Renormalization Group Study of a Driven Continuum Model for Molecular Beam Epitaxy

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We use the dynamic renormalization group technique to study a continuum model for molecular beam epitaxy for both one- and two-dimensional substrates. Relaxation of the growing film is due to surface tension and surface diffusion. In $1+1$ dimensions we find a purely diffusive strong-coupling fixed point with a dynamic exponent z different from that given by the linear theory as well as the Edwards-Wilkinson fixed point and a fixed point corresponding to unstable growth. In $2+1$ dimensions the purely diffusive fixed point is absent at the one loop order but the other two fixed points are still present.

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Deposition processes in which surface diffusion constitutes the dominant relaxation mechanism have been studied intensively during the last five years [1–13]. These studies were motivated by a desire to model technologically important growth processes such as molecular beam epitaxy (MBE) and to explore the notion of universality in nonequilibrium growth processes. In this context, an interesting class of models are solid-on-solid models driven by a flux of particles. If evaporation of particles is forbidden, a continuum description of these models begins with an equation of continuity

$$\frac{\partial h}{\partial t} + \boldsymbol{\nabla} \cdot \mathbf{j} = v_0 + \eta \ , \qquad (1)$$

since the absence of voids or overhangs allows a description of the surface in terms of a single-valued function $h(\mathbf{x}, t)$ and the absence of evaporation insures that surface rearrangement conserves volume. In Eq. (1) v_0 represents the average growth velocity and $\eta(\mathbf{x}, \mathbf{t})$ represents Gaussian white noise with $\langle \eta(\mathbf{x}, t) \rangle = 0$, and $\langle \eta(\mathbf{x}, t)\ \eta(\mathbf{x}', t') \rangle = 2D\ \delta^d(\mathbf{x} - \mathbf{x}')\ \delta(t - t')$. An important question is how the current $\mathbf{j}(\mathbf{x}, t)$ depends on the function $h(\mathbf{x}, t)$.

In this Letter we consider a particular model for the surface diffusion current [1,7,10] and study the Langevin equation

$$\frac{\partial h}{\partial t} = \nu \nabla \cdot \frac{\nabla h}{\sqrt{g(\nabla h)}} - \kappa \sqrt{g(\nabla h)}\ \tilde{\nabla}^2\ \nabla \cdot \frac{\nabla h}{\sqrt{g(\nabla h)}} + \eta(\mathbf{x}, t) \ , \qquad (2)$$

where the Laplace-Beltrami operator

$$\tilde{\nabla}^2 = \frac{1}{\sqrt{g(\nabla h)}} \nabla_i \left[\sqrt{g(\nabla h)} \left(\delta_{ij} - \frac{\nabla_i h\ \nabla_j h}{g(\nabla h)} \right) \right] \nabla_j$$

enters because the diffusion current is parallel to the interface rather than to the substrate. Here the function $g(x) = 1 + x^2$, ∇_i stands for $\partial/\partial x_i$, δ_{ij} is the Kronecker delta, and repeated indices imply summation. The second term in Eq. (2), with a positive coefficient, κ, describes relaxation through surface diffusion and was first derived by Mullins [14]. Finally, the first term in Eq. (2) is a surface tension term with a coefficient ν, which can be positive or negative. The reason for including this term in Eq. (2) is that numerical simulations seem to indicate that it is generically present when the deposition rate is nonzero [10]. The surface tension ν could be due to diffusion bias near step edges [3] and also arises if one takes into account the nonzero size of incoming particles [1]. Analytically, as will be seen below, this term must be included to obtain a consistent description of the long wavelength properties of this model.

The $(1+1)$-dimensional version of Eq. (2) has been studied by Golubović and Karunasiri (GK) [7]. By numerically integrating Eq. (2), GK found that the slope $\partial h/\partial x$ of the interface profile behaves like the order parameter of Ising-like systems in spinodal decomposition. GK also argued that this model can be *approximately* transformed into an equilibrium model possessing a double-well potential, suggesting that this model would behave like Ginzburg-Landau models for second-order transitions. We have performed a systematic renormalization group analysis on Eq. (2) for both $1+1$ and $2+1$ dimensions. This is an involved calculation, and in this Letter we present mainly the results. The technical details will be given elsewhere [15].

