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LETTER TO THE EDITOR

The dynamic variations of terrace length during growth on stepped surfaces

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Abstract. A recently derived theoretical method for modelling molecular beam epitaxy on stepped surfaces, which includes a nonlinear term for nucleation, has been extended so that large deviations from periodic step structure can be examined. The method is used in conjunction with Monte Carlo simulations to monitor the growth dynamics of a stepped surface with unequal terrace lengths and identify the stable configuration. We show that the equidistant step configuration is favoured even in growth regimes where nucleation on the terraces competes with atom incorporation at steps. Furthermore, we found a remarkable qualitative correspondence of the results obtained from the nonlinear diffusion equations and the simulations.

Recently there has been a renewed interest in the theoretical study of crystal growth, motivated in part by technological developments in precise methods for the fabrication of semiconductor crystals, such as molecular beam epitaxy (MBE). The use of vicinal (stepped) substrates at sufficiently high temperatures to allow growth by step propagation to dominate has proved particularly valuable for device construction. It is therefore vital to know what effect growth has on the step configuration, since the stability of the step positions is critical in obtaining well defined structures such as tilted superlattices [1].

Previous analytical models of crystal growth by step propagation are based on the near-equilibrium theory of Burton, Cabrera and Frank (BCF) [2], which assumes that a steady-state array of equidistant steps is stable under growth conditions. It is important to investigate the validity of this assumption, particularly when displacements from the equidistant configuration are large. The stability of an array of equal-length terraces to small length perturbations for the linear BCF theory has been analysed under the quasi-steady-state approximation, i.e. that the adatom fluxes to perturbed steps have the same form as in the steady-state solution [3-5]. Recently, Ghez *et al* [6] have included in BCF theory the effect of the moving boundary and departures from equilibrium at the step edges—both important under the highly non-equilibrium growth conditions of MBE—in a linear stability analysis of step motion and concluded that a train of equally spaced steps is stable to small perturbations under growth conditions.

In none of the aforementioned treatments, however, was nucleation on the terraces considered, despite the fact that island formation is an important part of MBE on stepped surfaces, even dominating over incorporation at the step edges under certain conditions [7]. Although it has been suggested that island formation may destabilize

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equidistant step arrays to perturbations [5], nucleation has not been included in any previous stability analyses. Certainly the nucleation rate, which is nonlinear in the adatom concentration [8, 9], precludes a straightforward linear stability analysis.

In this letter, we use Monte Carlo computer simulations and a nonlinear diffusion equation which includes diatomic nucleation, both developed previously to model MBE on stepped surfaces, to track the temporal growth of a step train with alternating unequal terraces. Unlike previous stability analyses which were restricted to either small perturbations or a quasi-steady-state approximation, both techniques follow the full time-dependent behaviour of the large initial perturbations and predict the approach to steady state as well as the final configuration. We show that step positions and velocities, and the surface coverage, exhibit temporal oscillations for certain growth conditions, but that a final steady state with equidistant steps is always achieved. Including island formation on the terraces in the nonlinear theory increases the speed of the approach to the final steady state. From these results we argue that a small amount of nucleation will actually help to *stabilize* perturbations in periodic step arrays. Description of each approach and its application to a highly perturbed step train is given below.

We use a Monte Carlo simulation, employed previously to model MBE [10], in which we can easily incorporate a vicinal substrate. The method, which is described in detail elsewhere [10], is based upon the solid-on-solid model proposed by Weeks and Gilmer [11]. During growth, material is deposited randomly onto the lattice and migration of atoms on the surface occurs via an Arrhenius diffusion term. The energy barrier to diffusion depends on the local configuration. The barrier consists of the energy bond to the substrate and the bonds formed with any nearest neighbours in the same horizontal plane of the lattice. In all the cases reported here, we performed our simulations on a 120×120 lattice with a substrate binding energy of 1.3 eV and an in-plane nearest-neighbour bond of 0.25 eV. Prior to growth, the lattice is constructed of steps with alternating short and long terraces.

