A Coupled Equations Model for Epitaxial Growth on Textured Surfaces

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Abstract

We have developed a coupled equations continuum model that explains the complex surface shapes observed in epitaxial regrowth on micron scale gratings. This model describes the dependence of the surface morphology on film thickness and growth temperature in terms of a few simple atomic scale processes including adatom diffusion, step-edge attachment and detachment, and a net downhill migration of surface adatoms. The continuum model reduces to the linear part of the Kardar-Parisi-Zhang equation with a flux dependent smoothing coefficient in the long wavelength limit.

Key words: Theory and models of crystal growth, Physics of crystal growth, GaAs surface morphology, GaAs homoepitaxy
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1 Introduction

The problem of the time evolution of the shape of crystal surfaces has a long history dating back to Mullins and Herring who considered relaxation during

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annealing above the roughening temperature [1]. More recently, shape relaxation below the roughening temperature has been studied extensively [3,4,5]. Below the roughening temperature the problem is complicated by the need to keep track of the dynamics of atomic steps and the fact that the surface free energy of crystal facets is singular. Biasiol et al. [6] have extended the theory of shape relaxation below the roughening temperature to include the effects of atom deposition, and use this theory to explain the self limiting V-grooves observed in organo-metallic chemical vapor deposition (OMCVD) growth on corrugated GaAs substrates. In this paper we present a new continuum model which we use to interpret measurements of the shape of corrugated GaAs (100) surfaces under growth conditions which do not produce faceting. Facets are not present in our experiments due to atomic scale roughness associated with atom deposition in the island growth mode, and the fact that the surface topography is sufficiently weak that the surface slope does not reach the low energy [111] facets. We show that this model reproduces the surface morphology that develops during molecular beam epitaxy (MBE) regrowth on 1D surface gratings.

2 Conventional Modeling of Weak Surface Texture

The evolution of long wavelength surface structures during GaAs homoepitaxy can be described by the Kardar-Parisi-Zhang (KPZ) equation [1,2]: \( \partial h/\partial t = \nu \nabla^2 h + \lambda (\nabla h)^2 + F + \eta(x,t) \). The coefficients in this equation are constants characterizing the microscopic atomic processes. The source term \( \eta(x,t) \) simulates the random arrival of atoms at an average rate \( F \). According to this equation, a textured starting surface will develop parabolic mounds that smooth with time separated by V-shaped valleys. Recent experimental work [7] has shown that the KPZ equation provides an accurate description of the morphology of epitaxially grown GaAs layers for surfaces with local slopes \( \lesssim 3^\circ \). The agreement with this simple continuum model suggests that the anomalous effects associated with the singular free energy of crystal facets are not important for the growth conditions in question.

In the case of GaAs molecular beam epitaxy (MBE) growth in which there is no re-evaporation, the simplest explanation for the linear term in the KPZ equation is that it is due to an inverse Ehrlich-Schwoebel (ES) effect [1] in which surface adatoms approaching a descending step are more likely to descend over the step rather than being reflected from it, due to a step edge potential barrier. This creates a downhill flux of adatoms (\( j \propto -\nabla h \)) and a smoothing term (\( \partial h/\partial t \propto -\nabla \cdot j \)) identical to the first term in the KPZ equation [1]. In practice, the atomic scale dynamics is complex with surface reconstructions, complicated step edge geometries, and a two component (Ga, As) surface [8,9,10]. The sign of the ES effect in GaAs is controversial [7,11,12],
but we show below that a negative ES effect, favoring downhill flow (stable growth) is consistent with the experimental data.

The nonlinear term in KPZ is associated with growth along the outward normal, as in chemical vapor deposition. In this case, \( \lambda \) should be equal to the growth rate \( F \). However, the value for \( \lambda \) needed to simulate the experimental results is almost two orders of magnitude larger than \( F \) [7]. Also, the KPZ nonlinearity is non-conservative, whereas MBE growth is conservative with a growth rate that is independent of the surface shape.

