Intermediate scaling regime for multilayer epitaxial growth

Richard S. Ross and Mark F. Gyure
HRL Laboratories, LLC, 3011 Malibu Canyon Road, Malibu, California 90265
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We explore the layer-by-layer (Frank–van der Merwe) growth regime within the context of a discrete solid-on-solid kinetic Monte Carlo model. Our results demonstrate a nontrivial scaling of the lattice step edge density, a quantity that oscillates about a nominally constant value prior to the onset of kinetic roughening. This value varies with the ratio of the surface diffusivity to the deposition flux, $R \equiv D/F$, as a nearly perfect power law over a wide range of R. This ''intermediate'' scaling regime extends in coverage from one to at least a few tens of monolayers, which is exactly the regime of most importance to the growth of device-quality semiconductor quantum heterostructures. Comparison with lowest-order linear theories for height fluctuations demonstrates the validity of the Wolf-Villain mean-field theory for the description of lattice step density and ''in-plane'' structure for all coverages down to the first monolayer of growth. However, the mean-field theory does not fully account for the surface width in this regime and consequently does not quantitatively predict the observed step density scaling.

Layer-by-layer (or Frank–van der Merwe) growth is by far the growth mode of choice for the production of device-quality semiconductor material by molecular beam epitaxy (MBE). This growth mode produces exceptionally high-quality epitaxial material with the extremely low defect densities required for most electronic and optoelectronic device applications. The layer-by-layer mode offers other practical advantages that make it desirable for device growth. Oscillations in the specular intensity of reflection high-energy electron diffraction (RHEED) patterns are often observed in this regime[1] and have been correlated both theoretically[2] and experimentally[3] to the instantaneous density of step edges on the surface. Oscillations in threshold photoemission[4,5] signals have also been observed in this regime and may also be correlated to surface steps.[6] These observations allow for the possibility of *in situ* monitoring and control of surface morphology. A typical scenario under device growth conditions is a gradual decay of oscillations for (at least) tens of layers to an asymptotic value that may remain constant for many more tens or even hundreds of layers. Possible explanations for this behavior are a transition to step flow growth[7] or the attainment of a steady-state roughness on the surface.

In spite of the practical implications, relatively little theoretical attention has been focused on obtaining a detailed understanding of growth in this regime. While well-developed theories now exist for homoepitaxial growth in the submonolayer and asymptotically rough regimes, very few models have attempted to describe the emergence of roughness and in-plane structure and the associated damping of growth oscillations in the technologically relevant regime. Notable exceptions are the work of Vvedensky and co-workers[8,9] who were able to match RHEED oscillation decays with a simple cubic solid-on-solid (SOS) model and the more recent work of Wolf, Krug, and co-workers[10–13] who have developed a scaling theory for the damping of growth oscillations. The former stopped short of examining any details of surface morphology associated with the RHEED oscillation decay while the latter has to date been almost exclusively restricted to one dimension.

In this paper we present a scaling result for the mean step density discovered while carrying out a series of two-dimensional kinetic Monte Carlo (KMC) simulations aimed at exploring surface kinetics and morphology during multilayer growth. In particular we find that the mean step edge density, which is constant from the very first layer of growth, varies with the ratio of the surface diffusivity to the deposition flux, $R \equiv D/F$, as a nearly perfect power law over a wide range of R. We use the Wolf-Villain linear MBE equation, which has been successfully applied to the damping of growth oscillations in this regime,[10–13] to analyze this mean-field behavior. We find that it is not only consistent with the existence of a time-independent step density, but correctly predicts the evolution of the in-plane surface structure as measured by the correlation length. However, the step density scaling exponent is not correctly predicted by the linear theory, because of its inability to correctly account for the surface width in these early stages of growth. This result is not only of practical significance for the monitoring of device growth, but also indicates that current theories underlying the damping of growth oscillations are incomplete and in need of modification.

We use a cubic pair-bond SOS KMC model.[8,14] This is in the class of Arrhenius-type growth models, wherein the probabilities for executing surface diffusion processes are governed by activated rates. The energy barriers for these processes are determined only by the local bonding environment for an atom, i.e., the number and configuration of nearest neighbors. We allow only for the following processes: (i) deposition at a constant rate F, (ii) surface adatom diffusion at a rate $r_S \equiv r_0 e^{-E_S/T}$, and (iii) surface diffusion along a step edge (one nearest neighbor) at a rate $r_e \equiv r_0 e^{-(E_S+E_N-E_e)/T}$. This defines an irreversible aggregation model in which E_N, the energy barrier to break a nearest-neighbor bond, must be very large. E_e serves to cancel off most of this barrier so that the difference $E_N - E_e$ is the relatively small edge diffusion barrier. Note that in this irreversible aggregation case, edge diffusion is only permitted when there is exactly one nearest-neighbor bond. A doubly bonded edge atom, such as in the cleft of an inside corner, must break an additional bond in order to diffuse along the edge, and is therefore rendered immobile. Diffusion around an outside corner of an island is treated as a special case in

