# Breakdown of Dynamic Scaling in Surface Growth under Shadowing

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Using Monte Carlo simulations and experimental results, we show that for common thin film deposition techniques, such as sputter deposition and chemical vapor deposition, a mound structure can be formed with a characteristic length scale, or "wavelength" $\lambda$, that describes the separation of the mounds. We show that the temporal evolution of $\lambda$ is distinctly different from that of the mound size, or lateral correlation length $\xi$. The formation of a mound structure is due to nonlocal growth effects, such as shadowing, that lead to the breakdown of the self-affinity of the morphology described by the well-established dynamic scaling theory. We show that the wavelength grows as a function of time in a power law form, $\lambda \sim t^p$, where $p \approx 0.5$ for a wide range of growth conditions, while the mound size grows as $\xi \sim t^{1/z}$, where $1/z$ varies depending on growth conditions.

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Thin film surface morphology controls many important physical and chemical properties of the films. It is therefore of great interest to understand and control the evolution of the surface morphology during thin film growth. The formation of a growth front is a complex phenomenon and very often occurs far from equilibrium. When atoms are deposited on a surface, atoms do not arrive at the surface at the same time uniformly across the surface. This random fluctuation, or noise, which is inherent in the process, may create surface growth front roughness. The noise competes with surface smoothing processes, such as surface diffusion, to form a rough morphology if the experiment is performed at either a sufficiently low temperature or at a high growth rate.

A conventional statistical mechanics treatment cannot be used to describe this complex phenomenon. About two decades ago, a dynamic scaling approach [1] was proposed to describe the morphological evolution of a growth front. Since then, numerous modeling and experimental works have been reported based on this dynamic scaling analysis [2,3]. In this analysis, the surface is described by the equal-time height-height correlation function $H(\mathbf{r})$, defined as $H(\mathbf{r}) = \langle [h(\mathbf{r}) - h(\mathbf{0})]^2 \rangle$. Here, $h(\mathbf{r})$ is the surface height at a position $\mathbf{r} = (x, y)$ on the surface. The notation $\langle \cdots \rangle$ denotes a statistical average. The dynamic scaling hypothesis requires that $H(r) \sim r^{2\alpha}$ for $r \ll \xi$ and $H(r) \sim 2w^2$ for $r \gg \xi$, where $\xi$ is the lateral correlation length, $w$ is the interface width or root mean square (rms) roughness, and $\alpha$ is the roughness exponent, which describes how "wiggly" the local surface is. Both $w$ and $\xi$ grow as a power law in time, $w \sim t^{\beta}$ and $\xi \sim t^{1/z}$, where the exponents $\beta$ and $1/z$ are called the growth exponent and dynamic exponent, respectively. Dynamic scaling requires $z = \alpha/\beta$ [1].

One notes that in the dynamic scaling hypothesis, the surface vertical direction does not scale the same way as does the lateral direction. It is therefore not a self-similar surface, but rather a self-affine surface. An important feature of a self-affine surface is that the height-height correlation function reaches a constant value (equal to $2w^2$) at a large distance at a given time. This distance defines the lateral correlation length $\xi$, beyond which the surface height fluctuations are not correlated. This means that there is no long range characteristic length scale involved, the surface height fluctuation is random beyond the correlation length. This assumption is valid for a number of surface growth models [2,3] where local smoothing effects such as surface diffusion is operative to compete with the noise.