We first discuss some general properties of the model. Clearly, Eq. (2) is invariant under the transformation $h(\mathbf{x}, t) \to -h(\mathbf{x}, t)$ even though the particle beam breaks the up-down symmetry. Another interesting property of Eq. (2) is that it is considerably more complicated in $2+1$ dimensions than in $1+1$ dimensions. This can be seen from the structure of the Laplace-Beltrami operator, i.e., in $2+1$ dimensions, it contains cross terms $(i \neq j)$ that do not appear in $1+1$ dimensions. Following GK's notation of $\mathbf{m} = \nabla h$, we rewrite Eq. (2) in the form

$$\frac{\partial m_i}{\partial t} = \nabla_{ij}^2 \left\{ \nu \frac{m_j}{\sqrt{g(m)}} - \kappa \left[\sqrt{g(m)} \left(\delta_{jk} - \frac{m_j m_k}{g(m)} \right) \right] \times \nabla_{kl}^2 \frac{m_l}{\sqrt{g(m)}} \right\} + \nabla_i \eta(\mathbf{x}, t), \qquad (3)$$

where the function g is given above. Note that we have

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an extra symmetry: $\nabla_i m_j = \nabla_j m_i$. Obviously, expanding the right-hand side of Eq. (3) in powers of m, one has two linear terms, i.e., a Laplace term $\nu\nabla^2 m_i$ and a Laplace square term $-\kappa\nabla^4 m_i$, arising from surface tension and surface diffusion, respectively. For $\nu > 0$, power counting shows that all nonlinear terms are irrelevant to the long wavelength behavior of the model. In this case, Eq. (3) reduces to the well-known Edwards-Wilkinson (EW) equation [16] and the scale invariant solution can be easily obtained for this linear equation. On the other hand, when ν is negative, the system is linearly unstable for wave vectors q in the range $0 < q < q_c = \sqrt{-\nu/\kappa}$. Then both the other linear term $-\kappa\nabla^4 m_i$ and the higher order terms in the series become important for determining the long wavelength properties of the system. Finally, if $\nu = 0$, all nonlinear terms arising from surface diffusion have critical dimension $d_c = 2$ with respect to the Laplace square term $-\kappa\nabla^4 m_i$, indicating that all nonlinear terms in the power series are relevant (marginal) for $d < 2$ ($d = 2$). Therefore, the long wavelength properties of the system depend crucially on the Laplacian term. Now a natural question arises: Is this Laplacian term generated by the nonlinear terms in the large length scale limit and, if so, what is the sign of the coefficient ν for this model?

To answer this question, we have applied the powerful dynamic renormalization group (DRG) theory to Eq. (3). However, as mentioned above, all nonlinear terms are relevant in the physically interesting dimensions and a renormalization group analysis would seem to be a formidable task. We find that the calculation can be truncated at the one loop order and the results turn out to be consistent. In other words, the renormalization perturbation expansion can be controlled by the powers of the leading nonlinear coupling parameters; the coefficients of higher order nonlinear terms are higher powers of the leading nonlinear couplings and can be consistently ignored to the required order. We now proceed to sketch this renormalization group analysis. Since the structure of the equations is different for different dimensions, the traditional ϵ-expansion method cannot be applied here and the renormalization group analyses must be performed individually for each dimension.

We consider first 1 + 1 dimensions. Set $d = 1$ and expand the right-hand side of Eq. (3) in powers of m to obtain

$$\frac{\partial m}{\partial t} = \nu\frac{\partial^2 m}{\partial x^2} - \kappa\frac{\partial^4 m}{\partial x^4} + \frac{\partial^2}{\partial x^2}\left[-u_0 m^3 + u_1 m^2\frac{\partial^2 m}{\partial x^2} + u_2 m\left(\frac{\partial m}{\partial x}\right)^2 + \cdots\right] + \frac{\partial}{\partial x}\eta(x,t)\,, \tag{4}$$