The nonlinear growth theory for equal-length terraces has been described previously [8, 9]; here we consider its extension to a system with alternating terrace lengths. The surface is an infinite train of flat terraces alternating in lengths of h_1 and h_2 the x-direction moving with velocities $v_1(t)$ and $v_2(t)$, shown in figure 1. Since the initial step structure alternates between the two terrace lengths on each successive step, we need only consider the system of two adjoining terraces. We further assume that the step edges are straight on average in the y-direction; thus the concentration of single adatoms on the terraces $n(x, t)$ is one-dimensional and the substrate is a continuum, whereas the simulation model is fully two-dimensional and treats discrete lattice sites.

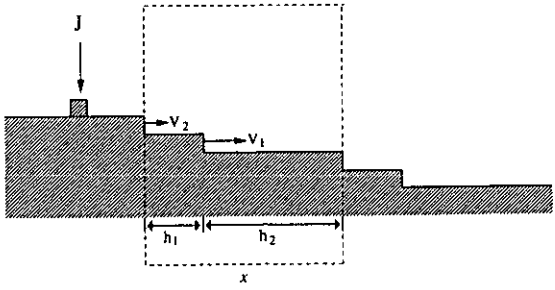

Figure 1. Schematic of a cross section through the crystal with alternating-length terraces.

In a stationary frame the diffusion equation is:

$$\frac{\partial n(x,t)}{\partial t} = D\frac{\partial^2 n(x,t)}{\partial x^2} + J - 2\sigma D n^2 - 2Jm\frac{n}{n_0} \tag{1}$$

where J is the adatom flux to the surface and D is the diffusion constant. The last two terms in (1) represent the formation of diatomic islands and are thus an approximation of adatom nucleation processes on the terraces. Here, σ is a capture efficiency for the collision of diffusing adatoms to form pairs, m is the number of sites around a single adsorbed adatom that will form a diatomic island when filled by impingement from the beam, and n_0 is the concentration of lattice sites.

It is convenient to utilize dimensionless variables so that the concentration is of order unity. Thus we define $y \equiv Dn/Jh_{av}^2$, where $h_{av} = (h_1 + h_2)/2$, and $x \to x/h_{av}$ so that $0 \leq x \leq 2$ over the two-terrace system. Time is scaled by the amount of time necessary to deposit one monolayer, $t \to t/(n_0/J)$, and the velocities are transformed as $v_i \to v_i n_0/Jh_{av}$, $i = 1, 2$. Note that for equal terraces ($h_1 = h_2$), the velocities are unity at steady state, where $v_i n_0 = Jh_i$. We rewrite (1) in terms of the new variables, and with reference to a coordinate system which moves with the velocity v_2 of the longer step:

$$\frac{\partial y}{\partial t} = \frac{\partial^2 y}{\partial x^2} + v_2\alpha\frac{\partial y}{\partial x} + 1 - 2\alpha\beta y^2 - 2\alpha m y. \tag{2}$$

The dimensionless parameter $\alpha = Jh_{av}^2/Dn_0$ represents the ratio of the diffusion time for an adatom to move an average terrace length (h_{av}^2/D) to the inter-arrival time of atoms per site (n_0/J), and the dimensionless parameter $\beta = \sigma n_0 h_{av}^2$ is related to the misorientation angle through the average terrace length with monatomic steps.

We denote $x^*(t)$ as the dimensionless position of the inner step, which moves at a velocity $v_1 - v_2$ relative to our reference frame. Since at $t = 0$, prior to growth, $x^*(0) = h_1/h_{av}$, we obtain $x^*(t) = h_1/h_{av} + \int_0^t (v_1 - v_2)\,dt$ for the position of the inner boundary with respect to the outer steps. The boundary conditions for $n(x, t)$ are determined by the assumption that the step edges act as perfect sinks, thus $n(0, t) = n(x_+^*, t) = n(x_-^*, t) = n(2, t) = 0$. Here, x_+^* and x_-^* denote respectively the inner boundary approached from below or above the step edge.

