

## Scaling and Universality of Self-Organized Patterns on Unstable Vicinal Surfaces

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We propose a unified treatment of the step bunching instability during epitaxial growth. The scaling properties of the self-organized surface pattern are shown to depend on a single parameter, the leading power in the expansion of the biased diffusion current in powers of the local surface slope. We demonstrate the existence of *universality classes* for the self-organized patterning appearing in models and experiments.

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The scale invariance of the fluctuations of growing or evaporating surfaces, known as kinetic roughening [1,2], has been extensively studied theoretically for both high-symmetry [3] and vicinal surfaces [4]. These surfaces are often found to be unstable [5]. In particular, vicinal surfaces may undergo a kinetic *step bunching* instability [6–9], when the regular step train decomposes into alternating low and high step-density regions, and then evolves into a self-organized, scale invariant pattern [10]. These patterns may be ideally used as a template for building anisotropic nanostructures, such as quantum wires. Hence, besides the fundamental interest, understanding the microscopic origin of step bunching is crucial for nanotechnology applications. Several scenarios for kinetic step bunching during growth or evaporation have been investigated: electromigration [11], Schwoebel barriers (in evaporation) or “inverted” Schwoebel barriers (in growth) at step edges [12], multispecies coupling by chemical reactions [13], dimer formation [14], and diffusion along step edges [15]. Using the continuum approach [10], we show that independently of the microscopic origin, the overall scaling behavior of the surface profile is uniquely determined by the nonequilibrium *biased diffusion current*, first discussed by Villain [5]. Specifically, the surface evolution is shown to depend on a single parameter, the power  $\rho$  of the leading term in an expansion of the nonequilibrium current in powers of the local surface slope. Since this expansion is dictated by the microscopic behavior of the system, the investigation of the scaling of step bunching reveals which class of microscopic models the macroscopic features observed belong to. This is the analog of the *universality classes* of equilibrium critical phenomena [16]. Step bunching is then characterized by scaling exponents, that take specific values for each universality class. The present work addresses all situations in which the current is a function of the local surface slope. This includes all the scenarios mentioned above, as well as impurity induced bunching. In the case when elastic constraints become important, as in heteroepitaxy, the current may be a nonlocal function of the slope, and/or of the surface curvature. Such situations may be treated by a similar approach as the one used here, but they fall outside

the scope of the present work. The examples studied in the present Letter cover both models and experiments, and reveal the existence of three universality classes for the self-organized patterning of unstable vicinal surfaces.

In the framework of the continuum approach [10], the crystal surface is described as a smooth function  $h(x, t)$ , whose time evolution is governed by an equation of the form

$$\frac{\partial h}{\partial t} = \Phi - \nabla \cdot \mathbf{J}, \quad (1)$$

where  $\Phi$  is the deposition rate, and  $\mathbf{J}$  the surface diffusion current. The latter contains in general a smoothing “equilibrium” part, and a destabilizing “nonequilibrium” part. The former is due to the Gibbs-Thomson effect [2]. Indeed, the surface free energy of a crystal at a vicinal orientation (next to a singular, nonrough facet) reads  $\mathcal{F} = \int dS \phi(|m_x|)$ , where  $m_x = \partial h / \partial x$  is the local slope, and the surface free energy density  $\phi$  has the form

$$\phi(m_x) = \phi_0 + \gamma|m_x| + A|m_x|^{n+1}. \quad (2)$$

The local surface slope  $m_x$  is equal (in absolute value) to the local step density  $1/\ell$ ,  $\gamma$  is the step stiffness,  $A/\ell^n$  is the step-step interaction potential, and  $\ell$  is the interstep distance. The  $n$  power generalizes the usual  $1/\ell^2$  form of the interaction that applies to entropic and elastic repulsion as well. Assuming that the steps are straight, any local deviation from a perfect staircase of equidistant steps is given by the Gibbs-Thomson formula  $\Delta\mu = -\partial\phi'/\partial x$  [2]. For a vicinal surface of positive slope,  $m_x > 0$ , Eq. (2) yields the excess chemical potential  $\Delta\mu = -(n+1)A\partial(m_x^n)/\partial x$ . Thus, the step-step interactions smooth out the fluctuations of the interstep distances, by creating a surface particle current

$$\mathbf{J}_{\text{eq}} = -\frac{D_s c_{\text{eq}}}{k_B T} \nabla(\Delta\mu) = \frac{D_s c_{\text{eq}}}{k_B T} (n+1)A\nabla^2(m_x^n). \quad (3)$$

The latter replaces the “Mullins-type” curvature driven current, that is appropriate for a surface above the roughening temperature [2].