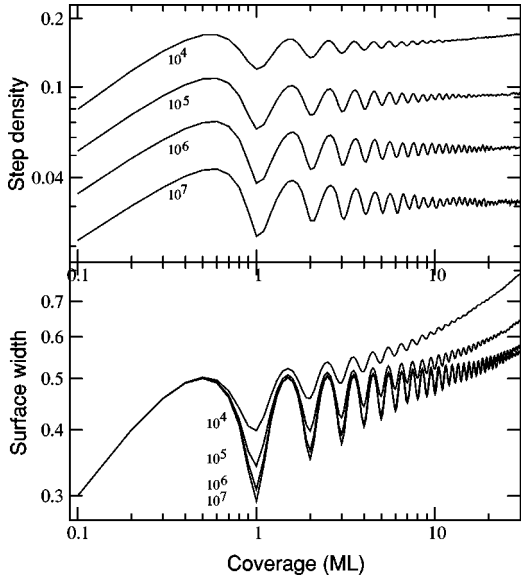

FIG. 1. Step edge density and surface width simulated for each value of D/F out to 30 ML.

which the edge atom makes a diagonal move at a rate equivalent to the edge diffusion rate. No additional step edge (Schwoebel) barrier is included, nor is evaporation allowed.

We have performed multilayer growth simulations on 500×500 lattices. Coverage extends to 300 ML and normalized surface diffusion rates $D/F\equiv r_S/F$ range from 10^4 to 10^7. For small values of D/F, 10^3 and lower, surfaces begin to roughen after the growth of only a few monolayers. Consequently, the early time scaling regime that is of particular interest here is not unambiguously present. For large values of D/F, 10^8 and higher, such simulations simply take too long in the parameter range studied to obtain good statistics. The edge diffusion rate r_e is fixed at 1/100 of the surface diffusion rate r_S. This is small enough to prevent contamination of the data by diffusion of dimers and large enough so as to still ensure compact islands for the smallest value of D/F included in the study. The results presented below represent averages over five independent runs for each parameter set.

In Fig. 1 we show the step edge density

$$\sigma=\frac{1}{N}\sum_{\langle i,j\rangle}[(1-\delta_{i,i+1})+(1-\delta_{j,j+1})], \tag{1}$$

which is computed by counting up all edges on the lattice at which there is a discontinuity, irrespective of the height difference,[15] and the surface width

$$W=\sum_{i,j}\langle h_{i,j}-\overline{h}\rangle^2. \tag{2}$$

The key observation to make is that the step density oscillates about a nominally constant value prior to the onset of kinetic roughening, while the mean surface width is an ever-increasing function of coverage. This *intermediate* regime is longer for higher D/F, and spans the growth of at least the first few tens of monolayers, consistent with experimental data generated under device growth conditions.[16]

Figure 2 shows the D/F dependence of the minima and

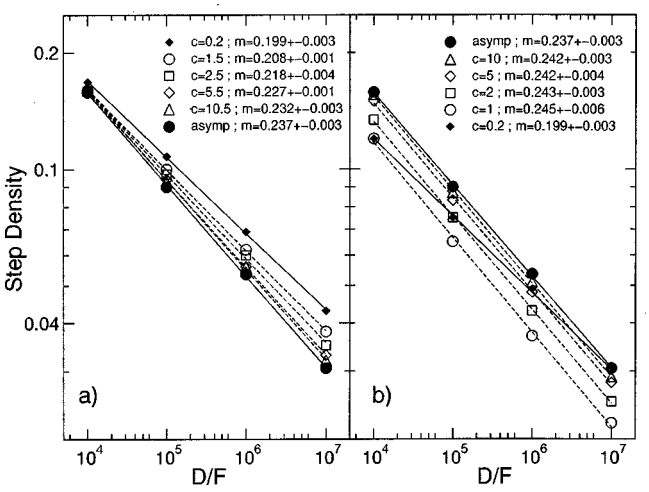

FIG. 2. Scaling of step edge density maxima (a) and minima (b). Shown are a submonolayer value along with those at 1-, 2-, 5-, and 10-ML coverage. The minima (b) correspond to integer values of coverage, while the maxima (a) correspond to coverages with an additional one-half monolayer of material. The data labeled ''asymp'' are the median values to which the step edge density oscillations first decay, prior to the onset of roughening. Solid and broken lines are linear fits, *m* gives the slope and goodness of fit.

maxima of the envelope of the step edge density σ at selected coverages. The submonolayer step edge density (small filled diamonds) scales as $(D/F)^{-0.19}$, consistent with the $-1/6$ power that is predicted by nucleation theory for the case of irreversible aggregation.[17] The constant ''mean asymptotic'' value (filled circles) scales as $(D/F)^{-0.25}$ in distinct contrast. Note that the maxima [Fig. 2(a)] evolve toward the asymptotic scaling over the first dozen or so monolayers. In contrast, the minima [Fig. 2(b)] display the same scaling as the asymptotic value, from the very first monolayer.