The dimensionless pair concentration $Y(x, t) \equiv DN/Jh_{av}^2$ is governed by a continuity equation similar to that for the adatom concentration:

$$\frac{\partial Y}{\partial t} = v_2\alpha\frac{\partial Y}{\partial x} + \alpha\beta y^2 + \alpha m y. \tag{3}$$

At the end of the steps, the step is being freshly formed and there can be no islands on top of it: $Y(x_-^*, t) = Y(2, t) = 0$.

Solution of (2)-(3) requires additional relations for the unknown step velocities $v_1(t)$ and $v_2(t)$. A local mass balance at the step edge which accounts for both the diffusive flux of the adatoms into the step and the convective flux of upswept islands yields:

$$v_1 = \left.\frac{dy}{dx}\right|_{x=x_-^*}^{x=x_+^*} + v_1\alpha Y(x_+^*, t) \qquad v_2 = \left.\frac{dy}{dx}\right|_{x=2}^{x=0} + v_2\alpha Y(0, t). \tag{4}$$

The equations (2)-(4) are discretized in time and space via an implicit finite difference method [12]. Since we have transformed to coordinates moving with velocity

v_2, the two outer boundaries are fixed. However, the inner step moves with respect to our fixed system with a velocity $v_1 - v_2$ and, in general, it will not coincide with a spatial grid point in successive time steps. This difficulty is overcome by allowing the inner step to move between grid points and using Lagrangian interpolation to discretize the equations around the moving inner step [13].

In figure 2 we show the relative position of the inner step $x^*(t)$ with and without nucleation as a function of α for an initial terrace distribution of $h_1/(h_1+h_2)=0.25$ and $h_{av}=10a$, where a is the distance between lattice sites. Under growth conditions, the alternating-length step train is unstable and will always eventually decay to a state where all terrace lengths are equal $[x^*(t\to\infty)=0.5]$. Note that when nucleation is neglected, recovery to the steady state is much slower and oscillations are absent.

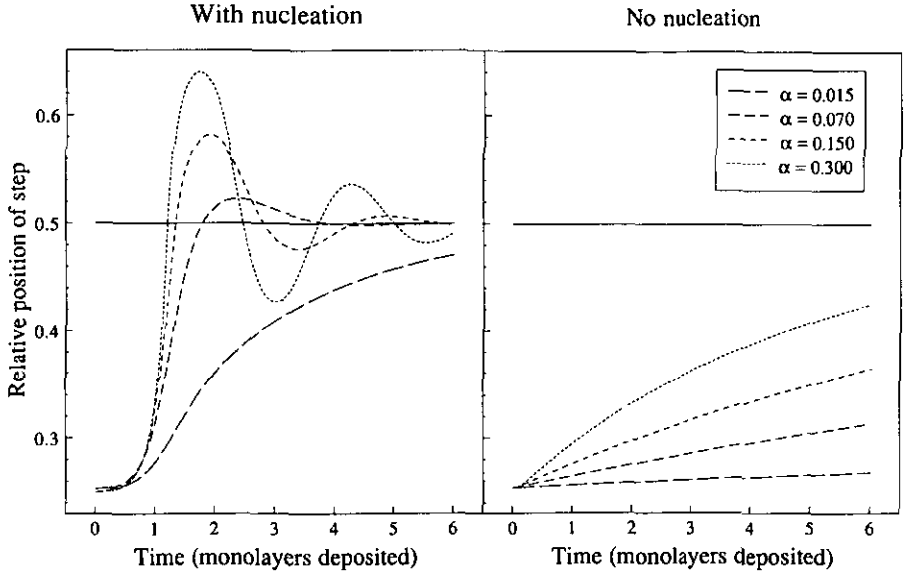

Figure 2. Relative step position $x^*(t)$ for initial configuration with $h_1/(h_1+h_2)=0.25$ calculated from the nonlinear theory with nucleation and $\beta=100$ (left panel), and without nucleation ($\beta=m=0$ in (1)) (right panel).

