

Molecular-Beam Epitaxial Growth and Surface Diffusion

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We investigate the statistical properties of the surface of thin films grown by molecular-beam epitaxy (MBE). We present and analyze a simple model of MBE growth which incorporates surface diffusion and deposition in a physically correct manner. The short-time behavior does not correspond to that predicted by the continuum model of Villain, Das Sarma, and others. At long times, the model is governed by Kardar-Parisi-Zhang dynamics.

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There has been much recent interest in the statistical properties of nonequilibrium surfaces. In particular, much attention has been focused on theoretical models to describe thin-film growth by molecular-beam epitaxy (MBE). This work is particularly germane given the very recent observation [1] of power-law scaling of surface roughness in iron films grown by MBE. A number of groups [2-4] have proposed that the statistical properties of MBE growth are given by the fourth-order continuum equation [2]

$$\dot{h} = -D_4 \nabla^4 h + \lambda_4 \nabla^2 (\nabla h)^2 + \eta, \quad (1)$$

where $h(\mathbf{x}_{\parallel}, t)$ is the height of the surface and η is a noise source with correlations

$$\langle \eta(\mathbf{x}_{\parallel}, t) \rangle = 0, \quad (2)$$

$$\langle \eta(\mathbf{x}_{\parallel}, t) \eta(\mathbf{x}'_{\parallel}, t') \rangle = S \delta^d(\mathbf{x}_{\parallel} - \mathbf{x}'_{\parallel}) \delta(t - t').$$

We shall argue in this paper that the theoretical arguments tying the above continuum equation to MBE growth possess a number of weak points. We shall then introduce a model which captures the basic physics of MBE in a computationally efficient manner so that the predictions of Eq. (1) can be put to a test. The results of the simulations of our model will be seen to indicate that Eq. (1) does not adequately describe the statistical properties of MBE growth. At long-time and length scales our model crosses over to a regime described by the Kardar-Parisi-Zhang (KPZ) equation [5]

$$\dot{h} = D_2 \nabla^2 h + \lambda_2 (\nabla h)^2 + \eta \quad (3)$$

as overhangs become important. However, even before this crossover, Eq. (1) does not correctly predict the scaling behavior. The nature of an appropriate continuum description for the pre-KPZ regime will be discussed in the conclusion.

Let us first review the physical motivation of Eq. (1), which lies in the fact that the essential mechanism of surface relaxation is surface diffusion (SD). In this case, as shown by Mullins [6], the growth of the surface should be described by

$$v_n = D_4 \nabla_{LB}^2 \kappa + \mathcal{F}, \quad (4)$$

where v_n is the normal velocity of the interface, κ the curvature, ∇_{LB}^2 the Laplace-Beltrami (intrinsic Laplacian) operator on the surface, and \mathcal{F} the (fluctuating) flux. Assuming the flux is normally incident so that growth is primarily in the transverse ($\hat{\mathbf{x}}_{\perp}$) direction and expanding about a flat interface yields

$$\dot{h} = -D_4 \nabla^4 h + \mathcal{F} + \dots \quad (5)$$

This is similar to Eq. (1) above, albeit with some important differences. First, there is *no* nonlinear term of the form $\nabla^2 (\nabla h)^2$. The physical basis of this fact is that the chemical potential is independent of the orientation of the surface (except for lattice effects, ignored in this continuum description). This symmetry argument would seem to imply that such a term cannot be generated by coarse graining, and so λ_4 should be zero to all orders in a renormalization-group expansion. This is consistent with the fact that λ_4 is not renormalized [4], at least to one-loop order [7], so that if the bare theory has λ_4 equal to zero, it cannot be induced by other nonlinear terms. In addition, Mullins' equation as well as its expansion equation (5) have the symmetry (in the frame moving with the average velocity of the interface) $h \rightarrow -h$. (Higher powers of κ in Mullins' equation which would break this symmetry are irrelevant for $d \geq 1$.) This symmetry is violated as well by the nonlinear λ_4 term. Furthermore, in the absence of the λ_4 term, the nonlinearities implicit in Eq. (4) are all relevant in $d=1$ and marginal in $d=2$ so that the scaling behavior of the surface diffusion dominated regime is in fact not obvious.