The instability appears due to kinetic, nonequilibrium effects that induce a *biased* diffusion current,  $J_{\text{neq}}(m_x)$ ,

that has to be computed from a microscopic model. Whatever its form, letting  $J \equiv J_{\text{neq}}(m_x)$  in Eq. (1), and linearizing it for small fluctuations about a uniform positive slope  $m_0$ , that is for  $h(x, t) = m_0x + \epsilon(x, t) + \Phi$ , yields

$$\frac{\partial \epsilon}{\partial t} = \nu_{\perp} \frac{\partial^2 \epsilon}{\partial x^2} \quad (4)$$

where  $\nu_{\perp} = -[dJ_{\text{neq}}(m_x)/dm_x]_{m_0}$ . Since Eq. (4) with a *negative* coefficient  $\nu_{\perp}$  implies an unstable increase of the fluctuations, it appears that a vicinal surface growing by step flow is unstable with respect to step bunching if

$$[dJ_{\text{neq}}(m_x)/dm_x]_{m_0} > 0. \quad (5)$$

The continuum description allows us to give a unified treatment of the step bunching instability for all systems characterized by a nonequilibrium current which is a local function of the slope. In particular, the scaling properties of the time evolution, as well as of the long-time self-organized bunch structure, can be computed. For these systems, the destabilizing, nonequilibrium current has the general form

$$J_{\text{neq}} = m_x J(m_x), \quad (6)$$

where  $J(m_x)$  is determined by the specific kinetics of the system. In general,  $J(m_x)$  vanishes with vanishing terrace width, and tends to a constant for a flat surface. The current induced by step-edge diffusion (SED) may differ slightly (see below). In the spirit of Landau's theory of critical phenomena [16], we expand the current in powers of  $m_x$  for small slopes, or of  $1/m_x$  for large slopes, and consider the leading term. In both cases, the current behaves then as  $J_{\text{neq}} \sim J_0 + Bm_x^{\rho}$ , with  $J_0 = 0$  (SED may differ having  $J_0 \neq 0$ ), and  $\rho$  taking either positive or negative values for the small and large slope limit, respectively. Since only the divergence of the current enters the growth equation, we can disregard the constant anyway and write, without loss of generality:

$$J_{\text{neq}} = Bm_x^{\rho}, \quad (7)$$

with  $B\rho > 0$  to satisfy the instability condition Eq. (5) for  $m_x > 0$ . Note that the *sign* of the nonequilibrium current itself determines the stability of the vicinal surface with respect to step meandering, and eventually mounding [10]. A growing (evaporating) surface of positive average slope is stable against meandering if  $J_{\text{neq}} < 0$  ( $J_{\text{neq}} > 0$ , respectively). Then, in general, a positive coefficient  $B$  in Eq. (7) implies that the growing (evaporating) surface is simultaneously unstable against step meandering and step

bunching if  $\rho > 0$  ( $\rho < 0$ , respectively). Again, SED is exceptional, since the sign of the constant  $J_0$  determines the stability with respect to meandering in this case. Putting together the equilibrium and the nonequilibrium currents, and removing the constant deposition rate by transforming to a uniformly comoving frame, we obtain the generic evolution equation for an unstable vicinal surface:

$$\frac{\partial h}{\partial t} = -B \frac{\partial}{\partial x} m_x^{\rho} - \frac{D_s c_{\text{eq}}}{k_B T} (n+1) A \frac{\partial^3}{\partial x^3} (m_x^n). \quad (8)$$

Differentiation of both sides with respect to  $x$  finally yields the evolution equation of the local step density  $m_x$ :

$$\frac{\partial m_x}{\partial t} = -B \frac{\partial^2}{\partial x^2} m_x^{\rho} - \frac{D_s c_{\text{eq}}}{k_B T} (n+1) A \frac{\partial^4}{\partial x^4} (m_x^n). \quad (9)$$