In addition, the KPZ description with constant coefficients is not consistent with experiments which show that the smoothing rate depends on the growth rate. For example, in Fig. 1 we show the scattered light intensity from a GaAs surface during an interruption in growth on a randomly textured substrate. The intensity of scattered light is proportional to the power spectral density of the surface topography at a spatial frequency \( q \) determined by geometrical factors [7] (in this case \( q = 41 \text{ } \mu \text{m}^{-1} \), corresponding to a lateral surface length-scale of about 150 nm). For low amplitude surface textures, in the KPZ model the surface should smooth exponentially with a characteristic rate given by \( \nu q^2 \) where \( q \) is the spatial frequency of the surface roughness [1]. As shown in the inset of Fig. 1, the smoothing rate responds immediately to changes in the growth flux; it is faster during deposition and slower during annealing,
suggesting that $\nu$ is flux dependent. This continued smoothing of the surface in the absence of an atom flux indicates that the physical mechanisms at play on the surface still favor a net downhill migration of surface adatoms, even after the flux of atoms from the vapor has been turned off.

Insight into why the smoothing rate depends on the flux can be obtained by comparing an atomic force microscope (AFM) image from a sample which is fast cooled (quenched) after growth with the AFM image of a surface which has been annealed (see Figs. 2a and b). The quenched sample (a) is covered with small islands, whereas the annealed sample (b) has broad terraces with few islands. The small islands must coalesce into the step edges during annealing. The kinetic barrier to the adatom coalescence into the step edges, causes the growth process to be non-local in space and time, in contrast to KPZ. A high density of steps at one location that absorb adatoms will affect the adatom density and hence the growth rate at another nearby location.

3 Coupled Growth Equation Model

The growth phenomena discussed above can be explained in a natural way if we extend the growth model to include the adatom dynamics explicitly with two coupled growth equations (CGE) [13]:
\[
\frac{\partial n}{\partial t} + \nabla \cdot j = F - \frac{\partial h}{\partial t}, \tag{1a}
\]
\[
\frac{\partial h}{\partial t} = 2Dn^2 + (\beta Dn - \kappa)S. \tag{1b}
\]

Eqn. 1a is a continuity equation for the adatom density \( n \) with source and sink terms, while Eqn. 1b describes the time dependence of the surface height \( h \), which depends on the dimer nucleation rate and the net adatom attachment rate at steps. The constants are defined in atomic units as follows: \( F \) - deposition rate from the vapor, \( D \) - adatom diffusion coefficient, \( S \) - density of steps, \( \kappa \) - rate of thermal evaporation of atoms from step edges into the adatom phase, and \( 0 < \beta < 1 \) is the sticking coefficient for an adatom at a step edge. An adatom is defined as a diffusing unit on the surface, which could be a Ga atom or a Ga-As complex. We also define:

\[
j = -D(\zeta n\nabla h + \nabla n), \tag{2a}
\]
\[
S = \sqrt{S_0^2 + (\nabla h)^2}. \tag{2b}
\]

where in Eqn. 2a, \( j \) is the surface current of adatoms and \( 0 < \zeta < 1 \) is a proportionality constant which describes the net downhill drift of adatoms. The second term in Eqn. 2a represents adatom diffusion. In Eqn. 1b, any adatom that attaches to a step edge is assumed to have incorporated into the film. The downhill drift parameter \( \zeta \) can be positive or negative: a positive value favors downhill drift of adatoms, consistent with the surface smoothing that is observed experimentally for GaAs (001) [11,7] (and also consistent with a negative value for the ES energy barrier).

