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Noise-assisted mound coarsening in epitaxial growth

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Abstract. Two types of mechanisms are proposed for mound coarsening during unstable epitaxial growth: stochastic, due to deposition noise, and deterministic, due to mass currents driven by surface energy differences. Both yield the relation $H = (RWL)^2$ between the typical mound height W, mound size L, and the film thickness H. An analysis of simulations and experimental data shows that the parameter R saturates to a value which discriminates sharply between stochastic ($R \simeq 1$) and deterministic ($R \ll 1$) coarsening. We derive a scaling relation between the coarsening exponent $1/z$ and the mound-height exponent β which, for a saturated mound slope, yields $\beta = 1/z = 1/4$.

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The appearance of large-scale structures ("mounds") during epitaxial growth has attracted considerable attention over the past several years. One reason for this is the widespread use of vapor-phase epitaxy for the fabrication of quantum heterostructures. Morphological control of both the substrate and the epilayers over a micron length scale is required for the reproducibility of such structures, particularly those involving narrow quantum wells. On a more fundamental level, mounds have been observed to form on several types of material and with various deposition techniques. An understanding of the kinetics of mound formation and evolution is therefore essential for a comprehensive description of epitaxial growth.

Mound formation during homoepitaxy can arise from a growth instability [1] due to the Ehrlich–Schwoebel (ES) barrier [2]. This barrier suppresses interlayer mass transport and thereby promotes multi-layer growth that is manifested either as a gentle modulation of the growth front [3] or as an array of pyramid-like structures that form immediately upon the initiation of growth [4]. Studies of this phenomenon by the numerical integration of continuum growth equations [3–5] and by kinetic Monte-Carlo (KMC) simulations of lattice models [3,5–7] show that after the initial instability is well-developed, the growth front evolves into an array of conic or pyramidal structures with a uniform slope (Fig. 1). The mound morphology is found to have a characteristic length scale L which coarsens with the film thickness H as

$$L \sim H^{1/z} \tag{1}$$

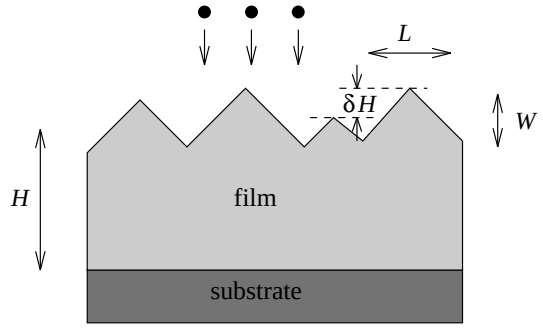

Fig. 1. Schematic mound morphology during unstable epitaxial growth, showing the film thickness H, mound height W, lateral mound size L, and mound height fluctuation δH.

where the coarsening exponent $1/z$ lies in the range 0.15–0.25. The typical mound height W also conforms to a power law,

$$W \sim H^{\beta} \tag{2}$$

where $0.25 \leq \beta \leq 0.5$. The calculated mound morphologies and exponents agree with *some* experimental findings [4,8], but some basic questions, such as why the mounds coarsen, remain unanswered.

In this paper, we propose two mechanisms to explain the coarsening phenomena described above. We show that, under physically reasonable assumptions, deposition noise alone can induce coarsening with scaling exponents consistent with simulational and experimental findings. Surface energetics, previously incorporated in deterministic

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continuum growth equations but whose role has not been clearly identified, also contribute to coarsening and give rise to the same scaling exponents. The relative importance of the two mechanisms is, for a given deposition flux, controlled by the substrate temperature. At sufficiently low temperatures, deposition noise dominates, entailing a universal relationship between the three geometrical parameters H, L, and W that characterize the mound structure (Fig. 1). At high temperatures, this relationship is a function of the substrate temperature, but the mound size distribution is more uniform than at lower temperatures. An analysis of KMC and experimental data confirms the appropriateness of these results and shows that available experiments correspond to the high-temperature regime. We provide estimates of the growth conditions that would enable the noise-dominated coarsening regime to be observed.