Many of these results can be explained within the context of a linear theory for height fluctuations. This linear theory has been used to successfully explain the scaling of the time at which oscillations decay in one-dimensional models of growth,[10,11] so the extent to which it can explain the scaling of step density observed here is another important test of its applicability to problems of this type. The lowest-order conservative equation of motion for height fluctuations in the presence of a deposition flux F and stochastic noise η is

$$\partial_t h=-K\nabla^z h+F+\eta. \tag{3}$$

The solutions of Eq. (3) for the correlation length ξ and interface width W are[18,19]

$$\xi\sim(Kt)^{1/z},$$

$$W\sim(\eta/K)^{1/2}\xi^{(z-2)/2}.$$

Assuming that there exists a single length and a single time scale, i.e., $K=l^z t$, the correlation length and surface width scale as

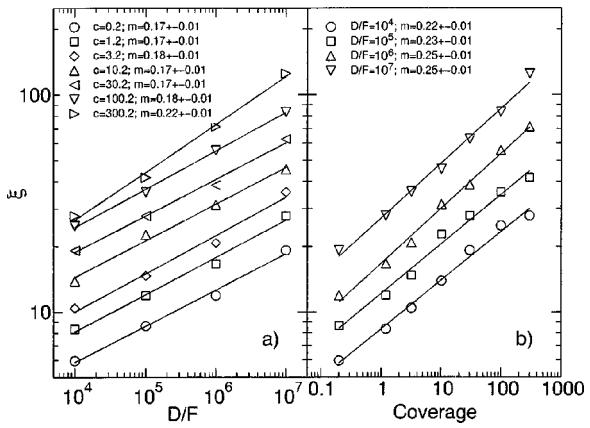

FIG. 3. Scaling of the correlation length ξ (as extracted from the peak of the structure factor of the height functions) with D/F (a) and coverage (b). Solid lines are linear fits; m gives the slope and goodness of fit.

$$\xi \sim l t^{1/z}, \tag{4a}$$

$$W \sim l^{-1} t^{(z-2)/2z}. \tag{4b}$$

For these relatively smooth morphologies, the step density should be satisfactorily described by the ratio of the surface width to the correlation length. Hence

$$\sigma = W/\xi \sim l^{-2} t^{(z-4)/2z}. \tag{5}$$

Note that if the order of the theory is $z=4$, corresponding to the Wolf-Villain linearized fluctuation theory (LFT),[20] the step density is *independent* of time (or coverage). This is consistent with our simulation data in the intermediate growth regime and invites comparison of the predictions of the linear theory for scaling of the step density with D/F in this regime. Choosing l to be the interisland spacing for irreversible aggregation as predicted by nucleation theory, $l \sim (D/F)^{1/6}$, the dependences for the correlation length, interface width, and step edge density on R ($\equiv D/F$) and coverage θ are

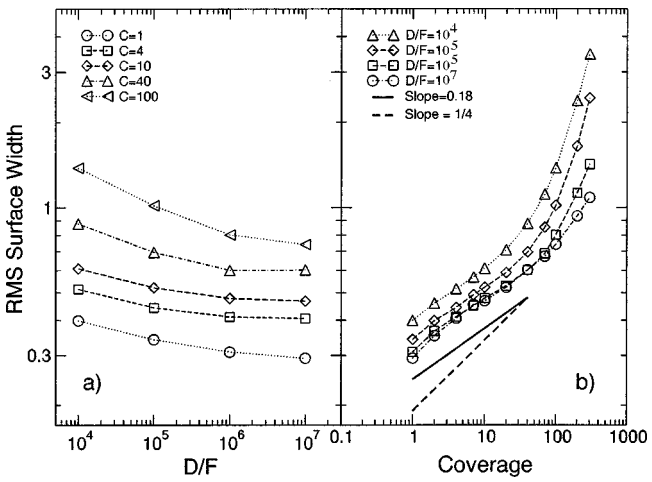

FIG. 4. Variation of the surface roughness (as computed from the surface height functions) with D/F (a) and coverage (b). Heavy lines in (b) have slopes of 0.18 and 1/4 for comparison.

