Interfacial Coarsening in Epitaxial Growth Models without Slope Selection

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We study the dynamics of an epitaxial growth in the presence of interfacial instabilities that induce a pyramidal or mound-type interfacial growth. We develop a collective coordinates method to *analytically* discuss the coarsening of growing interfaces in the regime when the mounds' slopes increase with time. We calculate the coarsening exponents characterizing the scaling behavior of the mounds' growth. [S0031-9007(96)01988-6]

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There is significant recent interest in the large-scale morphology of growing interfaces relaxing dominantly by surface diffusion, as in the molecular beam epitaxy (MBE) growth [1–9]. Recent studies of MBE grown films document the presence of a striking "tilting" instability forcing the interfaces to develop a *nonzero average slope* rather than to stay parallel to the substrate, i.e., perpendicular to the molecular beam [1–3]. This tilting of the interface manifests itself through the formation of growing pyramidal objects or mounds whose sides have a nonzero slope. The growth of these objects apparently dominates statistical properties of the growing interface—its coarsening and roughness.

Here we study the dynamics of an MBE growth in the presence of the interfacial instabilities inducing mound-type interfacial growth. We provide, for the first time, an *analytic* framework for the physical understanding of already numerous and challenging raw data obtained from numerical simulations and experiments. To address these problems, we introduce here a novel collective coordinates method inspired by modern theories of phase ordering [10]. We use it to discuss the statistics and calculate coarsening exponents of MBE growing interfaces in the *absence* of slope selection mechanism, when the mounds' slope indefinitely increases with time.

To illustrate our approach, we first consider the behavior of MBE interfaces described by the continuous model discussed by Johnson *et al.* [1,2]. (We focus first on this particular model for clarity and simplicity. We stress, however, that our approach is applicable to other continuous MBE growth models, as discussed later on.) In suitable units, it is of the form

$$\frac{\partial h}{\partial t} = -\nabla \mathbf{J}_{\text{destab}} - \kappa \Delta^2 h \,. \qquad (1)$$

$h(\mathbf{x}, t)$ is the interfacial height (in the frame comoving with the interface); $\mathbf{J}_{\text{destab}}$ is the *destabilizing* surface current produced by the so-called Schwoebel effect [6], of the form [1,2]

$$\mathbf{J}_{\text{destab}} = B \frac{\nabla h}{1 + (\nabla h)^2} \,. \qquad (2)$$

Parameter B in (2) is the measure of the strength of the Schwoebel effect. The second term in (1) is the surface diffusion relaxation discussed by Mullins [11]. The unit used in (2) for $\mathbf{x}$ is the mean distance between island nuclei, whereas the unit for h is the lattice constant. For the interface slopes $|\nabla h| \gg 1$ (i.e., terrace width $\ll$ distance between island nuclei), the current of Johnson *et al.* [1,2], Eq. (2), reduces to the form suggested by Villain [6]

$$\mathbf{J}_{\text{destab}} = B \frac{\nabla h}{(\nabla h)^2} \,. \qquad (3)$$

In the opposite limit, $|\nabla h| \ll 1$, the model (1) can be linearized. Then one easily reveals the instability of the flat interfacial configuration towards formation of a modulated structure. As the amplitude of this modulation grows, the interface eventually crosses over to the regime where (3) applies and the dynamics becomes strongly nonlinear. Our main concerns here are the statistical properties of interfaces in this *strongly nonlinear regime.* The model (1) can be put in the type-*A* dynamics form

$$\frac{\partial h}{\partial t} = -\frac{\delta F}{\delta h} \,, \qquad (4)$$

where F is an effective free energy of the form [for a d-dimensional interface]

$$F = \int d^d x \left(-\frac{B}{2} \ln[1 + (\nabla h)^2] + \frac{\kappa}{2} (\nabla \nabla h)^2 \right). \qquad (5)$$

F in (5) is like a Ginzburg-Landau (GL) model with the local slope $\mathbf{M} = \nabla h$ as an *order parameter*. In detail, our approach crucially depends on the possibility to express the dynamics in the type-*A* form (4). The logarithmic term in (5), which generates the destabilizing current (2), is the analog of the "local potential" term of a GL model favoring a phase with a nonzero $\langle \nabla h \rangle$. In the present case, however, this local potential is unbounded from below, reflecting the fact that the order parameter $\mathbf{M} = \nabla h$ will grow indefinitely with time. Thus, the dynamical model in Eqs. (1) and (2) is characterized by the absence of a preferred slope, i.e., there is no slope selection. We

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remark that there is no noise term included in (1). Noise is (generally) known to be irrelevant for the ordering process [10].