However, in practice, in many common modern deposition techniques, including sputter deposition and chemical vapor deposition (CVD), nonlocal effects such as shadowing [4,5] along with the redistribution of atoms reflected from the surface due to a nonunity sticking coefficient [6] can play an important role in defining the surface morphology during growth. We show that these nonlocal effects give rise to a mound structure that cannot be described within the context of self-affinity. A mound structure possesses a characteristic long range length scale $\lambda$, or wavelength, that is a measure of the average distance between mounds. (This mound structure is unrelated to that created by step barrier diffusion in molecular beam epitaxy (MBE) [7]. Since mounds in MBE are formed by a local growth effect, the growth dynamics can be described using a local continuum equation [8], whereas the growth effects considered in this Letter are nonlocal. The wavelength $\lambda$ for surfaces in MBE has been shown to behave as a power law $\lambda \sim t^p$, where $p$ ranges from 0.16 to 0.26 [9,10].) The quasiperiodic behavior quantified by this wavelength is distinctly different from the behavior of the mound size, or the lateral correlation length $\xi$. Using Monte Carlo simulations and experimentally deposited surfaces, we show that the separation of mounds grows as a function of time in a power law form, $\lambda \sim t^p$, where $p \approx 0.5$ for a wide range of deposition conditions under nonlocal shadowing and reemission effects. On the other hand, the growth exponent $1/z$ that is associated with the growth of the mound size, $\xi \sim t^{1/z}$, does depend on deposition conditions such as the sticking coefficient. We show that deviation in the growth of the mound separation and the mound size leads to a breakdown of the self-affinity and dynamic scaling of the system.

The primary nonlocal growth effect is the shadowing effect [4,5], where taller surface features block incoming flux from reaching lower lying areas of the film. This allows taller surface features to grow at the expense of shorter ones, leading to a coarsening of the surface. Shadowing is an inherently nonlocal process because the shadowing of a surface feature depends on the heights of all other surface features, not just close, or local, ones. However, the formation of mounds can be hindered by the reemission of particles during deposition. The reemission effect allows atoms to ''bounce around'' before they settle at appropriate sites on the surface [6]. Reemitted particles serve to change the overall particle flux incident on the surface, allowing previously shadowed surface features to receive particle flux. To describe the reemission effect, a sticking coefficient ($s_0$) is used which represents the probability an atom will stick to the surface when it first strikes. We assume reemitted particles will always stick. During deposition, shadowing tends to roughen the surface and reemission tends to smooth the surface [11].

The solid-on-solid $2 + 1$ dimensional Monte Carlo (MC) simulations used in this research have been designed to mimic the growth of thin films by normal incidence deposition, CVD, and sputter deposition. In normal incidence deposition, the incident flux is uniformly normal to the surface, whereas for CVD and sputter deposition, the incident flux has an angular distribution of $\cos\theta$, where $\theta$ is defined with respect to the surface normal. A cosine flux distribution is typical of CVD and sputter deposition at higher working gas pressures, where the mean free path of incident particles is small compared to the geometrical dimensions of the source-substrate separation. For a detailed description of the MC simulations used, see Karabacak, *et al.* [12]. To study the behavior of wavelength selection, we analyze the circularly averaged power spectral density function (PSD), which is the two-dimensional Fourier transform of the surface heights. The wavelength $\lambda$ is defined to be the reciprocal of the position of the peak $k_m$ in the PSD frequency spectrum. By measuring the time evolution of the position of this peak, we can capture the essence of the dynamics of mounds evolution.

The shadowing effect is active only when there exists an angular distribution of incident particle flux. If there is no angular flux distribution, taller surface features cannot block the incoming flux from the lower lying areas of the surface, and shadowing is not effective. Thus, in normal incidence deposition, there is no shadowing because the incident flux is uniformly normal to the surface. We find that in the normal incidence deposition simulations, no wavelength selection is seen for all values of the sticking coefficient $s_0$. However, once an angular distribution of flux is introduced, as in the CVD and sputter deposition simulations, wavelength selection is clear. Thus, when the shadowing effect is dominant, wavelength selection is manifested. This is consistent with the hypothesis that the nonlocal shadowing effect contributes to the creation of mound structures. To extract the wavelength exponent $p$, the time dependent behavior of the peak $k_m$ of the PSD is extracted and found to behave as a power law, $k_m \sim t^{-p}$. Figure 1 contains a plot of $\lambda$ as a function of time for the cosine flux MC simulation with $s_0 = 1$ where the exponent $p = 0.49 \pm 0.02$. When the sticking coefficient $s_0$ is reduced in the simulations, the value of the wavelength exponent remains relatively constant at $p \approx 0.5$. However, once the sticking coefficient is sufficiently small ($s_0 < 0.5$), the reemission effect is strong enough to redistribute a significant amount of particle flux to otherwise shadowed surface heights, which effectively cancels the shadowing effect and eliminates the wavelength selection. The fact that the wavelength exponent is independent of the sticking coefficient (for $s_0 > 0.5$) could suggest that these mounded surfaces may have a ''universal'' behavior when regarding wavelength selection.