The second important difference from Eq. (4) is that the flux \mathcal{F} has not been specified. On sufficiently large length and time scales, the flux \mathcal{F} *cannot* be modeled by a noise η with correlations as in Eq. (2), as a result of the physical process of "sticking on the sides" of surface features. This process generates overhangs and voids in the film, violating the volume conservation embodied in the surface diffusion dynamics of Eq. (1). If overhangs are significant, then the scaling properties of the interface are expected to be those of the KPZ equation. What has not yet been adequately addressed is the question of *when* overhangs are important and the physics of the crossover [8].

Given the above theoretical reservations regarding Eq. (1), we introduce a model to investigate these issues simulationally. To motivate our choice of model, let us start with a discussion of the models of Das Sarma and Tamborenea [9] and Wolf and Villain [3], which we shall refer to as the DT-WV model. In this model particles are dropped at a random value of x_{\parallel} onto the top of that column so that the height at x_{\parallel} is increased by 1. At this point the just-dropped particle is relaxed, moving to the top of a neighboring column if it increases the particle's coordination. In $d=1$ the width of the surface grows as t^{β} , $\beta=0.365$ for $t \ll L^2$, close to the value $\beta = \frac{1}{3}$ of the linear version of Eq. (1). Similarly, the width saturated at long times at a value proportional to L^{α} , with α close to the $\alpha = \frac{3}{2}$ of the linear theory. This model suffers from two defects which render it unsuitable for addressing the full range of MBE dynamics. First, the particles drop directly onto the top of a column, bypassing an arbitrary number of particles in neighboring taller columns on the way down. This "drop-through" rule is in contrast to the drop rule of the ballistic aggregation model [10], where the dropped particle sticks as soon as it encounters any particle of the aggregate. We emphasize that this sticking on the side is physically correct and would be present in any experiment or realistic (molecular dynamics, say) simulation [11] with finite-range forces. The second defect is that relaxation is allowed to occur between nearest-neighbor columns regardless of the difference in heights involved. This corresponds to an extremely anisotropic and unphysical diffusion, where diffusion is finite in the directions parallel to the substrate but infinite in the transverse direction. Note that both of these defects arise from the solid-on-solid (SOS) restriction which is at the heart of the DT-WV model. Whereas the SOS constraint is appropriate in some contexts, including some models of KPZ-type growth [12], in the present case it does not serve our purposes, since it enforces a symmetry (volume conservation, on average) not present in the physical situation and we are interested in probing the dynamical violation of this symmetry.

We now turn to a model of ideal MBE growth in which both surface diffusion and deposition are treated physically. In this model, the drop-through rule is replaced by that of ballistic aggregation, where the dropped particle sticks upon first encountering the aggregate, either below or to one side. We also adopt a more physical surface diffusion rule, wherein a randomly chosen surface (not fully coordinated) particle moves to a new position chosen from among the best-coordinated sites in a hypercubical box with sides of length $2L_D + 1$ (in all $d+1$ dimensions) centered on the particle's current position. The diffusivity is governed by L_D and D , the number of diffusion steps a surface particle performs on average in the time it takes to lay down a monolayer, and scales for small D as $L_D D^{1/2}$. This surface diffusion is not to be confused with height diffusion, which has also been studied [13] in the