At low α (since α is inversely proportional to D, this corresponds to high temperature (high D)) there is very little nucleation on the terraces. Growth is dominated by step propagation since most adatoms are incoporated at the step edge. However, there is slightly more nucleation on the larger terrace than on the smaller one, thus leading to a larger incorporation rate of islands at the short step and faster growth relative to the large step. Additionally, due to the moving boundary, the single adatom concentration is slightly skewed so that more migrating adatoms enter the step from below than above. Since this effect is more pronounced for the larger terrace, the net result is also a higher flux at the shorter step. The step position slowly and monotonically increases to a steady-state equidistant configuration.

At higher α (lower temperature) the situation is markedly different. Island formation on the terraces competes with incorporation of adatoms at the step edges. Since the dimensionless nucleation rate depends on the square of the terrace length, islands build up far more densely on the large terrace than on the small one. Eventually, after

the deposition of close to a monolayer, the build-up of islands on the large terrace is such that the short step rapidly picks up speed and moves swiftly across the large terrace, leaving behind a sparsely populated region. The relative terrace lengths are now reversed and the island build-up process is repeated on the alternately larger step until an equidistant steady state is reached. Thus the velocities of each step oscillate out-of-phase, leading to an oscillation of the step position around its steady-state value. In the absence of nucleation, a similar effect occurs at extremely high α (very low temperatures). In this case, however, the build-up of material on the terraces is due to an unrealistic mechanism, namely an accumulation of practically immobile adatoms on the terrace without any nucleation of the atoms.

Due to the discrete nature of the two-dimensional simulations, calculation of adatom concentrations and step positions from the simulations is somewhat ambiguous. Instead, we calculate the coverage of the surface with odd-height sites for comparison with the continuum model. For the initial configuration $h_1/(h_1+h_2)=0.25$, coverage of the surface with odd-height steps is 0.75. The temporal evolution of the surface is shown for decreasing temperatures in figure 3; for comparison the solution of the nonlinear model (increasing α which corresponds to decreasing temperature)is also shown. The noise present in the simulated curves is due to the finite size of the lattice. The noise fluctuations increase with temperature and lead to uncertainty in the exact position of the maxima and minima of the oscillations for high temperatures. The qualitative agreement between the simulations and the nonlinear theory is remarkable; both exhibit non-monolayer oscillations in the coverage for low temperature (high α) which disappear at increased temperature (lower α). In both cases, the steady-state odd-height coverage is 0.5. Comparison with figure 2 shows that the period of the coverage oscillations is approximately the same as the period of the oscillations in the step positions, as one would expect since the coverage is related to the relative sizes of the alternating terraces. The minima for the coverage oscillations are displaced to lower times, however, than those of the step position oscillations, since coverage also

