Self-Organized Quantum-Wire Lattice via Step Flow Growth of a Short-Period Superlattice

Lugang Bai,[1] J. Tersoff,[2] and Feng Liu[1]
[1]*University of Utah, Salt Lake City, Utah 84112, USA*
[2]*IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598, USA*
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We develop a theoretical model for step flow growth of multilayer films, taking into account the interlayer step-step interaction induced by misfit strain. We apply the model to simulate the growth of strain-compensated short-period superlattices. Step-bunch ordering improves in successive layers, leading to self-organized growth of a lattice of quantum wires. This quantum-wire array has some similarities to the "lateral composition modulation" observed experimentally in short-period superlattices.

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Smooth epitaxial films are often grown by step flow on a vicinal surface, so that adatom attachment at step edges preempts island nucleation. Even in step flow growth, the film morphology may become rough due to step bunching. Early studies had focused on understanding the bunching mechanisms in an attempt to suppress step bunching. Recently, however, it has been recognized that self-organized step bunching can lead to relatively uniform step-bunch arrays, with potential applications for nanofabrication [1,2].

Theoretical models of step flow growth have generally been limited to the surface of a semi-infinite solid [1–7]. However, when the bunching is driven by strain-mediated step interactions (rather than kinetic factors), surface steps can interact with buried interface steps. This interaction is particularly important for thin layers or short-period superlattices.

Here, we examine theoretically the step dynamics when such interlayer interactions are included. We find that these interactions contribute to step bunching, and, more importantly, lead to correlations between successive layers. This opens the possibility of three-dimensional ordering. In particular, we simulate the growth of short-period superlattices with alternating tensile and compressive layers. We find strong long-range ordering of step bunches, not only within a layer, but also between layers. The resulting structure is, in effect, an ordered lattice of quantum wires, as seen in Fig. 1. This structure also has some similarities to the lateral composition modulation seen experimentally in short-period superlattices [8–13].

We begin with a 1D model for step flow growth at a surface under stress. Integration of the adatom diffusion equation with appropriate step boundary conditions leads to the following step velocity [1,3]:

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

where x_i is the position of the ith step, F is the adatom flux, and f_i is the force per unit length on the ith step. B is a constant related to the adatom diffusion coefficient and the adatom formation energy.

For a semi-infinite solid,

$$f_i = f_i^{(1)} = \sum_{j \neq i}\left(-\frac{\alpha_1}{(x_j - x_i)} + \frac{\alpha_2}{(x_j - x_i)^3}\right). \tag{2}$$

The first term is the misfit strain-induced long-range monopole-monopole attraction between steps [3]. The second term is the short-range dipole-dipole repulsion between steps. Here $\alpha_1 = C_s F_1 F_2$ is the interaction strength between two force monopoles (F_1 and F_2). For

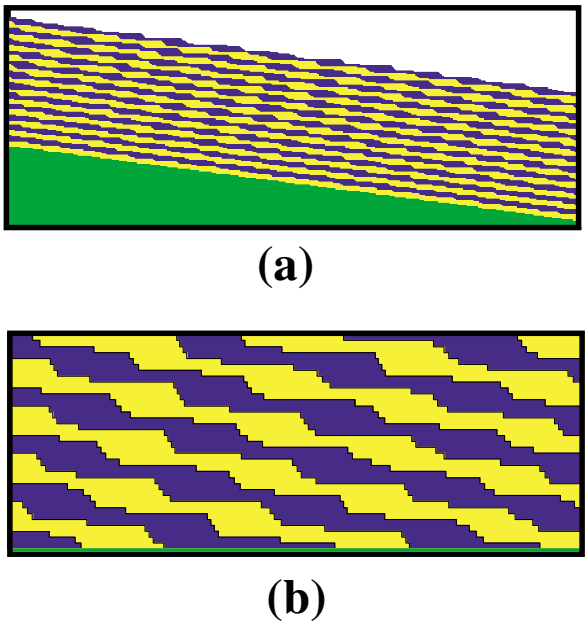

FIG. 1 (color online). Cross section of the final structure in our simulation, consisting of 12 bilayers of alternating layers of equal tension and compression, with each layer 4 ML thick. Vertical scale is expanded by ~10 times for clarity. (a) Entire system. (b) Expanded view of a particularly well-ordered region of (a), showing more clearly the individual steps. Note the excellent uniformity of bunch size and spacing.

