New Nonlinear Evolution Equation for Steps during Molecular Beam Epitaxy on Vicinal Surfaces

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(Received 17 December 1997)

A nonlinear evolution equation for a vicinal surface growing in the step flow mode is derived in the limit of weak desorption. This limit turns out to be singular, and nonlinearities of arbitrary order need to be taken into account. The meandering instability caused by step edge barriers leads to a step morphology with a fixed meander wavelength, cusp singularities, and a step width increasing as $\sqrt{t}$ without bound. Full lattice gas simulations confirm this picture, and the observed step morphology is derived analytically from the evolution equation. [S0031-9007(98)06044-X]

PACS numbers: 68.35.Ct, 81.15.Hi

Many semiconductors are produced by molecular beam epitaxy (MBE). The ability to grow crystals with sharp interfaces on the atomic scale is of considerable importance in the manufacturing of devices. One possibility for achieving this goal is to grow a vicinal surface in the step-flow mode, where deposited adatoms attach directly to preexisting steps. Ideally, the surface grows without changing its shape, by the advancement of a uniform, stable step train [1].

In reality, a number of additional effects interfere with this scenario. Fluctuations in the molecular beam lead to the kinetic roughening of the surface [2], and impurities can pin the steps [3]. Here we focus on a deterministic instability related to energy barriers suppressing interlayer transport (the Ehrlich-Schwoebel effect [4–6]), which induces step meandering. This phenomenon was predicted on the basis of linear stability analysis [7,8], and has been observed experimentally in several systems [9,10]. An important parameter governing the behavior is the ratio between the step spacing, ℓ, and the distance ℓ_D between nucleation centers on a singular surface; step flow requires $\ell/\ell_D \ll 1$. Otherwise, a transition to three-dimensional growth can be triggered by nucleation on the terraces [11,12], and the description in terms of step motion breaks down. In this Letter we address the nonlinear evolution in the regime prior to this secondary instability, corresponding effectively to the limit $\ell/\ell_D \to 0$.

We present a systematic derivation of the step dynamics from the underlying microscopic principles (conservation and kinetic laws). Our starting point is the Burton-Cabrera-Frank (BCF) [13] model supplemented with noninstantaneous kinetic attachment at the steps. In preceding works [14,15] this type of treatment was performed in a situation where desorption is not negligible. The meandering instability then sets in at a nonzero threshold flux [7,8], and the dimensionless distance from the threshold provides a small parameter ϵ. An expansion in powers of ϵ was performed, and to leading nontrivial order the steps were found to obey an equation of the Kuramoto-Sivashinsky type. In the absence of desorption—which is the relevant case for MBE—meandering occurs at arbitrarily small fluxes. We discover here that in this limit the expansion in ϵ is singular, which is a nonstandard situation in nonlinear dynamics. A careful analysis allows us nevertheless to derive the appropriate evolution equation, which turns out to be highly nonlinear. The solutions of this equation show an in-phase step meander of fixed wavelength, in agreement with recent experiments [10].

Let c be the areal adatom density and F the deposition rate. Mass conservation implies

$$D\nabla^2 c + F = 0 \tag{1}$$

where the quasistatic approximation (valid for most practical purposes) is adopted, and D is the diffusion constant. At the steps mass currents are related to departure from equilibrium by

$$\pm D\,\frac{\partial c}{\partial n} = \nu_{\pm}(c - c_{\mathrm{eq}})\,, \tag{2}$$

where $+$ and $-$ refer to the lower and upper terraces respectively, ν is a phenomenological kinetic coefficient, and c_{eq} is an equilibrium concentration taking into account the step curvature effect: $c_{\mathrm{eq}} = c^0_{\mathrm{eq}}(1 + \kappa\Omega\gamma/k_BT)$, where c^0_{eq} is the equilibrium concentration for a straight step, κ is the step curvature, γ the line tension, Ω the atomic area, and k_BT the thermal energy. Finally, mass conservation at the step yields

$$v_n = D\Omega\left[\left(\frac{\partial c}{\partial n}\right)_+ - \left(\frac{\partial c}{\partial n}\right)_-\right], \tag{3}$$

where v_n is the step normal velocity, and $\partial/\partial n$ stands for the normal derivative. Equations (1)–(3) completely

specify the step dynamics. In general the resulting evolution equations are nonlocal and nonlinear [15]; however, at sufficiently large scales local dynamics prevail.