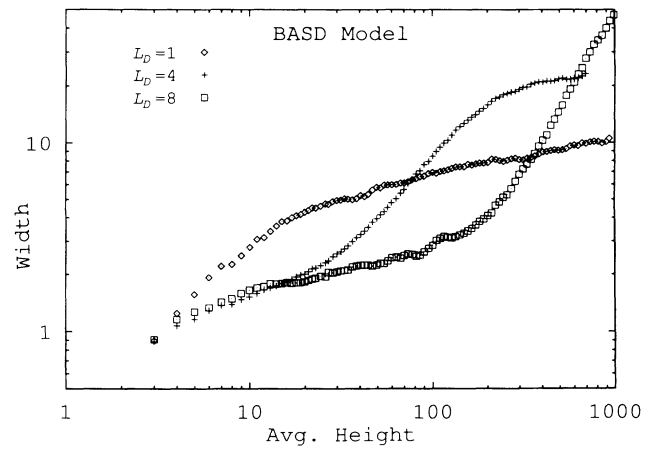


FIG. 1. Width vs average height for the BASD model, averaged over nine runs, with varying $L_D=1, 4,$ and 8 . Other parameters are $L=500, D=5$.

context of ballistic aggregation drop rules. We claim that this new model, ballistic aggregation with surface diffusion (BASD), is a physically correct analog of ideal MBE growth and should correctly describe the statistical properties of surfaces formed in this manner. Furthermore, while other more realistic models involving activated hopping [14] could in principle elucidate the issues posed, their computational complexity makes them unsuitable for a detailed study of scaling properties.

The results of simulation of our BASD model in $1+1$ dimensions for varying L_D are presented in Fig. 1. After a short transient, there is a regime, lengthening with L_D , where w grows with time, apparently as t^{β} , with $\beta \approx 0.25$ for the larger L_D . Then there follows a period of rapid growth of the width, after which power-law growth again sets in. This latter regime then terminates in a saturation regime. The aggregate in the first power-law regime is almost perfectly dense, with essentially no defects or overhangs. After the crossover, on the other hand, the aggregate resembles a coarsened version of a standard ballistic aggregate. We thus interpret the first power-law regime as representing surface diffusion scaling, whereas the second power-law regime and subsequent saturation are described by the KPZ equation.

One interesting and unexpected feature to note in Fig. 1 is the sharp rise in width separating the two regimes. This is in contrast to the results of an exactly solvable linear model with both $\nabla^2 h$ and $\nabla^4 h$ terms, where the crossover is much smoother. We have performed a data collapse of this crossover, rescaling width and time (or equivalently average height) to overlay the data for various L_D , presented in Fig. 2. This collapse indicates that as long as L_D is larger than a few lattice units, the crossover is governed only by L_D . While our values of L_D are not yet sufficiently large to determine the asymptotic scaling of the crossover time, t_c with L_D , it roughly behaves

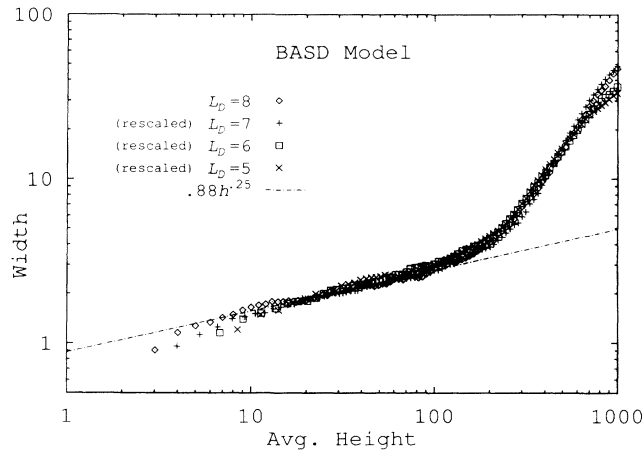


FIG. 2. Collapse of scaled width vs scaled height for the BASD model, averaged over nine runs, for $L_D = 5, 6, 7,$ and 8 . Other parameters are $L = 500, D = 5$.

as $t_c \sim L_D^2$ in the range studied.