two surface steps, $F_1 = F_2 = F_s = h_s \sigma_{xx}^{(A,B)}$, where h_s is the step height, σ_{xx} is the stress of the topmost layer (A or B) and $C_s = (1 - \nu^2)/(2\pi E)$, where ν is the Poisson ratio and E is Young's modulus. α_2 denotes the strength of the short-range interaction between the two surface dipoles.

For multilayer growth, we include also the force $f_i^{(2)}$ on surface steps due to buried interface steps:

$$f_i = f_i^{(1)} + f_i^{(2)} \tag{3}$$

and

$$f_i^{(2)} = \sum_{kl} \left\{ \frac{\alpha_l (x_{kl} - x_i)[(z_{kl} - z_i)^2 - (x_{kl} - x_i)^2]}{[(x_{kl} - x_i)^2 + (z_{kl} - z_i)^2]^2} - \frac{\beta_l (x_{kl} - x_i)}{(x_{kl} - x_i)^2 + (z_{kl} - z_i)^2} \right\}. \tag{4}$$

The force $f_i^{(2)}$ arises from the additional force on the ith surface step due to its elastic monopole-monopole interactions with the buried steps at the interfaces between previous layers. x_{kl} and z_{kl} denote the position of the kth step in the lth layer [14]. Here $\alpha_l = -C_l F_s F_l$ and $\beta_l = (1 - 2\nu)\alpha_l$, where $C_l = \frac{C_s}{4(1-\nu)}$ and F_s is the surface monopole. $F_l = \pm h_s(\sigma_{xx}^A - \sigma_{xx}^B)$ is the interface monopole at the lth buried layer, proportional to the stress *difference* between the two layers (A and B) at the buried step. (We neglect any difference between the elastic moduli and step heights of the two materials.) Obviously, the additional interlayer step-step interaction will influence the step flow growth of subsequent layers. It is particularly interesting to see whether such interaction can improve the self-organized ordering of the strain-induced step bunching.

In general there could also be dipole interactions between surface steps and buried steps. However, the step dipole arises largely from the reconstruction of the surface (and of the step itself), and will be much weaker at an interface step. For this reason, and because of the short-range nature, we neglect any dipoles at buried steps. In contrast, the interface-step monopole is equal to the difference between the surface monopoles for the respective layers, so for alternating tensile and compressive layers, the interface monopole is the sum (in absolute magnitude) of the two surface monopoles.

It is well known that various intermixing processes can occur at steps and interfaces during heteroepitaxy, and these can lead to a less abrupt interface. As long as the mixing does not extend through the entire layer, this broadens the monopole associated with each step, but does not affect its magnitude. If the step bunches are well separated laterally compared to this broadening, such intermixing should have little effect on the overall dynamics.

At a vicinal surface, the strain-induced monopole-monopole interaction between steps is always attractive [3]. In a multilayer growth, the interlayer monopole-monopole interaction may be either attractive or repulsive, even when the monopoles have the same sign, depending on the step-step separation. From Eq. (4), we deduce that the lateral force in the x direction between the ith step on the surface and the kth step in the lth buried layer is attractive (repulsive) beyond the range of $|\Delta x| > \sqrt{\nu/(1-\nu)}|\Delta z|$ but repulsive (attractive) within, if the two monopoles point at the same (opposite) direction, where $\Delta x = x_i - x_{kl}$ is the lateral separation between the two steps, and $\Delta z = z_i - z_{kl}$ is the vertical separation [14]. The range of attraction and repulsion between a surface step and a buried step depends on the depth of the buried step. Thus, the effect of buried steps on the growth and ordering of surface steps can be tuned by changing layer thickness.