where u_0, u_1, u_2 are nonlinear parameters whose bare values are $\nu/2$, 2κ, 3κ, respectively. As Eq. (4) stands, the leading nonlinear terms are cubic terms and the conserved lateral driving force proportional to $\partial^2 m^2/\partial x^2$ [9,17] does not appear. As has been pointed out previously [10], this is a consequence of the assumption that deposition does not change the nature of the diffusion process; i.e., it is still driven by energy differences and can be described using a surface Hamiltonian. If only the lowest order nonlinear terms are taken into account, the structure of Eq. (4) is quite similar to that of the Langevin equation describing dynamic critical phenomena. This similarity implies that the diffusive system will have an instability similar to a second-order transition. Therefore, a standard strategy used in the discussion of dynamic critical phenomena [18] can be directly applied to study Eq. (3). Namely, regarding ν and u_α ($\alpha = 0, 1, 2$) as the same order perturbation parameters, we determine the fixed point values ν^* and u^*_α to leading order.

Because of the complexity of the nonlinear terms, it is convenient to use Nozières and Gallet's (NG) dynamic renormalization [19] technique. Generally, the interface height function $h(\mathbf{x}, t)$ is a functional of the noise $\eta(\mathbf{x}, t)$. Therefore, according to NG, the coarse graining procedure of the DRG can be realized by performing the following steps. First, we split the noise $\eta(\mathbf{x}, t)$ into two parts $\eta(\mathbf{x}, t) = \bar{\eta}(\mathbf{x}, t) + \delta\eta(\mathbf{x}, t)$ so that $\bar{\eta}$ and $\delta\eta$ are statistically independent. Next, we perform a partial average over $\delta\eta$ on the height function h and define $\bar{h}(\mathbf{x}, t) = \langle h[\bar{\eta}(\mathbf{x}, t) + \delta\eta(\mathbf{x}, t)]\rangle_{\delta\eta}$, $h(\mathbf{x}, t) = \bar{h}(\mathbf{x}, t) + \delta h(\mathbf{x}, t)$. As a consequence, the original equation is decoupled into two equations of motion for $\bar{h}$ and δh, respectively. Finally, we solve the equation of motion for δh formally and substitute this formal solution into the equation of motion for $\bar{h}$. Because of the nonlinear terms, the parameters in the equation of motion for $\bar{h}$ are renormalized.

Applying the above procedure to Eq. (4) with only the cubic terms included we find that, to order $O(u^2)$, no new terms of different form are generated. This means that our original parameter space $\{\nu, \kappa, D, u_\alpha, \alpha = 0, 1, 2\}$ is sufficient to this order. In other words, the higher order terms are of order $O(u^3)$ and can be consistently ignored. Including all terms to leading order and using the traditional rescaling of $x' = xe^{-\delta l}$, $t' = te^{-z\delta l}$, and $h' = he^{-\chi\delta l}$, on taking the limit $\delta l \to 0$, we find the following flow equations:

$$\frac{d\ln u_\alpha}{dl} = z + 2\chi - 6 + 2\delta_{\alpha 0} + \frac{\triangle_\alpha}{u_\alpha}, \quad \alpha = 0, 1, 2\,, \tag{5}$$

$$\frac{d\ln\nu}{dl} = z - 2 - 3\kappa\frac{\bar{u}_0}{\nu}, \tag{6}$$

$$\frac{d\ln\kappa}{dl} = z - 4 - \bar{u}_1, \tag{7}$$

$$\frac{d\ln D}{dl} = z - 2\chi - 1, \tag{8}$$

where $\bar{u}_\alpha$ are reduced perturbation parameters $\bar{u}_\alpha = u_\alpha D/\pi\kappa^2$ and $\triangle_\alpha$, $\alpha = 0, 1, 2$, are given by

$$\triangle_0 = 3u_0(3\bar{u}_0 + \bar{u}_1) + 6u_1\bar{u}_0 - 3u_2\bar{u}_0 \ ,$$

$$\triangle_1 = 3u_0(3\bar{u}_0 - \tfrac{1}{2}\bar{u}_1 + 3\bar{u}_2) + u_1(12\bar{u}_0 + \bar{u}_1 + 6\bar{u}_2) + u_2(-\tfrac{3}{2}\bar{u}_0 + \bar{u}_1 - 3\bar{u}_2) \ ,$$

$$\triangle_2 = 3u_0(3\bar{u}_0 - \tfrac{5}{2}\bar{u}_1 + 4\bar{u}_2) + u_1(9\bar{u}_0 - 4\bar{u}_1 + 8\bar{u}_2) + u_2(\tfrac{9}{2}\bar{u}_0 + 5\bar{u}_1 - 4\bar{u}_2) \ .$$

