6) that in the continuum limit $a \to 0$, we do not encounter late-stage growth.

In Fig. 4, we show the result of a numerical calculation in $d=1$ starting from an initial surface with Gaussian randomness. Without noise (zero noise level), $p \approx 0.48$. For high noise levels, $p \approx 0.75$, in excellent agreement with the predicted value. For low noise levels, we see a slow crossover from early-stage growth to late-stage growth. These results appear to given a good account for the experiments of RM on silicon carbide. The measured low-temperature p value (0.73) is close to $\frac{3}{4}$, while for higher temperatures, lower exponents were measured. The increase of p with h due to crossover from early- to late-stage growth was also noted.

As we noted in the introduction, the validity of the HP construction applied to amorphous film growth is based on two conditions. First, "shadow" competition between columns should be unimportant. This requires that $\delta h/\xi \approx \xi/t \approx t^{p-1}$ should be small, and as long as $p<1$, this condition is satisfied. Second, we demanded that $l_0/\xi \to 0$. Since l_0 is independent of time, while ξ grows with time, this condition will eventually be satisfied. Typically, l_0 is less than 1 μm while ξ can be as large as 1 mm.

In summary, the HP construction predicts scaling laws for the coarsening of the amorphous thin-film growth of zone I which are in good agreement with experimental observations. The singularity networks of the HP construction correspond to the groove networks of amorphous films. At low temperatures, the experimentally measured exponent agrees well with our "universal" value $\frac{3}{4}$ for late-stage coarsening. At higher temperatures, the apparently smaller exponents observed experimentally are, according to our theory, due to crossover effects from early- to late-stage coarsening.

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[a]Permanent address: The Racah Institute of Physics, The Hebrew University, Jerusalam, Israel, and Department of Physics, University of California, Los Angeles, CA 90024.

[b]Permanent address: Department of Physics, University of California, Los Angeles, CA 90024.

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[9]For the shadow model of Refs. 6–8, it can be shown (see Ref. 7) that if one neglects the shadowing effect from neighboring columns, then in the case of uniform exposure, the broad tops in Fig. 1 evolve with a normal velocity $v_n = v(1+\cos\theta)/2$, where θ is the angle of the surface normal with respect to the vertical direction. We can decompose v_n as the sum of a uniform translation, with growth velocity $v\cos\theta/2$, and of a normal growth, with growth velocity $v/2$. The uniform translation does not affect the growth morphology. The normal growth is the HP term.

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[12]This is only valid if we assume that the noise does not produce *new* groove defects. If that were an allowed process, then a bump would spread as t^p instead of t. For the groove networks of Ref. 3, this appears to be a valid assumption.