A typical configuration of a growing interface governed by (1) is characterized by a growing interfacial width $H(t)$ (typical mound height) and a growing coarsening length scale $L(t)$ (typical mound lateral size). In the absence of a slope selection, the typical mound slope $M(t) = H(t)/L(t)$ will also grow rather than approach a preferred value. Here we discuss the laws governing growths of these quantities. Our approach is similar in spirit to recent theories dealing with phase ordering processes such as the spinodal decomposition [10]. Problems of the type considered here are similar to phase ordering processes with the order parameter $\mathbf{M} = \nabla h$ [7–9]. In general, we thus expect that $\langle \mathbf{M}(\mathbf{x} + \mathbf{r}, t)\mathbf{M}(\mathbf{x}, t)\rangle = M^2(t) f(r/L(t))$, or, equivalently,

$$\langle h(\mathbf{x} + \mathbf{r}, t) h(\mathbf{x}, t)\rangle = H^2(t)\phi\left(\frac{r}{L(t)}\right), \qquad (6)$$

where f and ϕ are some structure functions characterizing the phase ordering process. In terms of the Fourier transform of $h(\mathbf{x}, t)$, Eq. (6) reads

$$\langle h_{\mathbf{k}}(t) h_{-\mathbf{k}}(t)\rangle = H^2(t) L^d(t) g(kL(t)), \qquad (7)$$

with g, the Fourier transform of ϕ in (6), and $k = |\mathbf{k}|$. Equation (7) is the equal-time version of

$$\langle h_{\mathbf{k}}(t) h_{-\mathbf{k}}(t')\rangle = H(t)H(t')k^{-d} G(kL(t), kL(t')), \qquad (8)$$

with $g(p) = p^{-d} G(p, p)$. We assume that $L(t)$ and $H(t)$ are the only long length scales characterizing the interface. Thus, in particular, the probability distribution of the local slope, ∇h, must have the form

$$P(\nabla h) = \frac{1}{M(t)}\,\psi\left(\frac{|\nabla h|}{M(t)}\right), \qquad (9)$$

with $\int dy\, \psi(y) = 1$ and $M(t) = H(t)/L(t)$.

In the following, we discuss the model (1) in the strongly nonlinear regime with $|\nabla h| \sim H/L \gg 1$, when the destabilizing current assumes the form in Eq. (3) and the local potential term in (5) reduces to $-B\ln(|\nabla h|)$. By Eq. (1), one can easily verify that

$$\frac{d\langle h^2\rangle}{dt} = 2\langle(\nabla h)\mathbf{J}_{\mathrm{destab}}\rangle - 2\kappa\langle(\Delta h)^2\rangle, \qquad (10)$$

holds exactly. Thus, in the strongly nonlinear regime ($M = H/L \gg 1$), by (3), (7), (9), and (10)

$$c_1 \frac{dH^2}{dt} = 2B - 2c_0\kappa \frac{H^2}{L^4}, \qquad (11)$$

where

$$c_1 = \int \frac{d^d p}{(2\pi)^d}\, g(p),$$

and

$$c_0 = \int \frac{d^d p}{(2\pi)^d}\, p^4 g(p),$$

are numerical constants.

Equation (11) is an ordinary differential equation for $H(t)$. Let us now derive a similar equation for $L(t)$. To this end, we consider the average density $\epsilon = \langle F\rangle/A$ of the effective free energy (5) (with A the substrate area). This density satisfies, by (4), the exact equation

$$\frac{d\epsilon}{dt} = -\int \frac{d^d k}{(2\pi)^d}\,\langle \partial_t h_k(t)\partial_t h_{-k}(t)\rangle,$$

which can be combined with (8) to arrive at

$$-\frac{d\epsilon}{dt} = c_1\left(\frac{dH}{dt}\right)^2 + 2c_2 \frac{dH}{dt}\frac{H}{L}\frac{dL}{dt} + c_3\left(\frac{H}{L}\frac{dL}{dt}\right)^2, \qquad (12)$$

with

$$c_2 = \int \frac{d^d p}{(2\pi)^d}\, p^{1-d}\left(\frac{\partial G(p_1, p_2)}{\partial p_1}\right)_{p_1=p_2=p} = 0;$$

see Ref. [12], and

$$c_3 = \int \frac{d^d p}{(2\pi)^d}\, p^{2-d}\left(\frac{\partial^2 G(p_1, p_2)}{\partial p_1 \partial p_2}\right)_{p_1=p_2=p}.$$

