

Self-Organization of Steps in Growth of Strained Films on Vicinal Substrates

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Computer simulations show that if stress is present, steps on a vicinal surface can self-organize into a regular array of step bunches. Such self-organization can provide templates for subsequent fabrication of “quantum wire” nanostructures. The size and spacing of the bunches can be controlled independently. We analyze the requirements for optimal ordering. [S0031-9007(97)05273-3]

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The continued miniaturization of electronic devices is leading us into a realm of nanostructures, which exhibit novel electronic and optical properties, and have a wide range of potential applications such as single-electron transistors, quantum-dot lasers, and quantum computing. Quantum well devices are already widespread. Recent effort has been devoted to fabricating quantum wires and quantum dots, respectively, one- and zero-dimensional analogs to the quantum well. Just as uniform thickness is a prerequisite for a film in a quantum well device, uniform size and spacing are prerequisites for applications of wires and dots.

Different routes to fabricating wires and dots are possible. One can perform direct substrate patterning or lithography [1], or wirelike and dotlike structures can be induced to “self-assemble” during the growth of thin films. These self-assembled structures sometimes exhibit surprising uniformity [2–5], suggesting that their application in actual devices is a real possibility.

An approach that is, in a sense, a hybrid between deliberate nanopatterning and self-assembly is the use of vicinal surfaces as templates for the growth of wires and dots. Such surfaces are cut at a small angle to the atomic planes, creating a staircase of atomic-height steps that can serve as preferential sites for the growth of wires [6,7] and the nucleation of clusters [8,9]. But single steps tend to meander [10], and their spacing is often irregular [7]. These drawbacks have caused difficulties in previous efforts to use steps to grow semiconductor and metallic wires [6,7] or vertical superlattices [6].

Step bunches, on the other hand, tend to be much straighter than individual steps, due to their greater “stiffness” [10]. They can also have different heights depending on the number of steps in the bunches, allowing the possibility of different thicknesses for quantum wires, or direct control of the size of quantum dots that nucleate on the step bunches [11]. Recently it was shown that the steps on a vicinal surface can be induced to bunch by introducing stress, through the deposition of a thin layer of a lattice mismatched film (e.g., a SiGe alloy on Si) [12,13]. The problem is to *control* the bunching, in order to achieve a useful template structure.

In this Letter we show, through simulations, that stress-induced step bunching can be controlled to achieve uniformly sized and uniformly spaced step bunches. We demonstrate that, when growing in the step-flow mode, one should in principle be able to grow step bunches of any desired size and spacing by adjusting the growth rate, the growth temperature, the substrate step density, or the composition of the alloy.

Step-flow growth is perhaps the simplest mode of growth imaginable, with deposited atoms diffusing and attaching to existing steps. Yet it can exhibit complex behavior. Steps are sometimes observed to bunch, most often due to kinetic factors [14–16]. Even highly simplified models of kinetic step bunching exhibit a rich behavior that is not completely understood [14].

For a strained layer, there exists an attractive interaction between steps [13,17]. This leads to a bunching instability that is thermodynamically driven, and thus distinct from kinetic bunching. We focus here on the steady-state growth morphology resulting from such stress-induced step bunching, and on the prospect of systematically controlling this morphology.

We use a one-dimensional (1D) model [18] for step-flow growth of a surface under stress, such as the surface of a heteroepitaxial layer. Steps move only by attachment and detachment of adatoms, which diffuse across the terraces driven by gradients in the chemical potential. These gradients arise because the steps act as sinks for adatoms, and the elastic forces cause some steps to be better sinks than others. One can derive an equation of motion for the steps by integrating the diffusion equation for adatoms between steps, with a boundary condition determined by thermodynamic equilibrium with the steps [13]. This gives

$$\frac{dx_i}{dt} = F \left(\frac{x_{i+1} - x_{i-1}}{2} \right) - B \left(\frac{f_{i+1} - f_i}{x_{i+1} - x_i} - \frac{f_{i-1} - f_i}{x_{i-1} - x_i} \right). \quad (1)$$

Here x_i is the position of the i th step in the direction perpendicular to the step; F is the adatom flux per site; and f is the force per unit length on the step due to elastic

step-step interactions. B is closely related to the surface diffusion constant,

$$B = \frac{DA}{2kT} e^{-E_d/kT}, \quad (2)$$

where D is the adatom diffusion coefficient, A is the area per surface site, and E_d is the detachment energy of an adatom from a step.