We apply this model to investigate a strategy for growth of a self-organized lattice of quantum wires. We consider a strain-compensated superlattice, where the layers all have equal thickness, and the two alternating materials have tensile and compressive misfit of equal magnitude. (This could be, e.g., layers of $In_xAl_{1-x}As$ of different x, grown on an InP substrate.) Consequently, the total stress is zero; as long as the individual layers are thin, there should be no tendency to introduce misfit dislocations. Then the interlayer interaction can drive the surface step bunches to align with the step bunches at buried interfaces, forming a three-dimensional array of step bunches (a two-dimensional lattice in cross section). If the superlattice layers are thin compared to the bunch size, the result is a lattice of quantum wires.

This behavior is clearly seen in Fig. 1. For this simulation, we use a simulation cell containing 120 steps in each layer, with periodic boundary conditions. The average step spacing is $L_{av} = 52$, with a step height of 0.5, corresponding to a vicinal surface of 0.55° miscut. Other parameters are $B = 1.0$, $\alpha_1 = 1.0$, $\alpha_2 = 100$, and $\nu = 0.28$. We start with a substrate having random step distribution, and grow the first strained A layer using the same procedure as in earlier work [1]. Then we continue with the overgrowth of the first strain-compensating B layer. The cycle of alternating A and B growth is repeated to form a multilayer film, and the surface steps interact with the buried steps in all previous layers.

During the growth, steps at the surface (whether A or B) are always under a long-range attractive interaction, due to strain-induced elastic monopoles at the surface steps. Their interaction with the buried steps underneath is long-range attractive and short-range repulsive between the same types of layers (i.e., between A-A or B-B layers), while the opposite is true between the different type of layers (i.e., between A-B or B-A layers). This is because force monopoles at steps have opposite signs for the two types of layers. For our specific parameters, the interlayer step-step interaction between two different type layers is attractive for $|\Delta x| < 0.62|\Delta z|$ and repulsive for $|\Delta x| > 0.62|\Delta z|$. Also, the magnitude of monopoles at the buried steps are twice as large as those at the surface steps.

Figure 1 shows a film cross section that resulted from the evolution of simulated step configurations of a 12-bilayer multilayer film, using a flux of $F = 38$. The thickness of each layer is chosen to be 4 monolayers (MLs). The periodic lateral thickness modulation of the A and B layers, due to step bunching and ordering, forms automatically a lattice of quantum wires. This is clearer in Fig. 1(b), which gives an expanded view of a particularly well-ordered region from Fig. 1(a).

The modulation in Fig. 1 is also strongly reminiscent of the lateral composition modulation that has been observed experimentally in several compound semiconductor systems [8–13]. In both cases, the local composition averaged over alternating A and B layers corresponds to A-rich and B-rich columns (or sheets in 3D). We believe that the basic driving force is the same in both cases—the strain-mediated interaction between the morphology of the surface and of the buried interfaces [15]. However, the systems studied experimentally are presumably not grown in step flow mode, and the columns are roughly vertical. In our simulations, the columns are at an angle to the surface normal, reflecting the role of step flow dynamics.

The ordering mechanism and process observed here are similar to those found in step flow growth of a single-layer strained film [1]. As in that case, there is a competition between strain-induced bunching and flux-induced debunching. The flux-induced debunching becomes progressively more important with increasing bunch size. As a result, the bunch size initially increases with time, but this leads to stronger debunching, and eventually the system reaches a steady-state bunch size determined by the competition between strain-induced step bunching and flux-induced debunching. Because we have thin layers, the bunching requires several layers to reach a steady state. (In multilayer growth, the meaning of "steady state" is slightly different than for a simple surface, because the degree of bunching must vary between the beginning and the end of growth of each layer. This may also affect the degree of order, since optimal ordering is obtained when the average bunch size is an integer [1].)