The usual picture of coarsening assumes an exchange of atoms between neighboring mounds that depends on local surface configurations [9]. Deposition noise, on the other hand, introduces a new effect: the uncorrelated random arrival of atoms on the surface causes the volume (and, thus, the height) of a mound to fluctuate in time even when inter-mound mass flow is absent. This generates a height fluctuation δH which is uncorrelated from mound to mound. From Figure 1, we see that coarsening occurs when δH becomes comparable to the mound height W, at which point bigger mounds overwhelm their smaller neighbors. This process is irreversible as long as individual mounds are dynamically stable when in isolation [10].

The coarsening time under the noise mechanism can be estimated by considering a one-mound model in which all inter-mound mass transport is neglected. After H layers of material have been deposited, the number of deposited atoms in the column under the mound is given by $N \simeq HL^d/\Omega$, where L is the typical lateral mound size (Fig. 1), d is the dimension of the surface, and Ω is the volume of a single deposited particle. According to the central limit theorem, the fluctuation of N under random deposition is given by

$$\delta N \simeq N^{1/2} \simeq (HL^d/\Omega)^{1/2}. \tag{3}$$

This yields an estimate for the height fluctuation:

$$\delta H \simeq \Omega \delta N/L^d = (\Omega H/L^d)^{1/2}. \tag{4}$$

Coarsening occurs when $\delta H \simeq W$. From equation (4), we obtain a coarsening time (in units of deposited layers) of

$$H \simeq W^2 L^d/\Omega. \tag{5}$$

In the regime where the power laws (1) and (2) are well obeyed, (5) yields:

$$\frac{2\beta}{d} + \frac{1}{z} = \frac{1}{d}\,. \tag{6}$$

If the mound slope $s \simeq W/L$ saturates to a constant, this equation produces

$$\beta = \frac{1}{z} = \frac{1}{d+2}\,. \tag{7}$$

Kawakatsu and Munakata [11] studied a noise-driven coarsening model for $d = 1$, which can be mapped to our problem [12]. In that model, the mound slope saturates to a finite value after an initial transient. From (7), one obtains $z = 3$, which agrees with the analysis of reference [11]. In contrast, a noiseless model for unstable growth yields a much slower coarsening law in $d = 1$ [13].

For $d = 2$, we have checked that (6) is consistent with existing numerical studies, irrespective of whether the mound slope saturates. For models that exhibit slope saturation, several groups [5,12] have concluded that $\beta = 1/z = 1/4$, as suggested by (7). Higher values of β [6] (and, hence, lower values of $1/z$) are obtained if the mound slope continues to increase with the film thickness, in agreement with (6).

The foregoing analysis neglects completely any mass flow between mounds. Interestingly, studies [12,14] of certain deterministic growth equations which incorporate vertical mass transport up and down a mound (see below) have also produced a coarsening exponent of $z = 4$ in $d = 2$. However, no consensus has emerged on whether the coarsening law is sensitive to the form of the growth equation, *e.g.*, the inclusion of crystal anisotropies. The matter is further complicated by the existence of both fast ($z = 2$) and slow ($z = 4$) soft modes in this type of equation, as pointed out recently by Siegert *et al.* [15]. Clearly, a better understanding of the underlying mechanism(s) of coarsening is needed to resolve some of these puzzles.

To identify such a mechanism, we find it useful to distinguish two types of surface mass flow during growth: (i) vertical transport up and down a mound, and (ii) lateral transport around a mound. Recent work by Politi and Villain [13] on a one-dimensional growth model shows that the vertical current produces mounds but essentially no coarsening. The reason is that such currents only stabilize individual mounds but do not discriminate mounds of different size, and hence are not directly involved in coarsening.

In contrast, process (ii) transports mass between mounds in a way which depends on the neighboring mound configuration. To illustrate this idea, we consider the mass flow between two neighboring mounds in a typical late-stage situation [16], as depicted in Figure 2. The central part of each mound consists of approximately concentric rings of steps. The two mounds are joined by a "ridge terrace," the outer rim of which has convex parts on either side and concave parts in the middle. Sites on the concave parts offer on average better lateral bonding, and are therefore energetically more favorable. This effect can be modelled by a chemical potential difference $\Delta\mu$ between the convex and concave regions. Assuming that the steps are in local thermal equilibrium, we may write,