In order to introduce the small parameter ϵ whose value is a measure of locality, we write first the linear dispersion relation for small fluctuations around a regular train of straight steps [7,8]. For ease of presentation we consider an in-phase train, corresponding to the fastest growing linearly unstable mode [8], and one-sided attachment at the steps (only atoms coming from lower terraces are incorporated). We look for perturbations in the form of $e^{ikx+\omega t}$, where k is the wave number along the step, and ω the amplification rate. The linear dispersion relation takes the form

$$\omega = \frac{\epsilon D}{2} k^2 - D\Gamma\ell k^4 \equiv \alpha k^2 - \beta k^4, \qquad (4)$$

where $\epsilon = \Omega F \ell^2/D \ll 1$ is the Péclet number [1], ℓ is the interstep distance, and $\Gamma = \gamma\Omega^2 c_{\rm eq}^0/k_B T$. The steps are morphologically unstable against modes with wavelengths larger than $\lambda_c = 2\pi\sqrt{\beta/\alpha}$. The fastest growing mode has a wavelength $\lambda_u = 2\pi\sqrt{2\beta/\alpha} \sim \epsilon^{-1/2}$, and the time for development of instability scales as ϵ^{-2}.

In the spirit of a multiscale expansion [14] this suggests the introduction of slow spatial and temporal variables $X = \epsilon^{1/2}x$ and $\tau = \epsilon^2 t$, as in the case where desorption is present [14,15]. One would then naively be tempted to expand both the concentration c and the step position $\zeta(x,t)$ in power series in $\epsilon^{1/2}$. This standard analysis leads to the conclusion that there is no nonlinear term to counterbalance the linear terms. A close inspection [16] shows that the leading term in the step position must be singular, in the sense that $\zeta = H/\epsilon^{1/2}$, where H is of order 1. It might seem that this behavior is pathological and that therefore the expansion looses its meaning. However, the problem is resolved by noting that due to translational invariance only derivatives of ζ should enter the final equation. Indeed, since space scales as $\epsilon^{-1/2}$, then $\partial\zeta/\partial x = \partial H/\partial X \sim O(1)$, i.e., the slope is of order 1. As a consequence we expect nonlinearities of arbitrary order to enter the final equation. After the recognition of the singular behavior, and upon introduction of the "intrinsic" variable $\partial\zeta/\partial x$ as a natural representation of the step, the calculation follows the same route as in Ref. [14]. At the nontrivial $\epsilon^{3/2}$ order a solvability condition must be satisfied which results in the evolution equation we are seeking [16]:

$$\partial_t\zeta = -\partial_x\Bigg[\frac{\alpha\,\partial_x\zeta}{1+(\partial_x\zeta)^2} + \frac{\beta}{1+(\partial_x\zeta)^2} \times \partial_x\Bigg\{\frac{\partial_{xx}\zeta}{(1+(\partial_x\zeta)^2)^{3/2}}\Bigg\}\Bigg]. \qquad (5)$$

Because of our assumption of an in-phase train it takes the form of a conservation law, $\partial_t\zeta + \partial_x J = 0$, where the current J is the expression between braces.

It is instructive to compare Eq. (5) to the phenomenological equation for the height $h(x,z,t)$ of the surface above the substrate plane [6,12,17]. It is written as

$$\frac{\partial h}{\partial t} = -\nabla\Bigg[\frac{F\Omega\ell_D^2\nabla h}{1+(\ell_D/a)^2(\nabla h)^2} + K\nabla(\nabla^2 h)\Bigg], \qquad (6)$$

where $K > 0$ is a constant. Inserting the profile with an in-phase step train [12] $h(x,z,t) = (a/\ell)[\zeta(x,t) - z]$ into (6) and taking the limit $\ell/\ell_D \to 0$, one finds that the first term on the right hand side of (6) exactly coincides with the corresponding term in Eq. (5). In contrast, the second term on the right hand side of (5) contains nonlinearities not present in (6). While the nonlinearity inside the curly brackets is the standard form for the chemical potential of a stepped surface [18], the prefactor $\beta[1+(\partial_x\zeta)^2]^{-1}$ is new: It describes a reduction of the mobility due to the crowding of the deformed steps. We will see below that this term qualitatively changes the step dynamics.