We note that the interlayer interactions effectively increase the driving force for step bunching in the strain-compensated system, so that to obtain a given bunch size, a larger flux is needed for the multilayer system. For example, for the case shown in Figs. 1 and 2, the average bunch size is four steps, the same as achieved in a single-layer film in Ref. [1]. However, a larger flux of 38 is needed for the multilayer growth than 30 for the single-layer growth [1], because effectively a larger step-step attraction is present in the multilayer film, resulting from the additional interlayer step-step interactions. Such a condition is further confirmed by the growth of multilayer films with either too large a flux ($F = 40$) or too small a flux ($F = 30$); both lead to a decrease of bunch ordering because they result in a noninteger average bunch size, the former less than 4 and the latter larger.

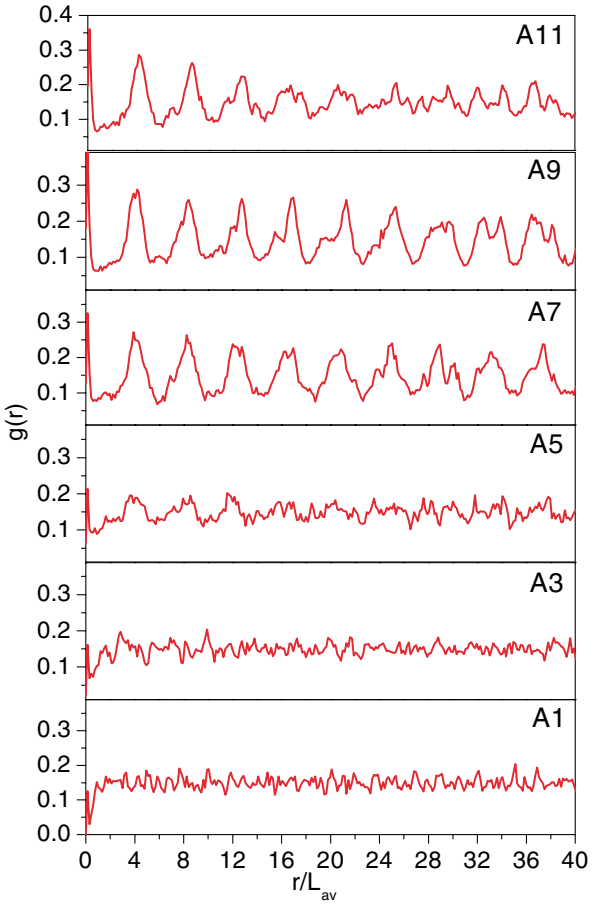

FIG. 2 (color online). The pair correlation function of the steps at the surface of A layers in A-B superlattice, for the $A1$, $A3$, $A5$, $A7$, $A9$, and $A11$ layers of the 12-bilayer film of Fig. 1. The results are each averaged over ten configurations well separated in time. Note the progressive improvement of ordering during growth of successive layers.

As the step bunches grow with successive layers, they also become increasingly ordered, and good ordering is achieved at around the same point where the bunches reach their asymptotic size. The improvement of step-bunch ordering with successive layers can be clearly seen in the step pair correlation functions shown in Fig. 2. By the seventh bilayer (i.e., seventh superlattice period, an overall growth of 56 ML), step bunches self-organize into an array with an average bunch size of four steps and very good long-range order. The good bunch order is then maintained in all the subsequent layers. The same behavior is observed for the B layers.

The 2D quantum-wire superlattice exhibits a striking degree of order. This is illustrated by the plots of 2D correlation function for steps from the 7th to the 11th layer shown in Fig. 3. The bunches in these layers are maintained with four steps, displaying very good long-range order in both the x and z directions. This is