Furthermore, by (5), (7), and (9), we get $\epsilon = -B\ln(H/L) + c_0\kappa H^2/2L^4 + \text{const}$, for $M \gg 1$. This, combined with (12) and (11), yields

$$c_3 \frac{H^2}{L}\frac{dL}{dt} = -B + 2c_0\kappa \frac{H^2}{L^4}. \qquad (13)$$

Equations (11) and (13) are first order in time ordinary differential equations for the evolution of the interface width $H(t)$ and the coarsening length $L(t)$. In the following we use these equations to extract the scaling behavior of the collective coordinates $H(t)$ and $L(t)$ characterizing the interface. To this end, we introduce the dimensionless quantity

$$x = \frac{c_0\kappa}{B}\frac{H^2}{L^4}. \qquad (14)$$

Then, by (11) and (13)

$$c_1 \frac{dH^2}{dt} = 2B(1 - x), \qquad (15)$$

$$\frac{H}{x}\frac{dx}{dH} = \frac{a_1 - a_0 x}{1 - x}, \qquad (16)$$

where $a_1 = 2 + 4c_1/c_3$ and $a_0 = 2 + 8c_1/c_3$. As the energy loss in Eq. (12) must be positive, $c_1 > 0, c_3 > 0$. Thus, $0 < a_1 < a_0$. This inequality restricts possible types of dynamical behaviors implied by (14) to (16) to a *unique* type in which, for large t, x approaches

$x^* = a_1/a_0 (0 < x^* < 1)$, by (16) [13]. So, for a large t, by (14) and (15), $H^2 = (Bx^*/c_0\kappa)L^4 = 2B(1 - x^*)t/c_1$. Thus, $H \sim t^\beta,\ L \sim t^n,\ H \sim L^\omega$, with

$$\beta = \tfrac{1}{2}, \quad n = \tfrac{1}{4}, \quad \omega = \beta/n = 2\,. \tag{17}$$

Thus, the ultimate scaling behavior of the continuous model (1)–(3) is characterized by the exponents in Eq. (17). The interface growth is superlinear, with $\omega = \beta/n > 1$, reflecting the presence of a *growing* interface slope $M(t) = H(t)/L(t) \sim t^\lambda$ with $\lambda = n(\omega - 1) = \beta - n = \frac{1}{4}$: By (6) and (7), the height-height difference $C(r,t) = \{\langle[h(\mathbf{x} + \mathbf{r}, t) - h(\mathbf{x}, t)]^2\rangle\}^{1/2}$, behaves, for $r \ll L$, as

$$C(r,t) = (c_4/d)^{1/2} M(t) r \sim t^\lambda r\,, \tag{18}$$

with

$$c_4 = \int \frac{d^d p}{(2\pi)^d}\, p^2 g(p)\,.$$

Thus, whereas, by (18), the "roughness" exponent α *is* one, the growing slope reflects itself in the presence of the *growing* prefactor

$$M(t) = \text{const} \times B^{1/2}(t/\kappa)^{1/4}. \tag{19}$$

Scaling behavior in Eq. (17) is *superuniversal,* i.e., independent of the interface dimension d, in agreement with numerical data of Hunt *et al.* ($d = 1$) [2], and of Somfai and Sander ($d = 2$) [14]. These numerical works on the *continuous* model (1)–(3) yield exponents which agree with our analytic theory results in Eq. (17). In particular, we find that the interfacial width H grows as a square root of the deposition time. Such a growth of H with $\beta = \frac{1}{2}$ has been observed in numerical simulations of *discrete* growth models by Zhang, Detch, and Metiu [15], and, more recently, by Amar and Family [16], and Smilauer and Vvedensky [17]. Smilauer and Vvedensky find $\beta \approx 0.52$, $n \approx 0.24$, and, thus, $\lambda = \beta - n \approx 0.28$ [17], whereas Amar and Family find $\beta \approx 0.45$, $n \approx 0.16$, and, thus, $\lambda = \beta - n \approx 0.29$ [16], in the no-slope-selection regime (see below). We remark that a growth with $\beta \approx \frac{1}{2}$ has been observed in the experiments of Ernst *et al.* in the homoepitaxy on Cu(100) surface [3], and, more recently, in the experiments of Elliott *et al.* in the homoepitaxy on Ag(111) surface [18].