From the flow equations, we have a renormalization picture to $O(u)$ for the $(1+1)$-dimensional system. The nonlinear couplings, due both to surface tension and surface diffusion, renormalize each other and ultimately determine the fixed-point value of ν^*. The parameter κ is renormalized but its fixed-point value cannot be determined at the present order. This is a common feature of the renormalization of cubic nonlinear terms [18]. This does not, however, affect our discussion of the linear stability of the system as long as we assume that the positive sign of the bare κ is not changed by the DRG transformation [20]. The noise strength D is not renormalized in the one loop calculation. From Eq. (6), the fixed-point value of the surface tension ν^* is proportional to $\bar{u}_0^*$. Therefore, the values of $\bar{u}_0^*$ determine the stability of the system in the long wavelength limit.

To find the fixed points it is convenient to consider reduced flow equations for $\bar{u}_\alpha, \alpha = 0,1,2$. Taking derivatives with respect to l on both sides of $\ln \bar{u}_\alpha = \ln u_\alpha + \ln D - 2\ln\kappa$ and using Eqs. (5)–(8), we readily arrive at

$$\frac{d\ln \bar{u}_\alpha}{dl} = 1 + 2\delta_{\alpha 0} + 2\bar{u}_1 + \frac{\triangle_\alpha}{u_\alpha} \ , \ \alpha = 0,1,2. \qquad (9)$$

It is easy to check that there are three strong-coupling fixed points of Eq. (9): (I) $\bar{u}_0^* = 0$, $\bar{u}_1^* = \bar{u}_2^* = -\frac{1}{7}$; (II) $\bar{u}_0^* = 0.2397$, $\bar{u}_1^* = \bar{u}_2^* = -0.6447$; and (III) $\bar{u}_0^* = -1.0633$, $\bar{u}_1^* = \bar{u}_2^* = 0.8212$. The fixed point (I) is stable with respect to the Gaussian fixed point and controls the scaling behavior of purely diffusive (but nonlinear) relaxation. At this fixed point the dynamic exponent $z = 4 - \frac{1}{7} \approx 3.867$ which may be compared to the value of 3.6 ∓ 0.3 found in simulations of the driven discrete Gaussian model [10]. The fixed point (II) with positive ν^* is in fact unstable with respect to the Edwards-Wilkinson fixed point at which all nonlinear terms are irrelevant in the long wavelength limit. On the other hand, at fixed point (III), one has a negative ν^*, and the system is linearly unstable. Under this circumstance, a finite system will eventually evolve into a steady state with large slopes. The precise morphology may depend on details such as boundary conditions. This fixed point is stable and has a rather large basin of attraction which is bounded by the $\bar{u}_0 = \nu = 0$ axis. The renormalization group flow diagram is shown schematically in Fig. 1 in the $(\bar{u}_0, u_{12})$ plane, where $u_{12} = (\bar{u}_1 + \bar{u}_2)/2$. At this level of approximation it therefore seems that, although the entire $\nu = 0$ axis is unstable, finite ν is *not* produced by renormalization. Furthermore, since the flow cannot cross the $\nu = 0$ axis, a negative surface tension does not become positive under renormalization nor does a positive become negative. Note that the ratio of the two nonlinear couplings arising from the surface diffusion, $\bar{u}_1/\bar{u}_2$, has changed from the bare value 2/3 to the fixed point value 1, implying that GK's transformation [7] is *exact* at the fixed points. Now we turn to the $(2+1)$-dimensional case, in which the algebra is more involved than for the $(1+1)$-dimensional case. Setting $d = 2$ and expanding the nonlinear terms in power of m, Eq. (3) becomes