$$\Delta\mu \simeq \gamma_s/L \tag{8}$$

where γ_s is the step free energy per unit length and L is the typical lateral scale in question (*e.g.*, the distance between two centers). This chemical potential difference induces an inward mass current (Fig. 2) of magnitude

$$j_s \simeq D_s \Delta\mu/L \tag{9}$$

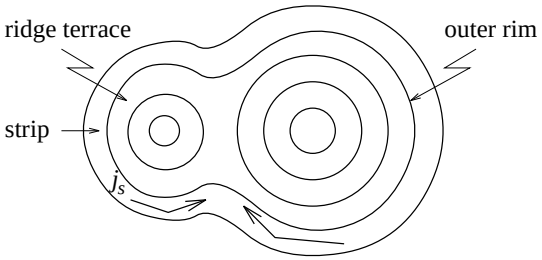

Fig. 2. Top view of two neighboring mounds of unequal size. Better lateral bonding for surface atoms is achieved at the concave regions of the closed steps. This mechanism results in an inward mass current from the outer rim.

where D_s is a kinetic transport coefficient [17]. The same mechanism is also appropriate in other layers below the ridge terrace, though j_s decreases due to the decreasing curvature of the steps. For Q layers with significant inward transport, the total inward mass current is given by

$$J_s \simeq Q j_s \simeq D_s \Delta\mu (Q/L). \tag{10}$$

The process just described yields a gradual outward expansion of the neck region connecting the two mounds. When the amount M of transported mass reaches a value comparable to the volume needed to fill the gap, WL^2, merging is complete. This leads to an estimate of the coarsening time due to lateral transport alone:

$$\tau_s = M/J_s \simeq L^4 \frac{W}{\gamma_s D_s Q} \cdot \tag{11}$$

With $Q \simeq W$, this again yields $z = 4$ [18].

To test these ideas, we have analyzed previously published and newly-generated data from KMC simulations of a solid-on-solid model that describes mound formation [6]. This model includes random deposition, nearest-neighbor hopping at a rate that depends on the local environment of the hopping atom (which thereby allows for bond-breaking), and an ES barrier. Two sets of model parameters are used, denoted by I and II, which describe systems with relatively weak and strong ES barriers, respectively. A detailed discussion of the model, its parameters, and a quantitative evaluation of the resulting morphologies can be found in reference [6].

The two mechanisms proposed above for coarsening can be distinguished by considering the ratio between δH given in (4) and the mound height W,

$$R = \frac{\delta H}{W} = \frac{\Omega^{1/2} H^{1/2}}{W L^{d/2}} \cdot \tag{12}$$

When deposition noise plays the dominant role, we have $R \simeq 1$, independent of the details of surface dynamics. On the other hand, a much smaller R is obtained when the surface-energy driven lateral transport dominates the coarsening process. In both cases, a constant R as a function of film thickness is itself a measure of the $z = 4$ scaling when the mound slope saturates.

The numerical values of R obtained from simulations under different growth conditions are plotted against layer thickness in Figure 3a. The quantities L and W are extracted from the shape of the height-height correlation function, as described in reference [4]. Apart from statistical fluctuations, R evolves to a constant value after an initial transient in all cases, thus confirming the validity of the scaling law (6).