It is important to note that the lateral correlation length $\xi$, or mound size, and the average mound separation $\lambda \propto k_m^{-1}$, describe different aspects of the surface morphology, and therefore may behave differently. The lateral correlation length $\xi$ can be obtained from the height-height correlation function $H(\mathbf{r})$, and the exponent $1/z$ can be obtained from the log-log plot of $\xi$ versus deposition time. In Fig. 1, the time dependence of the average mound separation $\lambda$ and lateral correlation length $\xi$ is plotted for the cosine flux MC simulation with $s_0 = 1$. Since the exponents $p$ ($\approx$0.49) and $1/z$ ($\approx$0.33) are not equal, a clear difference in behavior can be seen. Unlike the constant value of $p \approx 0.5$, we have found that the value of $1/z$ does vary as a function of $s_0$, ranging from 0.1 to 0.6,

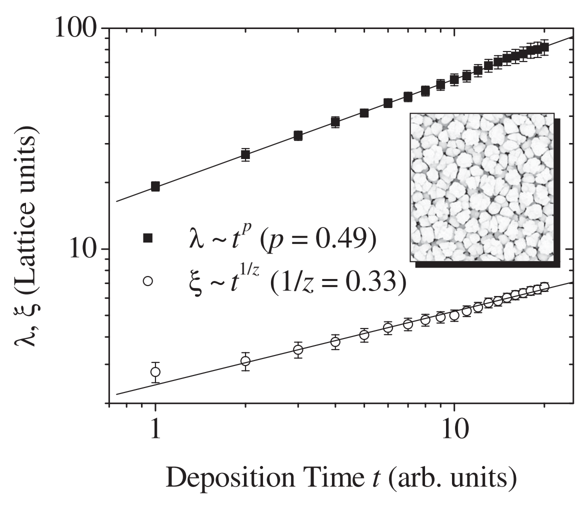


FIG. 1. Plot of average mound separation $\lambda$ and mound size (or lateral correlation length) $\xi$ for the cosine flux MC simulation with sticking coefficient $s_0 = 1$. The deposition time $t$ is defined such that one time step corresponds to an average of 50 deposited particles per lattice point. Inset: A top view of the simulated surface at $t = 10$.

indicating a nonuniversal behavior. In general, $1/z$ tends to decrease as $s_0$ decreases when wavelength selection is present.

Previous studies on the effects of shadowing [4,5] and reemission [6] did not examine quantitatively the behavior of the time evolution of the wavelength $\lambda$. It is important to note that some authors have used the variable $p$ to describe the time evolution of the lateral correlation length as opposed to wavelength selection. Using a model based on the Huygens principle (HP), Tang, *et al.* [4] examined the evolution of the lateral correlation length $\xi$ of simulated surfaces. The exponent $1/z$ associated with the lateral correlation length depends on the initial surface configurations, and ranges from $1/4$ to 1. However, under the HP, mounds grow next to each other without gaps, and the spacing between mounds is the same as the mound size, or $\xi = \lambda$, which implies $1/z = p$. A continuum model presented in Yao, *et al.* [5] accounted for shadowing during the growth process which predicted $1/z \approx 0.33$ [13], consistent with our prediction under the specific condition of $s_0 = 1$. Experimentally, the value of $1/z$ associated with the lateral correlation length reported in the literature scatters between 0.13 to 0.85 [12,14–23]. Therefore, it is reasonable to believe that the value of $1/z$ is not universal and strongly depends on deposition conditions.