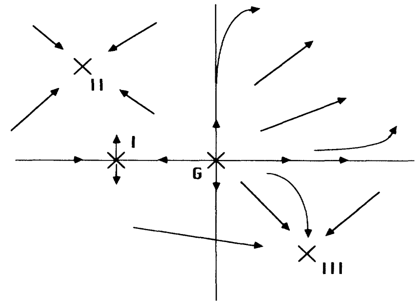

FIG. 1. Schematic renormalization group flow diagram in the $\bar{u}_0$ and u_{12} plane for $d = 1$.

$$\frac{\partial m_i}{\partial t} = \nu\nabla^2 m_i - \kappa\nabla^4 m_i + \nabla_{ij}^2(-u_0 m_j m^2 + u_1 m_j m_k \nabla^2 m_k + u_2 m_k m_l \nabla_{kl}^2 m_j + u_3 m_k \nabla_j m_k \nabla_l m_l + u_4 m_l \nabla_l m_k \nabla_j m_k + \cdots) + \nabla_i \eta(\mathbf{x}, t). \qquad (10)$$

The bare values of the nonlinear couplings are $u_0 = \nu/2$, $u_1 = u_2 = u_3 = \kappa$, and $u_4 = 2\kappa$. Compared to Eq. (4), two more cubic terms appear here due to the added complexity. Therefore, to order $O(u)$, the dimension of the original parameter space is 8, rather than 6, as in the $(1+1)$-dimensional case. Another important difference from the $(1+1)$-dimensional case is that the four cube terms with second-order derivatives are not "complete." That is, there are other terms, for instance, $m^2\nabla^2 m_j$, $m_j \nabla_k m_k \nabla_l m_l$, $m_j \nabla_k m_l \nabla_k m_l$, and so on which do not show up in the original expansion. After the manipulation of the DRG, these terms are generated by the coarse graining procedures. However, a close examination reveals that they are irrelevant at the one loop order. Following exactly the same method outlined above, the flow equations can be obtained without any essential difficulty, after some rather tedious calculations. Treating ν and $u_\alpha, \alpha = 0, ..., 4$ perturbatively, and including all relevant terms to leading order, we have the following flow equations:

$$\frac{d\ln u_\alpha}{dl} = z + 2\chi - 6 + 2\delta_{\alpha 0} + \frac{\triangle_\alpha}{u_\alpha}, \ \alpha = 0,1,2,3,4, \quad (11)$$

$$\frac{d\ln \nu}{dl} = z - 2 - 2\kappa\frac{\bar{u}_0}{\nu}, \qquad (12)$$

$$\frac{d\ln \kappa}{dl} = z - 4 - \frac{1}{2}(\bar{u}_1 + \bar{u}_2), \qquad (13)$$

$$\frac{d\ln D}{dl} = z - 2\chi - 2, \tag{14}$$

where $\bar{u}_\alpha = K_2 u_\alpha D/\kappa^2$ with $K_2 = 1/2\pi$ and the renormalization terms of the nonlinear couplings

$$\triangle_0 = u_0(\tfrac{9}{2}\bar{u}_0 + \tfrac{9}{2}\bar{u}_1 + \tfrac{15}{4}\bar{u}_2 - \tfrac{5}{4}\bar{u}_3 - \tfrac{5}{4}\bar{u}_4)\ ,$$

$$\triangle_1 = u_0(-6\bar{u}_0 - \tfrac{5}{4}\bar{u}_1 - \tfrac{31}{4}\bar{u}_2 + \tfrac{19}{4}\bar{u}_3 + \tfrac{29}{8}\bar{u}_4) + u_1(\tfrac{7}{8}\bar{u}_1 - \tfrac{5}{8}\bar{u}_2 + 2\bar{u}_3 + \tfrac{15}{16}\bar{u}_4) + u_2(-2\bar{u}_2 + \tfrac{17}{8}\bar{u}_3 + \tfrac{7}{8}\bar{u}_4) + u_3(-\tfrac{5}{8}\bar{u}_3 - \tfrac{9}{16}\bar{u}_4) + \tfrac{1}{16}u_4\bar{u}_4\ ,$$