The force f can be written as

$$f_i = \sum_{j \neq i} \left(\frac{\alpha_1}{(x_j - x_i)} - \frac{\alpha_2}{(x_j - x_i)^3} \right). \quad (3)$$

Here α_1 reflects the attractive interaction arising from the elastic relaxation around each step on a strained layer, and α_2 reflects the repulsion arising from the inherent stress (the “force dipole”) of each step. The elastic theory of these interactions has been discussed elsewhere [17].

Five parameters define our model: F , B , α_1 , α_2 , and the average step spacing L_{av} (which is determined by the angle of the overall surface relative to the atomic planes). However, with a rescaling of energy, length, and time, the behavior depends on only two independent ratios of characteristic lengths: L_0/L_{av} , where $L_0 = \sqrt{\alpha_2/\alpha_1}$ is the equilibrium spacing of an isolated step pair, and L_d/L_{av} , where $L_d = (B\alpha_1/F)^{1/3}$ is a length reflecting both diffusion and strain. L_0 can be adjusted through α_1 by changing the alloy composition, and L_d by changing the temperature or growth rate.

The parameters used here correspond to $L_0/L_{av} = 0.192$; the results should be qualitatively similar as long as $L_{av} \gg L_0$. For convenience, we refer to varying the flux F , with $B\alpha_1$ fixed at 10^5 , but it would be equivalent, and perhaps more practical in an experiment, to vary B through its exponential dependence on temperature.

We simulate the dynamics by direct integration of Eq. (1). The attractive interaction leads to step bunching [13], while the growth flux tends to break up large bunches [14]. Breakup of a bunch occurs when the large adatom current diffusing to the leading step and driving it forward overcomes the step-step attraction. After sufficient time, the system approaches a steady-state step morphology.

Figure 1(a) shows a portion of the surface after reaching steady-state growth for $F = 25$. It consists of rather well-ordered bunches of 3–4 steps, plus a comparable number of “free” steps. Individual steps are emitted by one bunch and captured by the next. The step bunches themselves are relatively static, with free steps dynamically attaching to and detaching from them.

The average bunch size decreases continuously with increasing F/B , reflecting the competition between thermodynamics (strong bunching for small F/B) and kinetics (suppression of bunching [14] for large F/B). Because larger bunches tend to emit steps, and smaller bunches to capture them, the tendency is for all bunches to converge to the same size. The spatial distribution of bunches also

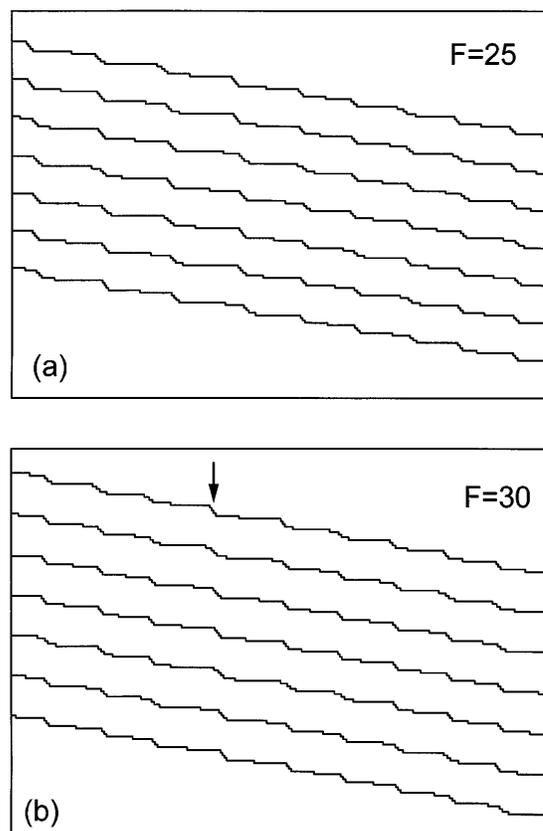


FIG. 1. Sequence of step configurations in the steady-state growth regime (from bottom to top) for deposition totaling about 1.2 monolayers (ML). The interval between subsequent configurations corresponds to about 0.2 ML deposition. (a) $F = 25$. A “phase boundary” separates the left and right halves of the figure: the left 4 bunches have 4 steps and the right 5 bunches have 3 steps. The spacings between 4-step bunches are consistently larger than those between 3-step bunches. (b) $F = 30$. All bunches have three steps except for one with four steps, marked by the arrow.

becomes rather regular, if one ignores the highly mobile free steps, as Fig. 1(a) shows.