Figure 1 shows a typical step profile starting from a random initial condition. (The final profile is found to be independent of the initial conditions; its basin of attraction is large enough.) The step develops a cuspy ordered structure whose amplitude grows unstably with time. The rms step width grows asymptotically as $\sqrt{t}$ (Fig. 2). This picture shares many features with that observed on Si(111) and Si(001) [9].

The method used to obtain Eq. (5) is based on an asymptotic expansion in ϵ. To check its range of validity, we have performed a numerical study of the full BCF model represented by Eqs. (1)–(3). We simulate a vicinal face with steps running on average in the x direction. Fluctuations of the step in the z direction are assumed to be described by a single-value function, i.e., overhangs and voids are excluded in the step configuration [19,20]. Atoms deposit on a square lattice of size 128×256, and diffuse on the vicinal surface containing four steps. We neglect the desorption process, corresponding to the MBE

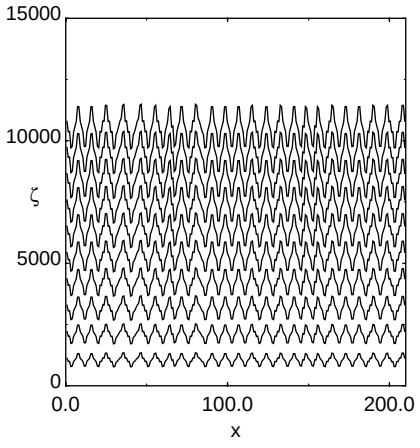

FIG. 1. A typical solution at different times.

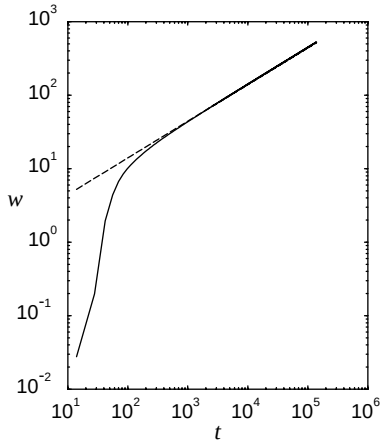

FIG. 2. The rms step width as a function of time.

situation. To realize the step-flow growth mode, nucleation on terraces is forbidden in the simulation. Periodic boundary conditions are assumed in the x direction, and helical ones in the z direction. The steps are initially straight. We assume (as in the analytical part) that the attachment occurs only from the lower terrace. The values of parameter sets are chosen to be the same as previously [19]: $D = 1$, $F = 10^{-4}$, $c_{\mathrm{eq}}^{0} = 0.119$, and the reciprocal of the step stiffness $k_B T/\gamma a = 0.362$.

The time evolution of one of the steps is shown in Fig. 3, and the associated variation of the rms step width w is shown in Fig. 4. In the early stage the step width seems to cross over from the random detachment behavior $w \sim t^{1/2}$, to the Edwards-Wilkinson behavior $w \sim t^{1/4}$ [21]. So far, each step has small fluctuations and behaves as an isolated one. At $t \approx 10^3$, the width becomes of order unity, and the step begins to sense the neighboring ones. The fluctuations increase abruptly in order to adjust the phase to the neighbors. After the phase adjustment $t > 10^5$, all the steps are synchronized in phase and the wavelength of fluctuation appears to remain constant [22]. The step profile is asymmetric in the z direction with flat tops and sharp valleys in contrast to Fig. 1. This suggests that symmetry-breaking terms [23] may appear in Eq. (5) at higher order. Still the step width increases as $w \propto t^{1/2}$ as shown in Fig. 4. It must be emphasized that this behavior is different from what is observed in some $(1 + 1)$-dimensional growth models with strong step-edge barriers [23,24] where it is essentially due to shot noise. In contrast, here the behavior reflects the interaction of steps through the diffusion field.