Experimentally deposited films have also been analyzed to investigate mound formation. A dc magnetron sputtering system was used to deposit amorphous Si on an initially flat Si(100) substrate. In all depositions, a power of 200 watts and an Ar pressure of $2.0 \times 10^{-3}$ torr was used. Depositions ranging from 7.5 to 960 min were performed at a deposition rate of approximately 8 nm/min. The surfaces were imaged using atomic force microscopy (AFM). For each deposition, statistics from four different AFM scans have been averaged, and the results are depicted in Fig. 2. The analysis gives $p = 0.51 \pm 0.03$, $1/z = 0.38 \pm 0.03$, and $\beta = 0.55 \pm 0.09$ consistent with the results of MC simulations with a sticking coefficient $s_0 \approx 0.7$, well within the regime of wavelength selection as predicted by simulation results. Even though shadowing is present in this deposition, $\beta < 1$ because reemission is also significant, which slows the growth of the interface width. In addition, amorphous SiN films have been deposited using a plasma enhanced CVD (PECVD) procedure. A similar analysis of the time evolution of the PSD gives $p = 0.50 \pm 0.06$, along with $1/z = 0.28 \pm 0.02$ and $\beta = 0.37 \pm 0.01$. Note that $\beta$ need not equal one under shadowing growth, although, from simulation results, $\beta = 1$ under pure shadowing growth with no reemission.

When speaking of the scaling behavior of a surface, it is important to distinguish between the scaling of the physical dimensions of the surface and the scaling of the statistical properties of the surface as a function of time. The term self-affine deals with the scaling of the physical dimensions of the surface. Self-affine surfaces are defined similarly to fractals in that the vertical and horizontal directions of the surface can be rescaled to yield a new

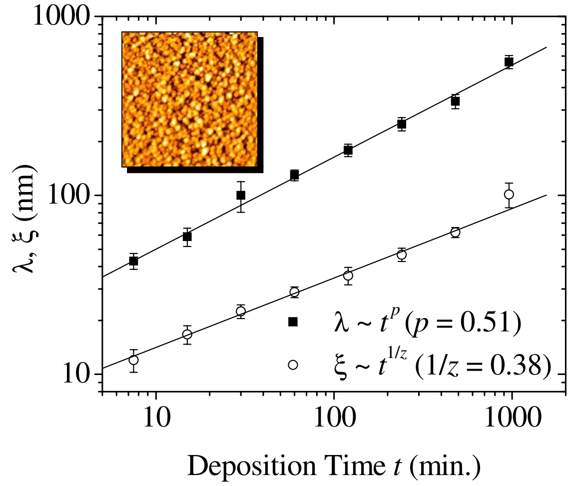


FIG. 2 (color online). Plot of average mound separation $\lambda$ and mound size (or lateral correlation length) $\xi$ for sputtered Si on Si. The extracted values for the exponents are $p = 0.51 \pm 0.03$ and $1/z = 0.38 \pm 0.03$. Inset: AFM top view image of the surface at $t = 120$ min. The AFM image size is $2\ \mu\text{m} \times 2\ \mu\text{m}$.

surface that is statistically identical to the original surface [2]. This self-affine property of the surface requires that the PSD have no characteristic peak [11]. However, this notion of scaling breaks down when the PSD has a peak as has shown to be the case in thin films created during CVD and sputter deposition, which precludes mounded surfaces from being characterized as self-affine.