For $F = 25$ ($L_0/L_{av} = 0.192$ and $B\alpha_1 = 10^5$), most bunches have either 3 or 4 steps. The presence of two different bunch sizes limits the degree of order. To quantify the ordering, Fig. 2(a) shows the pair correlation function for steps,

$$g(r) = \frac{1}{N_s} \sum_{ij} \delta(|x_i - x_j| - r), \quad (4)$$

of the $F = 25$ steady state, where N_s is the total number of steps. For a perfectly ordered step-bunch array, this would be a series of equally spaced sharp peaks, whose width corresponds to the (small) size of the individual bunches, and whose spacing reflects the spacing between bunches.

Figure 2(a) shows a split first peak, indicating the presence of two distinct bunch separations. These can

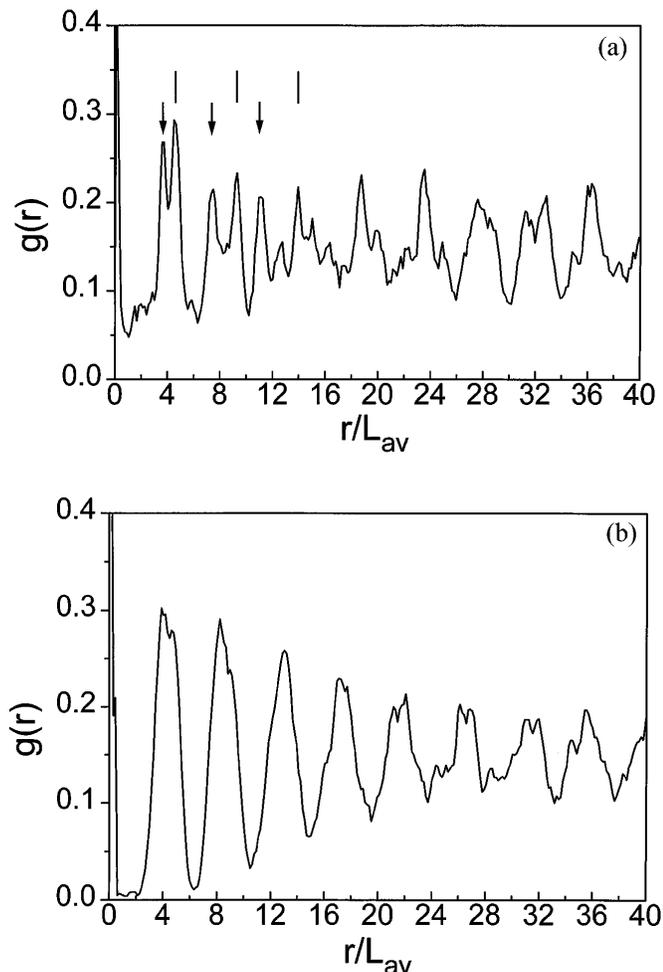


FIG. 2. Step-step correlation functions for steady state at different deposition fluxes F . The results are averaged over ten configurations well separated in time. (a) $F = 25$. The vertical lines with and without arrows mark two separate series of peaks, corresponding to two distinct spacings. (b) $F = 30$. The presence of equally spaced peaks up to high order reflects the strong long-range order of the bunch array.

be associated with bunches of 3 and 4 steps. Moreover, there are higher-order peaks for each spacing, but no low-order “mixed” spacings. Thus the system appears to be segregating into separate regions of 3-step bunches and 4-step bunches. This is confirmed by direct inspection of the steady-state configuration. Figure 1(a) shows the region around a “domain boundary,” with 4-step bunches on the left, and 3-step bunches on the right.

Our goal is not only to observe step-bunch ordering, but to understand how it may be controlled for nanofabrication. Perfect uniformity would require all bunches to have the same size. We therefore increased the flux to $F = 30$, to decrease the average bunch size. In the resulting $F = 30$ steady state [Fig. 1(b)] most bunches have 3 steps. The improved ordering is even more striking in the pair correlation function, Fig. 2(b). There is a

single characteristic bunch spacing, reflected in a series of equally spaced peaks. Moreover, the peaks remain quite distinct up to at least 13th order, limited only by the size of our simulation cell.