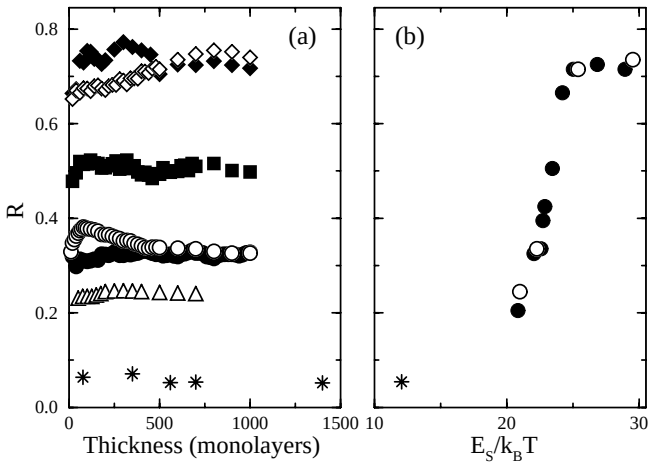

Fig. 3. (a) Data obtained from simulations and experiments for the time-development of R. Filled symbols denote KMC simulations with parameter set I at substrate temperatures $T = 728$ K (♦), 778 K (■), and 828 K (●). Open symbols denote simulations with parameter set II at $T = 350$ K (◇), 400 K (○), and 425 K (△). Experimental data is shown for Fe(001) [8] (⋆). (b) Temperature dependence of the saturated value of R for parameter set I (●), parameter set II (○), and experimental data (* as in (a)), plotted as a function of E_S/k_BT, where E_S is the single-adatom hopping barrier, and k_B is Boltzmann's constant.

Also shown in Figure 3a is the evolution of R obtained from scanning tunneling microscopy (STM) measurements during epitaxy of Fe(001) [8,19]. After a weak initial transient, the corresponding value of R saturates to a small value, indicating the dominance of the deterministic coarsening mechanism (*i.e.*, lateral transport driven by a chemical potential gradient) for the growth conditions employed. However, as we discuss below, we can prescribe growth conditions for this system that should enable stochastic coarsening to be observed.

In Figure 3b, we plot the temperature dependence of the saturated values of R for KMC simulations with parameter sets I and II and for the experimental data in Figure 3a. There are two important features of this diagram. Most apparent is the precipitous drop of R as the temperature is increased. This behavior can be understood by observing that the kinetic coefficient D_s in (9), which is associated with activated adatom hopping, is a strong function of temperature. Hence, the drop in R can be interpreted as a sharp crossover from the stochastic regime at low temperatures to the deterministic regime at high temperatures.

But a potentially more useful feature of Figure 3b is that by plotting the saturated values R against E_S/k_BT, where E_S is the adatom hopping barrier for the particular

system, the data collapse onto a single "universal" curve. This suggests that in our model, the lateral transport is controlled by terrace diffusion of single adatoms, $D_s \simeq k_0 \exp(-E_S/k_BT)$, where $k_0 \simeq 10^{13}$ s^{-1} [6]. Then, in view of the comments in the preceding paragraph, it may not be too surprising that the quantity E_S/k_BT emerges as an important parameter [20]. Assuming the same situation holds in the experiment [21] we estimate the temperature range over which noise-assisted coarsening can be observed for Fe(001) (at the growth rate used in Ref. [8]) as $T \approx$ 200 K and lower.

The main conclusion of our study is that both deposition noise and surface mass transport driven by bonding energy differences contribute to mound coarsening. The picture we have developed leads to an important geometrical relation between the parameters L and W characterizing the mound morphology, and the film thickness H, with a proportionality constant bounded from above by unity. A time-independent value of R implies a scaling relation between the mound coarsening exponent $1/z$ and the mound height exponent β, which agrees with previous work. KMC simulations and available experimental data do indeed show that R saturates during growth and, moreover, reveals that there is a clear discrimination between the low-temperature stochastic and the high-temperature deterministic mound coarsening regimes. The apparent universality of saturated R values, when appropriately plotted, permits the identification of growth conditions where stochastic coarsening can be observed.

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16. By "late-stage", we have in mind a situation where the surface has acquired a quasistationary profile, *i.e.*, after depositing exactly one more monolayer of atoms, the surface, on average, looks "almost the same" as before.
17. Two kinetic pathways contribute to j_s: diffusion along the step and detachment of atoms from the step to the terrace followed by diffusion on the terrace. In both cases the transport is essentially one-dimensional process, so D_s does not depend appreciably on L, but it may depend on the strip width (which is inversely proportional to the mound slope) when the second process dominates.
18. That both mechanisms yield $z = 4$ is a peculiar feature at $d = 2$. Note that, in the presence of other mounds, there are competing tendencies for the surface mass flow, which may lead to an increase of τ_s estimated in (11).
19. R. Koch, personal communication.
20. Obviously the barrier to detachment of adatoms from step edges must be important as well. In our simulations, this barrier is of the similar magnitude for the both parameter sets, so its effect could not be established.
21. We assumed E_S equal to 0.45 eV for Fe(001) [J.A. Stroscio, D.T. Pierce, R.A. Dragoset, Phys. Rev. Lett. **70**, 3615 (1993)].