In Equation 2b, we present a physically plausible hypothesis for the dependence of the rms step density on the surface slope. In this expression the random local surface slope associated with the growth-induced step density \( S_0 \) is added to the deterministic macroscopic surface slope \( \nabla h \). Since the local slope associated with the background step density \( S_0 \) is random, the two terms add in quadrature. In Equation 2b, we assume that the background step density is independent of the macroscopic surface slope. We expect \( S_0 \) to depend on temperature and deposition rate, and on time in the case of growth interrupts (see Fig. 2a and b) [14]. The simple picture of a surface consisting of flat terraces separated by atomic steps, can be expected to provide a good description as long as the surface slope does not reach the next low index crystal planes, namely (110) and (111). These planes are 45° and 54.7° from the surface normal, and beyond the range of surface slopes that we have explored experimentally (\( \lesssim 30° \)). We assume that the density of random steps \( S_0 \) is independent of the topography. In this approximation, the average step density is proportional to the rms value of the local surface slope. The expression for \( S \) is then constructed by averaging over the random orientation of the local slope, and the rms step density is given by the incoherent sum of
Fig. 3. Film thickness dependence: (A) AFM scan lines for regrowth on 100 nm deep gratings oriented perpendicular to the [110] direction; (B) Scan lines from CGE calculation; (C) Scan lines from 2D kMC simulation of 10 nm high grating structure, where one $\Delta t$ equals 5.6 ML of growth. All offsets arbitrary.

the two contributing factors.

For low amplitudes and long wavelength ($\nabla h < S_0$), the adatom density will be nearly constant as a function of position and time, and approximately equal to $n_0 = (F + \kappa S_0)/\beta DS_0$. In this case, Eqns. 1a, 1b reduce to,

$$\frac{\partial h}{\partial t} = \frac{\zeta}{\beta} \left( \frac{F}{S_0 + \kappa} \right) \nabla^2 h + F. \tag{3}$$

This reproduces the linear part of the KPZ equation and shows explicitly the dependence of the linear smoothing coefficient $\nu$ on the deposition rate and the downhill drift parameter $\zeta$. In addition, it shows that in the absence of growth ($F = 0$) the linear smoothing term is independent of the background step density $S_0$. This agrees with the light scattering data in Fig. 1, which shows that the smoothing rate is relatively constant during a growth interruption even though the AFM images in Fig. 2 indicate that the step density drops dramatically during annealing. Extending Eqn. 3 to higher order, one finds non-linear terms with higher order spatial derivatives. We speculate that the higher order nonlinear terms can be approximated by the KPZ nonlinearity over a limited spatial frequency range if the surface topography is not too large.
Textured Surfaces: Film Thickness Evolution

Growth on substrates with larger amplitude surface slopes, up to $\sim 30^\circ$, show complex surface shapes before evolving into parabolic mounds, as shown in Fig. 3a. At intermediate times the valleys are V-shaped with concave rather than convex sidewalls and distinct shoulders near the top of the sidewalls. Note the absence of (100) facets which are predicted theoretically for annealing below the roughening temperature in the absence of deposition [5]. Equations 1 and 2 can be solved in seconds with a finite difference scheme and a coupled differential-algebraic system solver, and a 1D solution is shown in Fig. 3b with parameters adjusted to match the experimental data in Fig. 3a (see Table 1 for parameters). The agreement with the experimental surface shapes is striking. In particular, the model reproduces the inverted “Gothic window” shape of the valley for the 600 nm growth and the KPZ-like cusps in the 2600 nm growth where the grating amplitude has reduced sufficiently so that the structure is described by the KPZ equation.

A continuum model cannot include the microscopic details of the atomic scale phenomena, such as the geometry and density of step edges. We therefore compare the continuum model in Eqns. 1 and 2 with a kinetic Monte Carlo (kMC) simulation, which includes the same physical processes that are included in the Eqns. 1. We use a 2D, cubic grid, one-component, restricted solid-on-solid (SOS) model, with nearest-neighbor interaction. Each atom bonds to the surface with an activation energy $E_{\text{act}} = E_{\text{sub}} + mE_{\text{lat}}$, where $m$ is the number of lateral neighbors [15]. The kMC simulations produce a random step distribution automatically due to the statistical nature of the model. In kMC, the binding energy for an atom at a step edge depends on how many neighbors it has ($\sim mE_{\text{lat}}$), whereas in the CGE continuum model a single average value is used for the step edge binding energy.