These simulations suggest that the evolution equation (5) captures the essential features. Next, we explain the observed behavior through the study of Eq. (5). In a steady state, $J = 0$, it leads to Newton's equation for a fictitious particle with a coordinate $m(x) = \partial_x \zeta/\sqrt{1 + (\partial_x \zeta)^2}$ moving in time x as $\beta d^2 m/dx^2 = -dU/dm$ with the potential $U(m) \equiv -\alpha\sqrt{1 - m^2}$. Using energy conservation, the amplitude A and the wavelength λ of the step profile can be evaluated as functions of the turning point m_0 of the particle (corresponding to the largest slope in the step profile), giving the results:

$$A = \sqrt{8\beta(\alpha + \mathcal{E})}/\alpha\,, \tag{7}$$

$$\lambda = 4\sqrt{\beta/\alpha}\,[2\mathbf{E}(\sin(\theta_0/2)) - \mathbf{K}(\sin(\theta_0/2))]\,, \tag{8}$$

where $\mathbf{E}$ and $\mathbf{K}$ are complete elliptic integrals of the first and second kind, respectively, $\mathcal{E} = U(m_0)$ is the particle energy, and θ_0 is defined through the relation $m_0 = \sin\theta_0$. The amplitude is an increasing function of $\mathcal{E}$ ($-\alpha \le \mathcal{E} \le 0$) and remains finite even for a maximal slope $\partial_x \zeta$ going to infinity (i.e., for $m_0 = 1$, $\mathcal{E} = 0$): $A \to \sqrt{8\beta/\alpha}$. Conversely, λ is a *decreasing* function of the maximal slope, varying from λ_c for $\mathcal{E} = -\alpha$ to $\sqrt{8\beta/\alpha}\int_0^{\pi/2} d\theta\,\sqrt{\cos\theta}$ for $\mathcal{E} = 0$. In other words, stationary profiles exist only

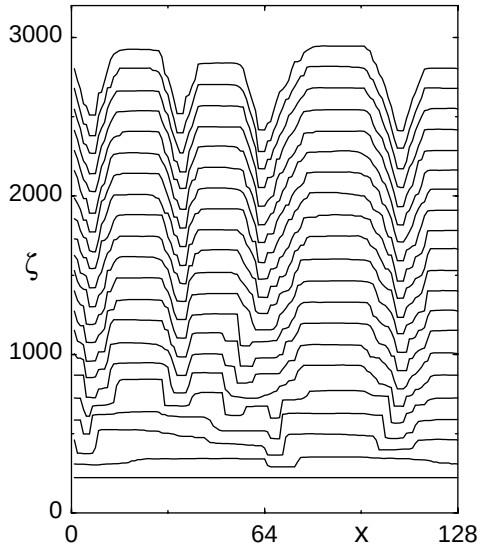

FIG. 3. Time evolution of one step in a step train.

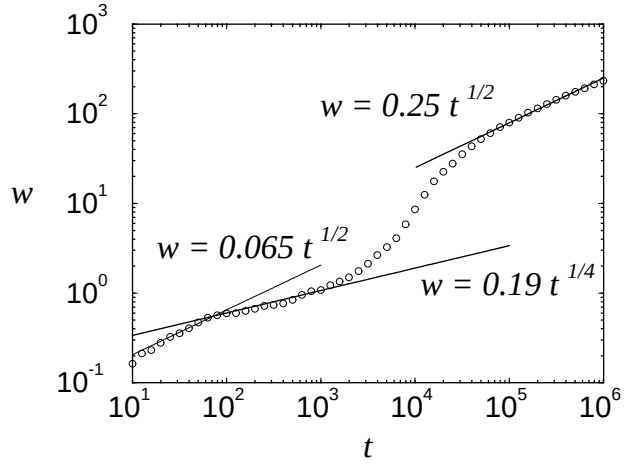

FIG. 4. Time dependence of the step width in double logarithmic scale. Full lines are guides to the eye.

for values of λ where the straight step is *stable* against small fluctuations. In marked contrast to the standard phenomenological equations [6,12,17], there are no stationary configurations in the linearly unstable regime $\lambda > \lambda_c$, which could support a coarsening process towards larger wavelengths. Instead, the most unstable wavelength λ_u is frozen in during the further evolution of the step. Indeed, using normalized variables ($x, \zeta, t \to \sqrt{\alpha/\beta}\, x, \zeta$ and $\alpha^2 t/\beta$) $\lambda_u = 2^{3/2}\pi = 8.89$, which is very close to the value (8.7–8.8) displayed in Fig. 1.