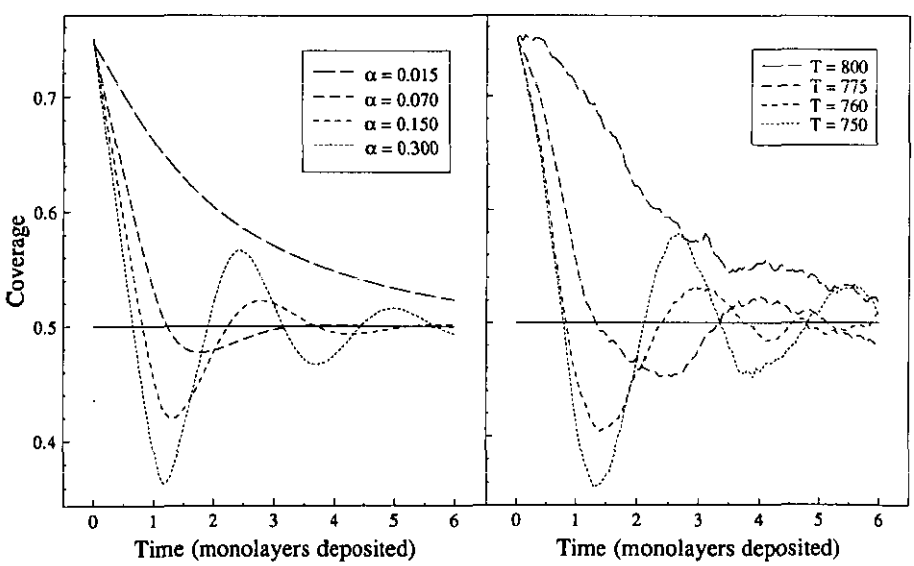

Figure 3. Coverage of odd-height steps during growth for nonlinear theory with $\beta=100$ (left panel) and from simulations (right panel).

includes the population of islands on each terrace as well as the relative sizes of the terraces.

The role of nucleation in driving the system toward equidistant step arrays can be understood as follows. More nucleation will occur on the larger terrace and a greater island concentration will build up. Since islands can only be incorporated below the step, this means that a shorter step will always have a higher velocity due to convective upsweep of islands on the long terrace below. It will thus move faster relative to the long step, which sweeps up relatively few islands from the short terrace below it. If overshoot occurs, the process is then reversed to force the system back to the equidistant steady state. Therefore we conclude that a small amount of nucleation will in fact act to *stabilize* step trains to perturbations in terrace length. We would however like to stress that this conclusion holds for only relatively small amounts of island formation; in regimes where nucleation is the dominant growth mechanism and growth may be distributed over more than one layer on a given terrace, this treatment, based on a step propagation growth mode, is no longer applicable. If nucleation is not included in the model (right panel of figure 2), the only mechanism for driving the system to an equidistant configuration is the imbalance in adatom fluxes above and below the step due to the moving reference frame. Since this effect is relatively weak, the perturbed system recovers much more slowly to the steady-state value than if nucleation were considered. For instance, for $\alpha = 0.015$, the relative step position has moved less than one tenth of the way to its final value after deposition of six monolayers, whereas with the addition of nucleation the step position has recovered almost ninety per cent of the distance to the steady-state value in the same time.

Of particular interest is the fact that the qualitative behaviour of the step motion as a function of growth conditions is the same for both simulations and theory. Since the simulations are fully two-dimensional and thus incorporate fluctuations and growth perpendicular to the step train, while the continuum model is one-dimensional and only approximates the complex nucleation processes, the correspondence between the results of the two models is remarkable. The only discrepancy between the simulations and the theory is a quantitative one; namely, using the same parameters, the nonlinear theory predicts that coverage oscillations are exhibited at lower temperatures than in the simulations. This is due to the fact that only diatomic nucleation is included in the nonlinear theory; at low temperatures we have shown that for a quantitative treatment higher-order nucleation must be considered [14].

In conclusion, by tracking the temporal evolution of alternating-length steps using simulation and a nonlinear continuum theory, we have shown that even under conditions where nucleation processes on the terraces are significant, step trains with terrace length perturbations will evolve to a stable, equidistant steady state. A small amount of island formation greatly hastens the recovery of systems with even very large perturbations, and will also increase the stability of an equidistant step train to disturbances in the terrace lengths during growth. Depending on the growth conditions, step positions may oscillate before settling to the steady-state equidistant array.

We plan to extend the nonlinear theory to track the full temporal evolution of step trains for complex systems where the final steady state may not be the equidistant one, such as monatomically stepped Si(100). Here, anisotropy of the surface layers may require inclusion of different adatom diffusivities on each terrace as well as attachment-detachment kinetics at step edge.

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