Table 1
Parameter table for CGE calculations. Atomic units were used with a lattice constant of 0.3 nm was used.

| Figure | T   | F   | D   | $\kappa$ | $S_0$ | $\beta$ | $\zeta$ |
|--------|-----|-----|-----|----------|-------|---------|--------|
| 2 (b)  | 580 | 1.0 | 180 | 3.0      | 0.075 | 0.3     | 0.075  |
| 3 (b)  | 420 | 0.8 | 0.2 | 0.00025  | 0.025 | 0.1     | 0.15   |
| 550    | 0.8 | 9.0 | 0.019 | 0.02 | 0.1 | 0.15 |
| 550    | 0.8 | 60  | 0.19 | 0.02 | 0.2 | 0.15 |
| 610    | 0.8 | 460 | 2.0 | 0.01 | 0.4 | 0.15 |

SOS simulations of MBE growth by kMC are limited by available computing power to small scale structures, and become intractable for realistic, high temperature growth scenarios where 2D systems have sides up to microns and
growth times on the order of hours. In Fig. 3c, we show a kMC simulation for a surface grating that is somewhat smaller than the experimental structures. The simulated grating profiles in Fig. 3c were obtained by projecting 2D kMC simulations onto a line at each time point by taking the average elevation perpendicular to the scan line. In this simulation, \( E_{\text{sub}} = 1.25 \text{ eV}, \ E_{\text{lat}} = 0.35 \text{ eV} \) and an ES step-edge barrier of \( E_{ES} = -0.05 \text{ eV} \) was used for the downhill drift mechanism. The agreement with the experimental shapes is excellent, reproducing all of the main features, except they are on a smaller size scale. The substrate and lateral binding energies are similar to values reported earlier in the interpretation of RHEED data \[16,17\] and compatible with the fitting parameters found in the continuum model. It is plausible that similar shapes could be obtained for the larger size scales relevant to the experiments by scaling the parameters appropriately. In the case of the CGE model (Eqns. 1, 2) we find that the parameters can indeed be scaled to yield similar surface shapes at different length scales \[18\].

5 Textured Surfaces: Temperature Evolution

In Fig. 4a, we show the dependence of the surface topography on growth temperature, for a fixed layer thickness together with (b) the simulated surface topography using Eqns. 1 and 2 and parameters as outlined in Table 1. The experimental data is obtained from growths on 100 nm deep gratings oriented perpendicular to [110]. This is the faster diffusion direction in this material system \[7\], and depends on the As\(_2\)/Ga ratio during growth, which was equal to three for all growths discussed in this work. This observation is consistent with the values used for the downhill drift parameter in our calculations, where the best fits were obtained using a larger \( \zeta \) when the gratings were oriented perpendicular to [110] (Fig. 4b) than for gratings perpendicular to the [110] direction (Fig. 3b). The diffusion constant \( D \), however, was considered isotropic in all calculations in this paper. There is some uncontrolled variation in the pitch and depth of the gratings in the experimental data in Fig. 4a. Nevertheless, the CGE model reproduces the main features in the temperature dependent data, namely the small secondary mound in the valley at 500°C, the KPZ-like cusp at 550°C and the inverse Gothic window shape for the valleys at 610°C. The shoulders at the edges of the ridges at 610°C are also reproduced by the model, although they are not as sharp as in the experimental data.

The parameters used in these calculations are based on the same energies used in the kMC simulations in Fig. 3c. The diffusion constant is related to the substrate binding energy through the expression \( D = (2kT/h) \exp (\frac{-E_{\text{sub}}}{kT}) \). The step edge detachment rate is calculated from \( \kappa = D \exp (\frac{-\hat{m}E_{\text{lat}}}{kT}) \), where \( \hat{m} \) is an average number of neighbors for atoms at a step edge which we set equal to 2.25. The declining value of \( S_0 \) with temperature is reasonable;
one might also expect $\zeta$ to decrease weakly with temperature. Satisfactory fits to the data were also obtained with a larger activation energy for $D$ (1.8 eV rather than 1.25 eV) together with a smaller prefactor and somewhat different (yet still physically reasonable) temperature dependences for the other parameters. Experimental and theoretical work suggests that the activation energy for $D$ is in the 1.5-2.0 eV range. [16,17,19,20].