$$\triangle_2 = u_0(-\tfrac{1}{2}\bar{u}_1 + 4\bar{u}_2 + \tfrac{1}{2}\bar{u}_3 + \tfrac{3}{4}\bar{u}_4) + u_1(-\tfrac{1}{4}\bar{u}_1 + \tfrac{1}{2}\bar{u}_2 + \tfrac{1}{2}\bar{u}_3 + \tfrac{5}{8}\bar{u}_4) + u_2(\tfrac{3}{4}\bar{u}_2 + \tfrac{3}{4}\bar{u}_3 + \bar{u}_4) + u_3(-\tfrac{1}{4}\bar{u}_3 - \tfrac{5}{8}\bar{u}_4) - \tfrac{3}{8}u_4\bar{u}_4\ ,$$

$$\triangle_3 = u_0(-\bar{u}_2 + \tfrac{9}{2}\bar{u}_3 + \tfrac{1}{2}\bar{u}_4) + u_1(-\tfrac{1}{2}\bar{u}_2 + \tfrac{7}{4}\bar{u}_3) + u_2(-\bar{u}_2 + 3\bar{u}_3 + \tfrac{3}{8}\bar{u}_4) + u_3(-\tfrac{3}{4}\bar{u}_3 - \tfrac{5}{8}\bar{u}_4) + \tfrac{1}{8}u_4\bar{u}_4\ ,$$

$$\triangle_4 = u_0(-2\bar{u}_0 - 2\bar{u}_1 - \tfrac{5}{4}\bar{u}_2 + \tfrac{5}{4}\bar{u}_3 + \tfrac{21}{4}\bar{u}_4) + u_1(-\tfrac{1}{2}\bar{u}_1 - \tfrac{7}{4}\bar{u}_2 + \tfrac{3}{4}\bar{u}_3 + \tfrac{9}{4}\bar{u}_4) + u_2(-2\bar{u}_2 + \tfrac{7}{4}\bar{u}_3 + \tfrac{15}{4}\bar{u}_4) + u_3(-\tfrac{1}{4}\bar{u}_3 - \bar{u}_4) - \tfrac{3}{4}u_4\bar{u}_4\ .$$

As for the (1 + 1)-dimensional case, we consider the reduced flow equations

$$\frac{d\ln\bar{u}_\alpha}{dl} = 2\delta_{\alpha 0} + \bar{u}_2 + \bar{u}_1 + \frac{\triangle_\alpha}{u_\alpha}\ ,\ \alpha = 0,1,2,3,4\ . \tag{15}$$

The fixed points of Eq. (15) are given by a group of four second-order simultaneous polynomial equations. We have solved this system of simultaneous equations numerically and found one solution with negative $\bar{u}_0^*$, which is $\bar{u}_0^* = -0.1509$, $\bar{u}_1^* = -0.2530$, $\bar{u}_2^* = -0.0108$, $\bar{u}_3^* = 0.0046$, $\bar{u}_4^* = -0.1021$. There are also several other solutions with positive $\bar{u}_0^*$. As argued above, these solutions are superseded by the Edwards-Wilkinson fixed point. An unfortunate feature of the $(2+1)$-dimensional case is the absence of the purely diffusive strong-coupling fixed point (I) although the Gaussian fixed point $\{\bar{u}_\alpha = 0\}$ is unstable. This is due to the fact that for $\nu = 0$ all nonlinear terms, including the ones retained in our expansion, are marginal rather than relevant at the Gaussian fixed point. To find the analog of fixed point (I) one would need to go to higher order in the perturbation expansion.

In conclusion, our dynamic renormalization group analysis of the surface diffusion relaxation system shows that it can evolve into two different morphological states. The first state with a positive surface tension is linearly stable, with the long-distance, late-time behavior of the EW universality class. The second state with a negative surface tension is linearly unstable and probably corresponds to the grooved state observed in numerical simulations [10]. However, in contrast to these discrete models, the drumhead model, driven by an external flux, does not evolve to a state with finite surface tension if the surface tension is initially zero it remains zero under renormalization.

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