Kinetic step bunching instability during surface growth

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We study the step bunching kinetic instability in a growing crystal surface characterized by anisotropic diffusion. The instability is due to the interplay between the elastic interactions and the alternation of step parameters. This instability is predicted to occur on a vicinal semiconductor surface Si(001) or Ge(001) during epitaxial growth. The maximal growth rate of the step bunching increases like $F^4$, where $F$ is the deposition flux. Our results are complemented with numerical simulations which reveals a coarsening behavior on the long time for the nonlinear step dynamics.

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The field of surface growth on semiconductors is very active due to its importance for both technological applications and fundamental science [1, 2, 3]. Under nonequilibrium growth a variety of experiments reveal rich crystal morphologies resulting from the nonlinear evolution of step flow instabilities [4, 5]. The kinetic and elastic effects drive the system towards self-organized states, characterized by the appearance of ordered structures on the vicinal surfaces. This self-organization can be exploited in semiconductor nanotechnology, for the development of devices having interesting quantum properties [6, 7]. A basic mechanism for pattern formation in vicinal semiconductor surfaces is the step bunching instability [8, 9, 10], whose origin is commonly attributed to the presence of impurities, to the inverse Ehrlich-Schwoebel effect, or to electro-migration (see [11, 12] and references therein). This Letter is motivated by the molecular beam epitaxy (MBE) experiments on Si(001) in which it was observed a new type of kinetic instability leading to the formation of step bunches [13, 14, 15]. Microscopic kinetic Monte-Carlo simulation showed that in the case of Si(001), step bunching is due to the coupling between diffusion anisotropy and differences in step kinetic parameters [16]. Here we provide a macroscopic instability mechanism for the step bunching instability that does not require any inverse Ehrlich-Schwoebel effect. We show that the interplay between the elastic step interactions and the alternation of kinetic parameters, characteristic of the Si(001) vicinal surface, induces a finite wavelength instability with maximal growth rate increasing as $F^4$ ($F$ is the deposition flux). Our results are complemented by numerical simulations which reveal a coarsening behavior on the long time for the non-linear step dynamics.

The Si(001) vicinal surface consists of a periodic sequence of terraces where rows of 2 × 1 dimerised adatoms (terrace of type $a$) alternate with 1 × 2 dimerised adatoms (terrace of type $b$), see Fig. 1. On the reconstructed surface adatoms diffuse preferentially along dimer rows, giving rise to an anisotropic diffusion. Therefore, the steps separating the terraces are of two kinds. The $S_a$ step is rather straight while the $S_b$ step is very corrugated [17].
boundary conditions at each step:

\[ C_n^a(x_n^{a-1}) = C_{eq,b} \]
\[ D_a \frac{\partial C_n^a}{\partial x}(x_n^a) = -\nu [C_n^a(x_n^a) - C_{eq,a}] \]
\[ D_b \frac{\partial C_n^b}{\partial x}(x_n^b) = \nu [C_n^b(x_n^b) - C_{eq,a}] \]
\[ C_n^b(x_n^b) = C_{eq,b} + 1 \]

where \( \nu \) is the step kinetic coefficient of step \( a; C_{eq,a} \) and \( C_{eq,b} \) are the adatom equilibrium concentration at each step. The velocity of each step is given by the conditions, \( v_n^a = \Omega D_b \frac{\partial C_n^b}{\partial x}(x_n^a) - \Omega D_a \frac{\partial C_n^a}{\partial x}(x_n^a) \)
\( v_n^b = \Omega D_a \frac{\partial C_n^a+1}{\partial x}(x_n^b) - \Omega D_b \frac{\partial C_n^b}{\partial x}(x_n^b) \),

with \( \Omega \) the unit atomic surface. The adatom equilibrium concentrations are determined by the elastic interactions between steps mediated by the terrace reconstruction 18 20 21.