Now, we look for scale-invariant solutions of Eq. (9) [1]. Scale invariance means that, if we compare two bunches whose widths differ by a factor  $\lambda$ ,  $W_2 = \lambda W_1$ , the respective bunch heights  $h_1, h_2$  differ by a factor  $\lambda^{\alpha}$ . In particular, if the scaling factor  $\lambda$  depends on time,  $\lambda = t^{1/z}$ , so that the bunch width grows with time as  $W \sim t^{1/z}$ , the bunch height also grows as  $h(\lambda t) \sim \lambda^{\beta} h(t)$ . These relations define the exponents  $\alpha, z$ , and  $\beta = \alpha/z$ , and can be summarized in the scaling form  $h(x, t) = t^{\alpha/z} H(x/t^{1/z})$ . The exponents are then determined by requiring Eq. (9) to be independent of  $\lambda$ , after rescaling of  $x, t$ , and  $h$ . For the physically most interesting case  $n = 2$  we obtain

$$\alpha = \frac{4 - \rho}{2 - \rho}; \quad z = 2 \frac{3 - 2\rho}{2 - \rho}; \quad \beta = \frac{1}{2} \frac{4 - \rho}{3 - 2\rho}. \quad (10)$$

The form of the exponents for any  $n$  is given in Table I. To describe the scaling of the local bunch slope  $m_x$ , it is convenient to introduce another exponent,  $\delta$ . Since  $m_x \sim x^{\alpha-1} \sim t^{(\alpha-1)/z} \equiv t^{\delta}$ , we obtain  $\delta = 1/(3 - 2\rho)$ . When the instability evolves into a self-organized state, the current may be made to vanish by a stationary surface profile  $h_0(x) = x^{\alpha}/\Theta$ , where the dimensionless coefficient  $\Theta$  contains the physical parameters. Inserting this scaling form into Eq. (8) and letting  $\partial h/\partial t = 0$ , we obtain  $\Theta \approx [6D_s c_{\text{eq}} A / (k_B T B)]^{1/(4-\rho)}$ . Another scaling relation that is quite important for the analysis of experimental results concerns the average distance between steps in a bunch,  $\ell_b$ , as a function of the number of steps in a bunch,  $N$ . If we assume that  $\ell_b$  scales like  $1/m_x$ , and define the exponent  $\gamma$  through  $\ell_b \sim N^{-\gamma}$ , we find  $\gamma = 2/(4 - \rho)$ .

TABLE I. Predicted exponents for the observed universality classes of step bunching, and for general step-step interactions.

	$\rho = -2$	$\rho = -1$	$\rho = 1$	$\rho, n$
$\alpha$	3/2	5/3	3	$(2 + n - \rho)/(n - \rho)$
$\beta$	3/7	1/2	3/2	$(2 + n - \rho)/[2(n + 1 - 2\rho)]$
$z$	7/2	10/3	2	$2(n + 1 - 2\rho)/(n - \rho)$
$\delta$	1/7	1/5	1	$1/(n + 1 - 2\rho)$
$\gamma$	1/3	2/5	2/3	$2/(2 + n - \rho)$

We conclude that all the exponents are uniquely determined once  $\rho$  is chosen. The determination of the exponents allows us to find out the universality class, that is the value of  $\rho$  and the leading behavior of the nonequilibrium current, for the system under scrutiny. In order to show how this may work, we analyze a number of situations where step bunching has been investigated in detail, either experimentally in real system, or in models. We will see that only *three universality classes* for step bunching seem to emerge, corresponding to  $\rho = -2, -1, 1$ , respectively. The scaling exponent values for these universality classes in case of  $n = 2$  are summarized in Table I.

*The  $\rho = -2$  universality class.*—It is well known that at least in metal systems, SED may be the fastest kinetic process. We follow Politi and Krug [15] in arguing that the SED would induce a destabilizing (uphill) current, independent of the slope in the large-slope limit, and vanishing linearly with  $m_x$  in the small-slope limit [15]. The characteristic length scale  $\ell_c$  that separates large from small slopes, is identified as the distance between islands on terraces: in other words, when  $m_x < 1/\ell_c$ , step flow is replaced by island nucleation on terraces. In the intermediate region between the large- and small-slope limit we propose an interpolation formula for the biased diffusion current based on the results of Ref. [17], that differs from Politi and Krug's [15]. It reads  $J_{\text{sed}}(m_x) = B\ell_c|m_x|\tanh[1/(\ell_cm_x)]$ . Note that step flow is always unstable towards bunching, since  $dJ_{\text{sed}}/dm_x$  is always positive. In the step flow regime  $m_x > 1/\ell_c$ , the current behaves asymptotically as  $J_{\text{sed}}(m_x) \sim J_0 - m_x^{-2}$ , so that  $\rho = -2$  is expected in this case. To check the corresponding exponents (see Table I), we have performed kinetic Monte Carlo (KMC) simulations, whose details will be reported elsewhere, of step-flow growth in the presence of strong SED. Step bunching is indeed observed. Following Ref. [18], we compute the mean distance between steps in the bunches,  $\ell_b$  as a function of the deposition time. The result is shown in Fig. 1 (circles), it compares well with the power-law behavior with  $\delta = 1/7$  of Table I.