Our phase ordering type theory of growing interfaces can be easily generalized to discuss surface relaxation different from the Mullins term $-\kappa\Delta^2 h$ in Eq. (1) [19,20]. For example, if $\kappa = 0$ (or, in practice, small), the surface relaxation is dominated by a higher order term $\sim\Delta^m h$, with $m > 2$, as documented by the recent study of Stroscio *et al.* of an MBE growth *with* slope selection [20]. For such a surface relaxation, in the *absence* of slope selection, by a direct generalization of our previous discussion we find

$$\beta = \tfrac{1}{2}, \quad n = 1/2m, \quad \omega = \beta/n = m\,, \tag{20}$$

and $\lambda = \beta - n = (m - 1)/2m$, for any $m > 1$. A notable feature of this result is that $\beta = \frac{1}{2}$ *regardless* of the actual value of m [21]. Thus, the "square-root of time" growth of the interface width is insensitive to important *qualitative* details of the surface diffusion relaxation, such as the value of m. Such a growth is common to systems in which slope selection mechanism effects are weak on the experimental time scale and $\mathbf{J}_{\text{destab}}$ has the Villain's form, Eq. (3). We remark that Amar and Family [16] find $n \approx 1/6$, in the "no-slope-selection" regime. By our Eq. (20), this indicates the presence of a *strong* $\Delta^3 h$ type surface relaxation in their model, $m = 3$. Recent experiments of Stroscio *et a.* [20] indicate that this might be a frequent feature in MBE growth.

In the presence of slope selection, the destabilizing ("uphill") surface current $\mathbf{J}_{\text{destab}}$, Eqs. (2) and (3), is replaced by a net local surface current $\mathbf{J}_{\text{loc}}(\nabla h) = \mathbf{J}_{\text{destab}}(\nabla h) + \mathbf{J}_{\text{downhill}}(\nabla h)$. For $|\nabla h| = M_0 =$ the preferred slope, $\mathbf{J}_{\text{loc}} = 0$, i.e., the downhill current balances the uphill current. For $|\nabla h| \ll M_0$, the uphill current dominates the net current, i.e., $\mathbf{J}_{\text{loc}}(\nabla h) \approx \mathbf{J}_{\text{destab}}(\nabla h)$. Thus, our no-slope-selection scaling behavior, Eq. (17), holds for $t \ll t_c$, where t_c is a crossover time scale at which $M(t) \approx M_0$. So, by (19).

$$t_c = \text{const} \times \frac{\kappa M_0^4}{B^2}\,. \tag{21}$$

For $t \ll t_c$, $H = \text{const}(Bt)^{1/2}$ and $L = \text{const}(\kappa t)^{1/4}$, i.e., one has our no-slope-selection behavior ($\beta = \frac{1}{2}$, $n = \frac{1}{4}$, $\alpha = 1$, and $\lambda = \frac{1}{4}$, for the Mullins-type surface relaxation). For $t \gg t_c$, the slope selection dominates ($M \approx M_0$) and one has a different scaling behavior studied before numerically [9,16], with $\beta_s = n_s \approx 0.25$, $\omega_s = \beta_s/n_s = \alpha_s = 1$, and $\lambda_s = \beta_s - n_s = 0$, reflecting the asymptotic approach to the preferred slope M_0. The crossover time scale t_c, Eq. (21), should, generally, *increase* with increasing temperature.

In our previous discussions, we assumed $\mathbf{J}_{\text{destab}}(\nabla h)$ to be of the Villain's form, Eq. (3). However, our theory can be applied to $\mathbf{J}_{\text{destab}}(\nabla h)$ of a *general* form [22]. From this generalization, we find that $\beta = 0.5$ *only* if $\mathbf{J}_{\text{destab}}(\nabla h)$ has the Villain's form; i.e., Eq. (3) is the *unique* form that can yield $\beta = 0.5$. Moreover, we find that β *not* equal to $\frac{1}{2}$ can occur only if $\mathbf{J}_{\text{destab}}(\nabla h)$ has a form *qualitatively* different from the Villain's form. For the model with a $\Delta^m h$-type surface relaxation, and $\mathbf{J}_{\text{destab}} \sim \nabla h/|\nabla h|^\eta$, *asymptotically* for large $|\nabla h|$, we find that $\beta(\eta, m) = (2m - 2 + \eta)/2m\eta$ for $0 < \eta < 3$, and $\beta = (2m + 1)/6m$ for $\eta > 3$, whereas $n = 1/2m$. Note that, interestingly, *only* for the Villain's type current ($\eta = 2$), the exponent $\beta(\eta = 2, m)$ does *not* depend on m and equals $\frac{1}{2}$.