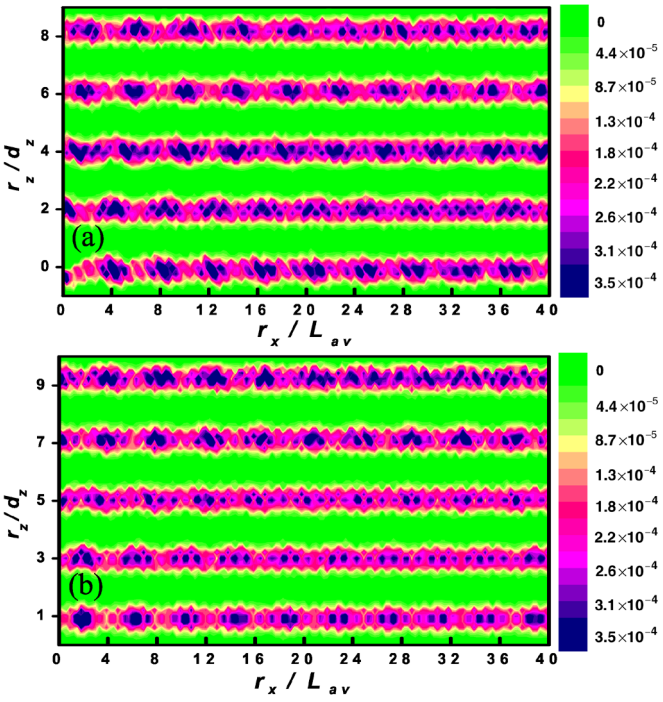

FIG. 3 (color online). 2D correlation function of steps from the 7th to the 11th layer, illustrating the formation of a 2D array of step bunches. (a) Between the same type of steps (*A*-*A*) + (*B*-*B*). (b) Between the different type of steps (*A*-*B*) + (*B*-*A*).

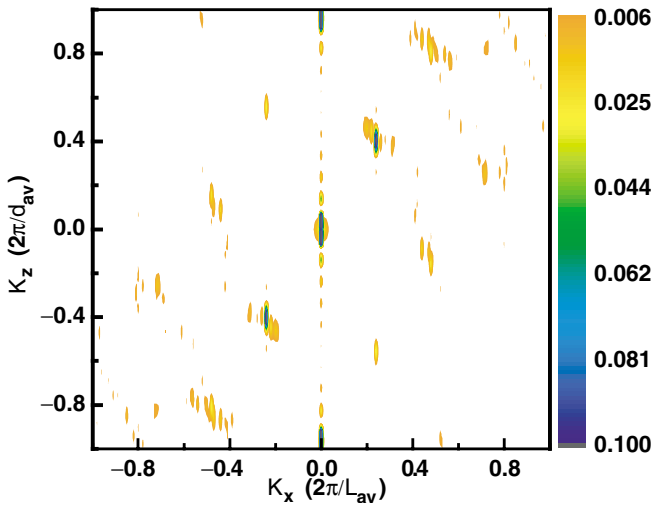

FIG. 4 (color online). Structure factor of steps from the 7th to the 11th layer, showing the high degree of symmetry and order of the 2D array of step bunches.

indicated by the equally spaced packed peak positions, with a lateral separation of $4L_{av}$ (4 times of average step separation) and a vertical separation of d_{av} (layer thickness), and by the well-defined high-order peaks. The high degree of symmetry of the 2D superlattice is shown by the plot of structure factor in Fig. 4, illustrating again the excellent ordering.

While we have used layers of equal thickness here, equally good strain compensation can be obtained with, e.g., one layer twice as thick but with half the misfit of the other layer. Thus the same approach could be used to obtain more isolated wires of the larger misfit material. This could be, e.g., InAs wires in an $In_xAl_{1-x}As$ matrix on an InP substrate, for optoelectronic applications. Also, different growth rates could be used for the respective materials, allowing independent control of the degree of step bunching. Thus there are many opportunities for tuning the self-organization to achieve a desired structure.

In conclusion, we have developed a theoretical model for step flow growth of strained multilayer film, taking into account strain-induced interlayer step-step interaction. We demonstrate that such interaction can help to progressively improve the step-bunch ordering in the growth of a multilayer film consisting of different layer sequences and thicknesses. Furthermore, we apply the model to simulate growth of strain-compensated multilayer films of alternating compressive and tensile strained layers, which provides a potentially useful method for fabricating quantum-wire superlattice. The self-organized step bunches with uniform size and spacing generate a quantum-wire superlattice with good long-range order. The method also effectively avoids dislocation formation and allows flexible choice of materials.

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