*The  $\rho = -1$  universality class.*—Until recently, it was believed that kinetic bunching during growth or evaporation might only be a consequence of the asymmetric sticking at steps, as first discussed by Schwoebel [12]. Indeed, assume that a diffusing particle attaches to a step with a probability that is larger from the upper step side than from the lower one (downhill diffusion bias). This assumption is usually called “inverse Schwoebel effect” (ISE). On the opposite, when the incorporation probability of a particle at a step is reduced from the upper side, with respect to the lower side, one has the uphill diffusion bias and “normal Schwoebel effect” (NSE). It is straightforward to show that the ISE leads to step bunching during growth, while the NSE leads to step bunching during evaporation of the step train. When the ISE and the NSE are very strong, the biased diffusion current behaves as  $J_{\text{SE}}(m_x) \sim m_x^{-1}$  [10], yielding  $\rho = -1$ . The same  $m_x^{-1}$  form of the biased diffusion current has been found by Stoyanov [19] for electro-

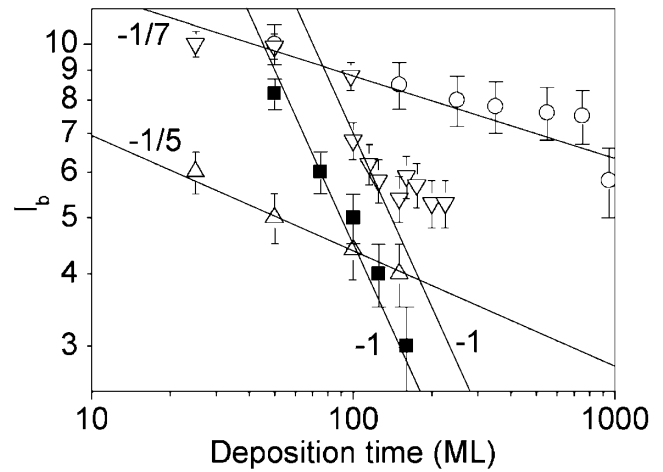


FIG. 1. The average terrace width inside a bunch,  $\ell_b$ , as a function of the deposition time, computed from KMC simulations assuming different microscopic models of epitaxial growth. (i) Step-edge diffusion (open circles). (ii) Inverse Schwoebel effect (open triangles). (iii) Two-particle model (open inverted triangles). (iv) Step-edge diffusion and attraction to kinks (solid squares). Lines are drawn according to the power laws predicted for the  $\rho = -2$ ,  $\rho = -1$ , and  $\rho = 1$  universality classes, respectively.

migration of adatoms during growth without desorption. To check the predictions of Table I, we investigate step bunching induced by strong ISE in KMC simulations. The resulting mean terrace width in the bunches,  $\ell_b$ , as a function of the deposition time is shown in Fig. 1 (open triangles). The exponent  $\delta = 1/5$  is in agreement with these data.

*The  $\rho = 1$  universality class.*—In many growth processes, several diffusing species exist on the surface [13]. Two of us [13] have shown that step bunching may occur if at least two kinds of diffusing particles are present on the surface, and these adparticles experience a NSE at step edges. Particles of one kind are deposited on the surface from the vapor or a beam (the “precursor”), while the other kind (the “growth unit”) is produced as the result of a chemical reaction involving the precursor. Step bunching is due to the coupling between the densities of precursors and growth units. This coupling gives rise to a nonequilibrium current that, in the region where step bunching shows up, behaves as  $J_{2p} = \Phi x_B^2 m_x$ , where  $\Phi$  is the deposition rate of precursors, and  $x_B$  is the precursor surface diffusion length. One sees that  $\rho = 1$  in this case. Another situation in which  $\rho = 1$  is expected, corresponds to growth (evaporation) with a weak inverse (normal) Schwoebel effect (WSE). Indeed, the current has in both growth and evaporation the form  $J_{\text{SE}} = -\Delta\Phi d_S(1 + d_S m_x)^{-1}$ , where  $d_S$  is a characteristic length scale given by the ratio  $D/\kappa$  between the adatom diffusion constant  $D$  and the (asymmetric) attachment coefficient  $\kappa$  to the step edge, and  $\Delta\Phi$  is the supersaturation [2]. Then, WSE implies  $d_S \ll \ell$ , so that  $J_{\text{WSE}} \sim -\Delta\Phi d_S(1 - d_S m_x)$ . We note that in the opposite limit  $d_S m_x \gg 1$  one recovers the  $J_{\text{ES}} \sim m_x^{-1}$