\[ C_{eq,b} = C_0 + E \left( \frac{1}{l_n^b} - \frac{1}{l_n^{a-1}} \right) \]
\[ C_{eq,a} = C_0 + E \left( \frac{1}{l_n^a} - \frac{1}{l_n^{b-1}} \right) \]

where \( E = \Omega C_b A/k_B T \) (\( C_b \) is the uniform equilibrium concentration, \( k_B \) the Boltzmann constant, \( T \) the temperature, and \( A \) is an energy per unit length related to the stress and elastic constants of the medium 21). The system behavior is controlled by three independent nondimensional parameters: \( f = F l_0^2/\nu E \), \( \alpha_a = \nu l_a/D_a \) and \( \alpha_b = \nu l_b/D_b \). We set the unit length to be the initial size of terrace \( a, l_n^a = 1 \). With this convention the velocity is measured in units of \( \nu E \Omega/l_n^a \). Under normal experimental conditions (\( \Omega F < 1 \text{s}^{-1}, A \approx 5 \times 10^{-12} \text{Jm}^{-1}, \Omega C_0 \approx 10^{-2} \) \( f \ll 1 \), \( \alpha_a \gtrsim 1 \) and \( \alpha_b \ll 1 \).

We first look for a uniform train of steps with \( l_n^a = 1 \) and \( l_n^b = l_b \) for all \( n \), traveling at a constant velocity. The relative velocity is

\[ v_b - v_a = \frac{4 - \nu l_b(4 - f)}{\nu l_b(1 + \alpha_a) + 4 - \nu l_b(4 - l_b f)} \]

It vanishes for terraces of size \( l_b \) given by

\[ l_b^\pm = 2 l_b = 2 \frac{2 - f/4 + \alpha_a - \alpha_b \pm \sqrt{\Delta}}{f(1 + \alpha_a + \alpha_b - 4 \alpha_b/f)} \]

where \( \Delta = (2 - f/4 + \alpha_a - \alpha_b)^2 - f(2 + \alpha_a)(1 + \alpha_a + \alpha_b - 4 \alpha_b/f) \). Depending on the values of the parameters (\( \alpha_a, \alpha_b, f \)) different situations may arise. The two solutions \( l_b^\pm \) represent terraces of nearly equal size (\( l_b^\pm \)), or double steps (\( l_b^\pm \)) for which terraces of type \( a \) almost disappear (Fig. 2). These two branches exist (\( \Delta > 0, l_b > 0 \)) for fluxes lower than \( f_c \). The upper branch characterizing the double state exists in the interval \((f_c^+, f_c)\), as shown in Fig. 2. This interval vanishes for the values of \( \alpha_b \) satisfying \( \alpha_b^2 > (1 + \alpha_a)(2 + \alpha_a) \).

We investigate now the stability of a train of steps traveling at the constant velocity \( V = v_a(l_b) = v_b(l_b) \), where \( l_b \) is one of the solutions \( l_b^\pm \). The perturbed step positions are given by \( x_n^a = V t + n(l_b + X_n^a) \) and \( x_n^b = V t + n(l_b + l_b + X_n^b) \). We expand the step velocities up to first order in the perturbation \( X_n^{a,b}(t) = X_{a,b} \exp(st + i k t) \), where \( s \) is the (complex) growth rate and \( k \) the wavenumber. After some algebra, we obtain the following expression for the growth rate \( s = \sigma + i \omega \), in the limit of small \( f \) and \( k \), for the lower branch \( l_b^- \),

\[ \sigma = A f^2 k^2 - B k^4, \quad \omega = \frac{1}{2} f(k - C f k^3) \]
where

\[
A = \frac{1}{16} \frac{(\alpha_a + \alpha_b + 3)}{\alpha_a + \alpha_b + 2}^3
\]

\[
B = \frac{1}{1536} \frac{1}{\alpha_a + \alpha_b + 2}^3 \times
\]

\[
[768(1 + \alpha_a + \alpha_b) + 192(\alpha_a + \alpha_b)^2 -
24(8 + 6\alpha_a + 14\alpha_b + \alpha_a^2 + 6\alpha_a\alpha_b + 5\alpha_b^2)f -
(72 + 86(\alpha_a + \alpha_b) + 19\alpha_a^2 + 80\alpha_a\alpha_b - 11\alpha_b^2)f^2]
\]

\[
C = \frac{1}{384} \frac{8\alpha_a + 8\alpha_b + 16 - 9(\alpha_a - \alpha_b)f}{2 + \alpha_a + \alpha_b}.
\]