Cohen *et al.* [23] note that $\beta = \frac{1}{2}$ growth occurs for the case of *infinite* Schwoebel barrier (SB). Our analysis recovers this result, because for a *finite* SB, the step-flow model of Elkinani and Villain [24] yields $\mathbf{J}_{\text{destab}} = B\nabla h/|\nabla h|(M^* + |\nabla h|)$, with $M^* \to 0$, in the limit of infinite SB (here $B = \frac{1}{2}$ flux density). In this limit, this current reduces to the original Villain's form (3). Moreover, even for a *finite* SB, the Villain's form is approached asymptotically, $\mathbf{J}_{\text{destab}} = B\nabla h/|\nabla h|^2$ (i.e., $\eta = 2$) for slopes $|\nabla h| \gg M^*$. According to the above discussion, one thus has, at the longest time, the regime with $\beta = \frac{1}{2}$ even for a system with *finite* SB. During the growth without slope selection, a system with a finite SB will eventually enter the regime with $\beta = \frac{1}{2}$ when $M(t) = H(t)/L(t)$ gets bigger than M^* [25]. In this regime, the deposition of atoms on a terrace *asymptotically* outnumbers the transitions across the Schwoebel barriers. Thus, interface width scales with time in the same way as in a system with infinite SB.

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[12] To see that $c_2 = 0$, note that, as $G(p_1, p_2) = G(p_2, p_1)$, one has

$$c_2 \sim \int_0^\infty dp \left[\left(\frac{\partial G(p_1, p_2)}{\partial p_1} \right)_{p_1=p_2=p} + \left(\frac{\partial G(p_1, p_2)}{\partial p_2} \right)_{p_1=p_2=p} \right] = \int_0^\infty dp \, \frac{d}{dp} [G(p, p)] = 0,$$

as $G(p, p) = p^d g(p) \to 0$ for both $p = 0$ and $p = \infty$.

[13] By integrating (16), $a_1 \ln(H) = \text{const} + \ln(x) - (1 - x^*) \ln|x - x^*|$. This implies $x \to x^*$ as $H \to \infty$.

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[21] For any m bigger than 1, the interface width satisfies an equation of the form of Eq. (15), with an $x = \text{const}\, H^2/L^{2m}$ which saturates to an $x^* < 1$ at long times. Thus, $H \sim \sqrt{t}$ for *any* m, and $L \sim H^{1/m}$, implying Eq. (20).

[22] For a local surface current of the form $\mathbf{J}_{\text{loc}} = \Phi(|\nabla h|)\nabla h$, where $\Phi(|\nabla h|)$ is an *arbitrary* function of $|\nabla h|$, we arrive at equations which have the form of our Eqs. (11) and (13), however, with B replaced by $G(M) = \langle \mathbf{J}_{\text{loc}} \nabla h \rangle = \langle \Phi(|\nabla h|) (\nabla h)^2 \rangle$, or by (9), $G(M) = \int dz\, \Phi(z) z^2 P(z) = M^2 \int dy\, \Phi(My) y^2 \psi(y)$.

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[25] For the slopes $|\nabla h| \ll M^*, \mathbf{J}_{\text{destab}} \sim \nabla h/|\nabla h|$, i.e., the Elkinani-Villain current behaves as an $\eta = 1$ current. So, the $\beta = \frac{1}{2}$ regime is *preceded* by an earlier regime ($M(t) < M^*$), with, according to our theory, $\beta(\eta = 1, m) = 1 - 1/2m = 1 - n$. In the simulations of Ref. [24], such an earlier regime has been observed. It has $\beta \approx 1$, what is in agreement with our result as there is essentially no coarsening (i.e., $n \approx 0$) in the full model of Ref. [24]. Such a growth, with $\beta \approx 1$ and a small n, was observed in the experiments on Si; see D. J. Eaglesham, H. J. Gossman, and M. Cerullo, Phys. Rev. Lett. **65**, 1227 (1990).