In addition, one can also consider the time evolution of a surface and any scaling behavior it may exhibit. As discussed earlier, the dynamic scaling hypothesis predicts a relationship between various exponents associated with the time evolution of surface statistics. It can be shown that when the dynamic scaling hypothesis holds, the PSD of a surface exhibits a time dependent scaling [11]. However, for mounded surfaces, the difference in behavior between the lateral correlation length and average mound separation has a profound impact on the scaling of the PSD. In the PSD of a mounded surface, the peak position $k_m$ is related to the wavelength $\lambda$ as $k_m \propto \lambda^{-1} \sim t^{-p}$. The full-width-at-half-maximum (FWHM) of the PSD is inversely proportional to the lateral correlation length [11], and thus behaves as $\xi^{-1} \sim t^{-1/z}$. Thus, the time evolution of the peak of the PSD is governed by the exponent $p$, whereas the time evolution of the spread of the PSD is governed by the exponent $1/z$. Therefore, if $p \approx 1/z$, the overall shape of the PSD will scale with time. However, as the values for $p$ and $1/z$ become separated, the overall scaling behavior of the PSD breaks down. This behavior is clearly seen in Fig. 3(a), which contains various PSD extracted at different stages in the evolution of surfaces created in the cosine flux MC simulation with $s_0 = 1$. A similar plot is shown in Fig. 3(b) measured from sputtered Si surfaces described earlier. The PSD curves are scaled so their peaks coincide, which results in the wave number axis multiplied by a factor of $\lambda \sim t^p$. Since the peak position defines the value

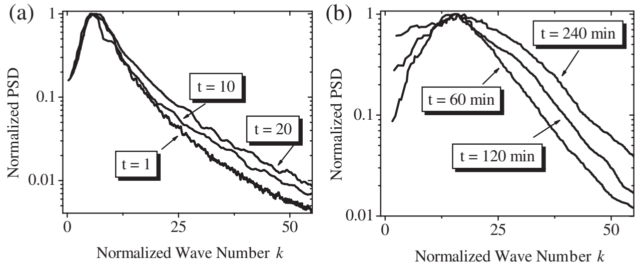


FIG. 3. (a) Rescaled PSD curves for the cosine flux MC simulation with sticking coefficient $s_0 = 1$. The curves are scaled according to peak position, which scales the long range (small $k$) behavior of the PSD. The overall spread of the curves (large $k$) does not scale, evidence of a breakdown of dynamic scaling. (b) Rescaled PSD curves for experimentally deposited sputter Si on Si.

for the wavelength, scaling the peaks of the curves corresponds to scaling the surfaces according to long range (small wave number) behavior. A clear deviation is observed in the spread of the curves. The behavior of the PSD for larger wave numbers corresponds to the short range behavior of the surface as represented by the lateral correlation length. Since $p \neq 1/z$ for these surfaces, these length scales do not evolve at the same rate, which leads to the behavior seen in Fig. 3. In the scaled curves, the spread is proportional to $t^{-1/z}t^{p} = t^{p-1/z}$, and since $p > 1/z$ in these examples, the widths of the scaled curves increase with time. It follows that measuring different values for the exponents $p$ and $1/z$ is evidence of the breakdown of dynamic scaling for mounded surfaces, as has been shown in both simulations and experimentally deposited mounded surfaces. Therefore, in general, the nonlocal effects that lead to mound formation do not allow the system to scale, and the system loses its self-affine and dynamic scaling behavior.

In conclusion, we have presented a study of mound formation during thin film growth. The wavelength exponent $p$ can be used to characterize the evolution of mounds on the film, defined in terms of the time evolution of the peak position of the PSD, $k_m \sim t^{-p}$. For the surfaces studied in this work, $p \approx 0.5$, independent, within error, of the strength of reemission. A comparison of the average mound separation $\lambda$ and lateral correlation length $\xi$ reveals that their behavior is not necessarily the same, evidence that the entire system does not scale as one. From our analysis, thin film deposition appears to be much more complex than originally anticipated due to nonlocal effects. However, the evolution of the wavelength that characterizes the separation of mounds appears to be universal.

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