6 Conclusions

We have shown that the complex surface morphology which develops during epitaxial regrowth on patterned GaAs (100) substrates, can be explained by the dynamics of the deposited adatoms, including step edge attachment and detachment, adatom diffusion, and downhill drift. Although we attribute the downhill drift to a negative Ehrlich-Schwoebel barrier we cannot rule out the possibility that this effect is caused by some other mechanism. This analysis is specific to GaAs, but the concepts are generic and may be applicable to other systems.
References

[1] See for example: A. Pimpinelli, J. Villain, *Physics of Crystal Growth* (Cambridge Univ. Press, Cambridge, 1998).

[2] A. Ballestad, B. J. Ruck, M. Adamcyk, T. Pinnington, T. Tiedje, *Phys. Rev. L.* **86**, 2377 (2001).

[3] *Dynamics of Crystal Surfaces and Interfaces*, ed. P. M. Duxbury, T. J. Pence, (Plenum Press, NY, 1997).

[4] J. Erlebacher, M. J. Aziz, E. Chason, M. B. Sinclair, J. A. Floro, *Phys. Rev. Lett.* **84**, 5800 (2000).

[5] V. B. Shenoy, A. Ramasubramaniam, L. B. Freund, *Surf. Sci.* **529**, 365 (2003).

[6] G. Biasiol, A. Gustafson, K. Leifer, E. Kapon, *Phys. Rev. B.* **65**, 205306 (2002).

[7] A. Ballestad, B. J. Ruck, J. H. Schmid, M. Adamcyk, E. Nodwell, C. Nicoll, T. Tiedje, *Phys. Rev. B.* **65**, 205302 (2002).

[8] P. Tejedor, F. E. Allegretti, P. Smilauer, B. A. Joyce, *Surf. Sci.* **407**, 82 (1998).

[9] J. Tersoff, M. D. Johnson, B. G. Orr, *Phys. Rev. Lett.* **78**, 282 (1997).

[10] Ch. Heyn, M. Harsdorff, *Phys. Rev. B.* **55**, 7034 (1997).

[11] V. R. Coluci, M. A. Cotta, C. A. C. Mendonça, K. M. I.-Landers, M. M. G. de Carvalho, *Phys. Rev. B.* **58**, 1947 (1998).

[12] M. D. Johnson, K. T. Leung, A. Birch, B. G. Orr, J. Tersoff, *Surf. Sci.* **350**, 254 (1996).

[13] Related coupled equations have been used earlier to describe growth on faceted surfaces [21,22]. In this work the adatom incorporation rate was dependent on the crystal facet orientation.

[14] W. Braun, B. Jenichen, V. M. Kaganer, A. S. Shtukenberg, L. Däweritz, and K. H. Ploog, *J. Cryst. Growth* **251**, 56 (2003).

[15] A. Madhukar, S.V. Ghaisas, *CRC Crit. Rev. Sol. State and Mater. Sci.* **14**, 1 (1988).

[16] B. A. Joyce, *J. Mat. Sci. - Mat. in Electronics*, **14**, 591 (2003);

[17] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave, and B. A. Joyce, *Phys. Rev. B.* **46**, 6825 (1992).

[18] A. Ballestad, PhD thesis, in preparation. University of British Columbia, Canada.

[19] H. Yang, V. P. LaBella, D. W. Bullock, Z. Ding, J. B. Smathers, and P. M. Thibado, *J. Cryst. Growth*, **201/202**, 88 (1999).
[20] P. Kratzer, E. Penev, M. Scheffler, *Appl. Phys. A* **75**, 79 (2002).

[21] S. Koshiba, Y. Nakamura, M. Tsuchiya, H. Noge, H. Kano, Y. Nagamune, T. Noda, and H. Sakaki, *J. Appl. Phys.* **76**, 4138 (1994).

[22] W. Braun, V. M. Kaganer, A. Trampert, H.-P. Schönherr, Q. Gong, R. Nötzel, L. Däweritz and K. H. Ploog, *J. Cryst. Growth* **227-228**, 51 (2001).