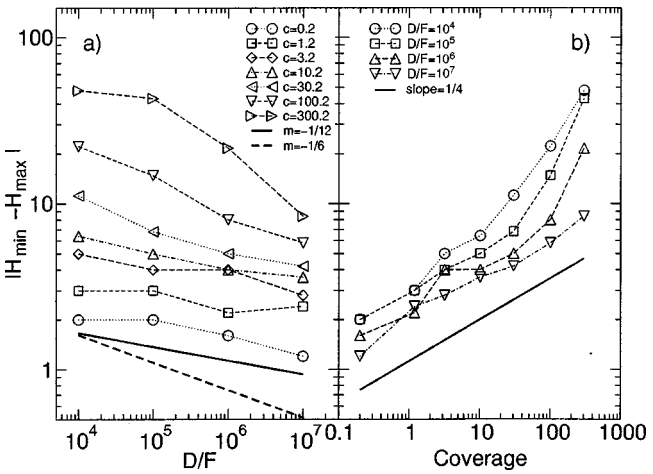

FIG. 5. Scaling of the peak-to-peak surface width (as computed from the surface height functions) with D/F (a) and coverage (b). The heavy lines in (a) have slopes of 1/12 and 1/6, while that in (b) has a slope of 1/4 for comparison.

$$\xi(R,t) \sim R^{1/6} \theta^{1/4}, \tag{6a}$$

$$W(R,t) \sim R^{-1/6} \theta^{1/4}, \tag{6b}$$

$$\sigma(R,t) \sim R^{-1/3} \theta^{0}. \tag{6c}$$

Interestingly, while the coverage dependence of the step edge density is in agreement with the predictions of the linear theory, the scaling exponent (R dependence) is *not*. In an attempt to understand this breakdown of LFT for the step density, we investigate further the behavior of the correlation function and two measures of the surface width.

Figure 3 shows the scaling of the correlation length with coverage and R as computed from the peak of the structure factor of the surface height function. The power-law fits to the data are all excellent, except for the R scaling at late times $\theta > 100$ ML when nonlinear effects begin to enter. The data indicate that ξ scales as $t^{0.25}$, independent of R, and as $R^{0.17}$, independent of coverage, in nearly perfect agreement with the prediction of the LFT. The surface roughness, as computed directly from the surface height function and shown in Fig. 4, appears to scale with coverage as $\theta^{0.18}$. This exponent is significantly lower than the value expected from LFT, 0.25. In addition, the roughness does not scale well at all with R for any coverage. A different measure of the surface width, the average peak-to-peak height H is shown in Fig. 5 and *does* display reasonable scaling with both coverage and R. So long as the surface width is below about 10 (roughness $\sim 4\%$), the scaling of this quantity with coverage

TABLE I. Theoretical and simulated exponents.

	Linear fluctuation theory		KMC simulation	
	R	θ	R	θ
ξ	1/6	1/4	0.17 ± 0.01	0.24 ± 0.01
W	$-1/6$	1/4		0.18 ± 0.01
H			-0.08 ± 0.04	0.25 ± 0.05
σ	$-1/3$	0	-0.25 ± 0.05	0.01 ± 0.05

is very consistent with a power of 1/4. The R scaling is weak, more consistent with $\sim 1/12$ power, as opposed to the 1/6 power predicted by the LFT.

Table I is a summary of the exponents for the Wolf-Villain LFT and those extracted from the simulation data. The data can be summarized as follows: (i) Wolf-Villain theory correctly predicts the time independence of step edge density. (ii) The correlation length scaling measured is as predicted by LFT. Therefore, the in-plane structure is correctly accounted for by the LFT for all coverages down to a single monolayer of growth. (iii) The rms surface width W does not display any correct scaling in this regime. The scaling of the peak-to-peak surface width H, however, is consistent with the LFT in coverage, but not in R. Consequently the ratio of H to ξ is time independent, as predicted by LFT, but displays a slightly different R scaling, 0.25 vs 1/3. Interestingly, the observed scaling of H and ξ (consistent with $\sim R^{-1/12}\theta^{1/4}$ and $\sim R^{1/6}\theta^{1/4}$, respectively) do correctly account for the full scaling of the step density σ assumed to be given by H/ξ, hence validating the assumption of a mean surface slope based on a single length scale, Eq. (5).

In conclusion, we have demonstrated that although the existence of a constant step edge density can be explained by a transition of the entire surface to step flow growth, this is not the only explanation. It can also be the subtle balance between lateral coarsening and the generation of surface width (roughness) that properly accounts for this behavior. The layer-by-layer growth regime is correctly described by the Wolf-Villain linearized theory for height fluctuations to the extent that it accounts for in-plane structure and the existence of a constant average step edge density. The linear theory fails, however, to account for the correct scaling of step density with R, but only because LFT does not describe surface width correctly. We, therefore, expect a breakdown of the previously observed scaling for oscillation decay times, as this behavior does depend on the generation of surface roughness, hence surface width. A further understanding of the role of surface width in these systems is clearly required to satisfactorily complete this picture.

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