Figure 4 shows the real and imaginary parts of \(s\) for typical values of the parameters. The imaginary part \(\omega = \Im s \sim f k\) is associated with the propagation of compression waves. The upper branch \(l_f^+\) displays a similar behavior, showing that both uniform solutions are unstable. The growth rate \(\sigma = \Re s\) of this instability increases like \(f^2 k^2\) for small \(k\) and is stabilized at larger \(k\) by elastic effects (independent of \(f\)). The most unstable mode \(k_{\text{max}}\) increases linearly with the flux \(k_{\text{max}} \sim f\), and its corresponding growth rate scales like \(\sigma_{\text{max}} \sim f^4\), as can be seen from Eq. (12) and illustrated in Fig. 4. Therefore, for small \(f\), the instability is almost vanishing. We have checked that this result is not qualitatively changed if a finite kinetic coefficient \(\nu_b\) for the \(b\) step is included into the model. In the particular case \(\nu_a = \nu_b\), the growth of the step flow instability vanishes, independently of the \(D_a/D_b\) ratio.

This step bunching instability is a consequence of kinetic and elastic coupling under crystal growth conditions. The asymmetry of kinetic coefficients, related to the alternation of smooth and rough steps, introduces a supplementary elastic coupling between non-neighboring terraces. Their combined action breaks the symmetry between the ascending and descending adatoms currents.

In contrast to the usual inverse Ehrlich-Schwoebel effect, the unbalanced currents concern the interaction between two double terraces.

We can obtain the non-linear evolution of the step positions by numerical integration of the set of differential equations (13-15). These equations can be written explicitly using (1) and its associated boundaries conditions.
where \( \nu^0 \equiv \mu^0_a + \frac{\mu^0_b f l_b^0}{1 + \alpha_b l_b^0} \) and \( \nu^b \equiv \mu^b_a + \frac{\mu^b_b f l_b^{n+1}}{1 + \alpha_b l_b^{n+1}} \),

\[
\nu^a_e = \mu^a_a + \frac{\mu^a_b f l_b^n (2 + \frac{3 \alpha_a l_a^n}{2})}{1 + \alpha_a l_a^n} - \nu^0_a \quad \text{and} \quad \nu^b_e = \mu^b_a + \frac{\mu^b_b f l_b^n (1 + \frac{3 \alpha_a l_a^n}{2})}{1 + \alpha_a l_a^n} - \nu^0_b,
\]

The typical evolution of the step flow on the vicinal surface is shown in Figs. 5 and 6. We choose as initial conditions different sets of slightly perturbed equal size terraces. The amplitude of the perturbations increases during the evolution while compression waves propagate as predicted by the linear analysis. The further evolution of the system shows the formation of bunches of steps separated by wider terraces. A coarsening regime sets in, characterized by the coalescence of these step bunches into larger ones. Ultimately the long time evolution leads to a surface composed of a single macro-step. We have checked that these results are not affected by the choice of initial conditions or by the size of the system. This behavior is illustrated in Fig. 3 by numerical simulations depicting the step train at two different times for \( f < f_c \). We have also checked that this scenario remains valid when \( f > f_c \). Figure 3(a) shows a modulated step pattern characteristic of the incipient formation of step bunches. Figure 3(b) shows the further evolution of these bunches during the coarsening regime. A variety of models of step bunching shows that the mean number of steps in a single bunch obeys a power law \( \langle n(t) \rangle \sim t^\beta \).

In the present case this behavior is expected to be more complex due to the presence of oscillations in the step size. This point deserves a specific investigation using a continuous description of the step dynamics, more appropriated for the study of the long time evolution.

In conclusion, we have proposed a kinetic mechanism of step bunching instability under non-equilibrium growth of a vicinal surface of Si(001)-like crystals. The essential ingredients are the alternation of rough and smooth steps, having different diffusion and attachment coefficients, and the coupling of non-neighboring terraces by elastic interactions. We have found that the rate of growth of the instability scales like \( t^4 \). It would be interesting to experimentally test this behavior, using for example measurements of the surface roughness evolution. Some indirect evidence supporting the present instability mechanism are provided by experiments on Si/Ge growth \[22\]. The addition of a small quantity of Ge atoms produces a substantial modification of the steps properties, and in particular shades off the distinction between rough and smooth steps. As a consequence, a stable step flow regime sets in even though the diffusion coefficient of successive terraces are still different. A generalization of the present model to two dimensions is currently under progress. We expect a rich phenomenology due to the coupling between a meandering instability induced by the alternation of the step parameters \[22\], the step bunching instability and the anisotropy of surface diffusion \[24\].

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