behavior leading to the  $\rho = -1$  universality class (see above). In yet another context, Stoyanov and Tonchev [11] derived from a modified Burton, Cabrera, and Frank (BCF) equation for electromigration-induced bunching a nonequilibrium current of the form  $J_{el} = -2Dc_{eq}Fm_x/(k_B T)$ , where  $F$  is the strength of the electromigration force. Bunching of steps is then predicted when the force  $F$  acts in the step-down direction. The last example of  $\rho = 1$  behavior is provided by a model in which SED is fast, and atoms are attracted to kinks from the two nearest neighbor sites on the lower side of the step. This asymmetry of the adatom incorporation at kinks creates a biased diffusion in the uphill direction of the form  $J_{neq} \sim m_x$ , that induces step bunching in KMC simulations of step-flow growth at high temperature (details will be given elsewhere). For this model, as well as for the two-particle model with NSE [18], we compute the average terrace width in the bunches,  $\ell_b$ , as a function of time (see Fig. 1, solid squares and open inverted triangles, respectively). The resulting behavior compares well with the prediction  $\delta = 1$  for the  $\rho = 1$  universality class. Note also that the predictions  $\gamma = 2/(n + 1)$  have been confirmed by the numerical solution of the BCF equations of step motion [11] for the electromigration problem with  $1/\ell^n$  step interactions.

*Experiments.*—Step bunching is very often observed in experiments, but rarely characterized quantitatively. We list in the following the studies we are aware of. Krishnamurthy *et al.* [6] have made a detailed study of step bunching evolution during the epitaxial growth of GaAs/AlAs multilayers on GaAs(110) substrates miscut towards [010]. They report measurements of the bunch density  $1/W \sim t^{-1/z}$  and the bunch height  $N \sim t^\beta$  as a function of deposition time. Their data agree with  $\beta = 1/2$ ,  $z = 10/3$ , as well as with  $\beta = 3/7$ ,  $z = 7/2$ , which would class this system either in the  $\rho = -1$  (ISE) or the  $\rho = -2$  (SED) universality class.

Schelling *et al.* [20] have measured the bunch height  $N$  as well as the period  $W$  of the bunch structure as a function of deposition time in Si(001) homoepitaxy on substrates miscut towards [110]. As for the previous case, inspection of their data (Fig. 3a of Ref. [20]) suggests that this system may fall either in the SED ( $\rho = -2$ ) or the ISE ( $\rho = -1$ ) universality class. The latter seems to be in contradiction with the observation of mounding at low temperatures in the same system, which is incompatible with an ISE, as well as with the investigation by Latyshev *et al.* [9] of step bunching during evaporation. Indeed, Latyshev *et al.* report that step bunching during direct current (dc) heating of vicinals of Si(001) is independent of the dc direction, which suggests that the instability is due to a NSE [2]. The exponents  $\beta$  and  $\gamma$  are found to be  $\beta = 0.53 \pm 0.05$  and  $\gamma = 0.46 \pm 0.05$ , respectively. Inspection of Table I shows that these exponents are not far from the values  $\beta = 0.5$  and  $\gamma = 0.4$  for  $\rho = -1$  (sug-

gesting the existence of a NSE, since they refer to evaporation). Of course, Si(001) is a complicated system with anisotropic diffusion and a detailed model study of its instabilities is obviously needed.

Step bunching induced by dc heating has been thoroughly investigated for vicinal surfaces of Si(111) [21]. The bunching appears here to depend in a complex way on the electric current direction, and on the temperature. When the instability is caused by an electric current in the step-down direction, experiments show that the facets between steps grow as  $W \sim t^{1/2}$ , so that  $z = 2$ . Furthermore, the experimental data fit the scaling relation  $\ell_b \sim N^{-\gamma}$  with  $\gamma = 0.69$ . Table I suggests that the system belongs to the  $\rho = 1$  universality class, at least when the electric current is step down.

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