Another surprise is the value of β characterizing the surface diffusion regime. The data collapse in Fig. 2 would seem to indicate that β is approaching a value of ≈ 0.25 with increasing L_D . While we would not expect $\beta = \frac{1}{3}$, since λ_4 is zero, it would appear that we are not seeing the $\beta = \frac{3}{8}$ characteristic of the linear version of Eq. (1) either. In fact, the apparent short-time exponent of the DT-WV model itself seems to vary strongly with L_D . This is illustrated in Fig. 3, where the effective $\beta \equiv \partial \log w / \partial \log h$, obtained from an average over 1000 runs, is plotted versus time for the DT-WV model with $L_D = 1$ and 2 . Such an effect is seen in the data of Ref. [9] [see Fig. 3(b) therein] though it was not commented on there. A value of β of 0.25 would seem to indicate that the Edwards-Wilkinson-equation (the $\lambda_2 = 0$) version of the KPZ equation is relevant here. It is hard to understand why a $\nabla^2 h$ would be generated, since the chemical potential driving surface diffusion should be independent of h . A possible explanation might be that lattice-pinning effects [15] are responsible. Clearly this issue bears further investigation.

The above implies that Eq. (1) is not an appropriate description of the physics. On short length scales, the governing dynamics is as yet unclear. On length scales larger than a diffusion length, the physics is that of the KPZ equation. This is so despite the absence of desorption. It is important to understand the origin of the terms in the continuum KPZ description of our BASD model. They do not arise from any explicit gravitational force or tendency for aggregated particles to move downhill. Rather, they arise from sticking on the sides leading to an \dot{h} which depends on the local slope, since the larger the slope, the higher up the next particle will stick. This produces the nonlinear $(\nabla h)^2$ term in Eq. (3). Upon coarse graining this term gives rise to a $\nabla^2 h$ term, as shown by

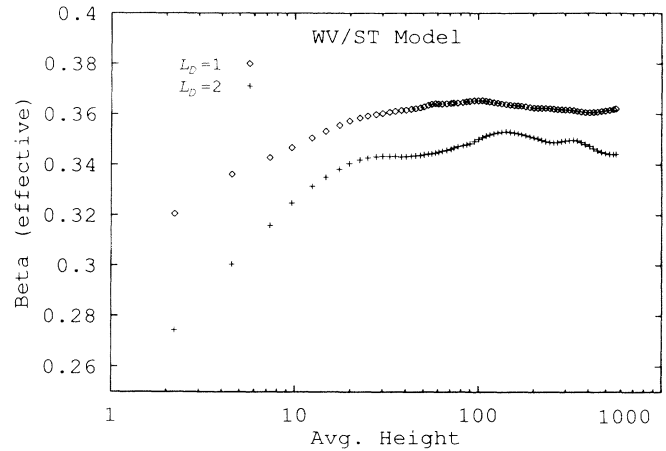


FIG. 3. Effective β vs average height for the WV-ST model, averaged over 1000 runs for $L_D = 1, 2; L = 1000$.

renormalization-group studies [5]. Note that even if our speculation of the generation of a $\nabla^2 h$ term in the surface diffusion regime is valid, the KPZ nonlinearity is forbidden by the $h \rightarrow -h$ symmetry.

A final point is the porosity of the films produced by the BASD model, which is not typical of real MBE films. It is important to note that growth conditions in the laboratory are carefully chosen to produce "good" films. For example, films are typically grown at high temperatures where diffusion lengths are of the order of microns. This is consistent with the initial compact nature of the SD regime. Furthermore, the width in the SD regime grows surprisingly slowly (and if the Edwards-Wilkinson-type scaling is maintained in $2+1$ dimensions, the width would in fact only grow logarithmically). Also, interrupted growth is frequently employed to improve the film quality. Low-temperature films do become porous if grown too thick [16]. In fact, the sharp experimental distinction between "good" and "bad" films is consistent with the violent nature of the crossover.

In conclusion, we have seen how a physically reasonable model for ideal MBE growth leads to a crossover from SD to KPZ scaling on a time scale which grows with diffusion length. Furthermore, the scaling in the surface diffusion regime is not in accord with the predictions of the previously proposed continuum theory. We maintain that similar results would obtain from a study of any physically correct model, including more realistic Monte Carlo or molecular-dynamics simulations. We believe further study of our BASD model, especially of the $(2+1)$ -dimensional case, will prove fruitful.

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