So far we have emphasized the average bunch size as controlling the degree of order. From a more fundamental perspective, we must consider the dynamical stability of a given step configuration. Consider for simplicity a system of straight steps having a short-range attractive interaction. These will form bunches of ν steps (including single steps, $\nu = 1$). It is a reasonable approximation to treat each bunch as having a chemical potential $\mu(\nu)$ which depends only on the size ν , and a corresponding equilibrium density $\eta(\nu)$ of adatoms near the bunch. The velocity of the m th bunch may then be obtained by generalizing the result for single steps [19],

$$\frac{dx_m}{dt} = \frac{F}{2\nu_m}(x_{m+1} - x_{m-1}) + \frac{D}{\nu_m} \left(\frac{\eta(\nu_{m+1}) - \eta(\nu_m)}{x_{m+1} - x_m} - \frac{\eta(\nu_m) - \eta(\nu_{m-1})}{x_m - x_{m-1}} \right), \quad (5)$$

where ν_m is the size of the m th bunch.

At large F , all the bunches move forward in the growth direction, with smaller bunches moving more quickly because of the factor $1/\nu_m$. At very small F , however, large bunches move more quickly in the forward direction because of their lower chemical potential, and the smallest bunches can actually retreat. In any case, there will be continual collisions between different-size bunches, leading to coalescence, fragmentation, and/or exchange of steps. Thus it seems natural to expect a complex, chaotic evolution of the surface morphology.

For a stable configuration with long-range order, it might seem necessary that all steps be in bunches of a single size, so that no collisions need occur. However, there is a competition between energetics, which favors large bunches, and kinetics, which favors small bunches or single steps, to better incorporate the arriving atoms. Our simulations suggest that the system automatically balances these factors by alternating bunches and single steps.

This alternation is clearly seen in Fig. 1, especially in the better-ordered case in Fig. 1(b). The alternation is also reflected in Fig. 2(b). The first-order peak falls almost exactly on an integer spacing (in units of L_{av}), corresponding to four steps per period, i.e., one 3-step bunch and one free step per period. There is also a small shoulder on the right side of the first-order peak, reflecting the fact that there are a few “defect” bunches containing four steps. These defect bunches are presumably responsible for much of the damping and broadening of the high-order peaks in Fig. 2(b). This alternation of bunches and free steps is consistent with previous 2D simulations of Kandel and Weeks [14], if

one notices that for a given lateral position there is only one “crossing step” (the 2D analog of our single steps) between bunches.

This arrangement of alternating single steps and bunches has a special behavior that may account for its occurrence. Consider a periodic structure with one bunch and one single step per period. In this case, from Eq. (5) the velocities of all the bunches are the same, as are those of all the single steps, independent of the positions of the single steps relative to the bunches. So one may in effect have two coexisting periodic arrays, one of single steps and one of bunches, moving independently of each other.

There is one further ingredient needed to make the arrangement of alternating bunches and steps dynamically stable: the bunches must remain in a dynamic equilibrium with the free steps. Whenever a single step collides with a bunch, the bunch must emit a single step from its other side, so that the distribution of sizes is preserved. Thus a bunch of some size M must be stable for the given conditions, while a bunch of $M + 1$ is unstable and immediately loses a step. In that way, single steps appear to pass through the bunch, although the actual event is capture followed by emission of a different step. If this special configuration acts as a sort of dynamical “attractor,” it may account for the two-phase behavior observed for $F = 25$.

We emphasize that we do not yet understand the dynamics in detail, especially the “phase separation.” Nevertheless, we have demonstrated that considerable control of the ordering is possible. In actual growth, such control would probably require *in situ* diffraction measurements, so that flux or temperature could be varied to optimize the degree of order. The two-phase behavior, in particular, could simplify this procedure. In that case two distinct periods would coexist on the sample, and one could simply vary the growth conditions until the diffraction spot for one period disappears.

In conclusion, in step-flow growth of strained layers there is a competition between thermodynamic step bunching and kinetic debunching. This leads to a dynamic steady state, with finite-size step bunches exhibiting considerable uniformity and long-range order. Most important, the bunch size and spacing can be directly controlled and the ordering can be systematically improved by tuning the temperature and/or the flux. Through control of growth parameters, degree of stress, and choice of substrate miscut, it should be possible to create any desired size and spacing of step bunches. Such self-assembled and self-organized step-bunch arrays hold considerable promise as templates for subsequent fabrication of quantum wires and quantum dots.

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