Solutions of fixed λ are described by the ansatz $\zeta(x,t) = A(t)g(x)$, where $A(t)$ is an increasing function of time. Inserting this into (5) it is seen that the second, equilibrium contribution to the current vanishes as A^{-3} relative to the first, nonequilibrium term. For $t \to \infty$, $A \to \infty$ and the equation reduces to $\dot{A}A = \alpha g''/[g(g')^2] = C$ where $C > 0$ is a constant and $\dot{A} = dA/dt$, $g' = dg/dx$. It follows immediately that the amplitude grows as $\sqrt{t}$. To solve the equation for g appropriate boundary conditions have to be specified. By symmetry the current J has to vanish at the maxima and minima of the profile. Since $J \sim 1/g'$ this implies that g' diverges at $x = 0$ and $x = \lambda/2$. The relevant solution reads [25]

$$g(x) = \lambda/(2\sqrt{\pi})\,\mathrm{erf}^{-1}(4x/\lambda - 1)\,, \qquad (9)$$

where $\mathrm{erf}(x)$ is the error function. This is consistent with the numerical profiles shown in Fig. 1, and the prediction for the step width $\sqrt{\langle \zeta^2 \rangle} = (2\alpha t)^{1/2}$ is in good agreement with the data in Fig. 2.

In summary, we have shown that, as in the case with desorption [14], we can extract from the full BCF model the step evolution equation during MBE. The absence of desorption is subtle and completely changes the mathematical structure of the problem. This difficulty could, nevertheless, be circumvented. The results show that there is no coarsening, but rather the step develops a spikelike ordered pattern. The overall shape together with the temporal behavior of the width of the step obtained from the continuum evolution equation and MC simulations agree well. A striking feature, a finite time singularity within the continuum approach, appeared. A physical cutoff must intervene. Does the next order contribution in ϵ capture that effect, or does the singularity survive at higher orders? This question must be elucidated in the future. Another subject of further investigation is the transient behavior of the out-of-phase train before the steps become synchronized. Of interest might also be to elucidate the effect of the ES effect at the step corners [24]. Finally when allowance is made for nucleation (low temperature or small misorientation angle) this should give rise to a rich interplay between unstable step-flow and 2D island formation. It is of importance to specify the time scale beyond which nucleation competes with step flow.

O. P-L is supported by NSF Grant No. DMR 96-32521. C. M. is grateful to J. Villain for fruitful discussions. P. P. gratefully acknowledges financial support from Alexander von Humboldt Stiftung. The work of J. K. was supported by DFG within SFB 237 *Unordnung und grosse Fluktuationen*.

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[1] J. Y. Tsao, *Material Fundamentals of Molecular Beam Epitaxy* (Academic Press, Boston, 1993).
[2] D. E. Wolf, Phys. Rev. Lett. **67**, 1783 (1991).
[3] D. Kandel and J. D. Weeks, Phys. Rev. Lett. **72**, 1678 (1994).
[4] G. Ehrlich and F. G. Hudda, J. Chem. Phys. **44**, 1039 (1966); R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. **37**, 3682 (1966).
[5] J. Villain, J. Phys. I **1**, 19 (1991).
[6] J. Krug, Adv. Phys. **46**, 139 (1997).
[7] G. S. Bales and A. Zangwill, Phys. Rev. B **41**, 5500 (1990).
[8] A. Pimpinelli, I. Elkinani, A. Karma, C. Misbah, and J. Villain, J. Phys. Condens. Matter **6**, 2661 (1994).
[9] R. T. Tung and F. Schrey, Phys. Rev. Lett. **63**, 1277 (1989); F. Wu, S. G. Jaloviar, D. E. Savage, and M. G. Lagally, Phys. Rev. Lett. **71**, 4190 (1993).
[10] L. Schwenger, R. L. Folkerts, and H.-J. Ernst, Phys. Rev. B **55**, R7406 (1997).
[11] J. Krug and M. Schimschak, J. Phys. I (France) **5**, 1065 (1995).
[12] M. Rost, P. Scămilauer, and J. Krug, Surf. Sci. **369**, 393 (1996).
[13] W. K. Burton, N. Cabrebra, and F. C. Frank, Philos. Trans. R. Soc. London A **243**, 299 (1951).
[14] I. Bena, C. Misbah, and A. Valance, Phys. Rev. B **47**, 7408 (1993).
[15] O. Pierre-Louis and C. Misbah, Phys. Rev. Lett. **76**, 4761 (1996); O. Pierre-Louis and C. Misbah (to be published).
[16] O. Pierre-Louis, Thèse d'Université, Université J. Fourier, Grenoble, France, 1997.
[17] A. W. Hunt, C. Orme, D. R. M. Williams, B. G. Orr, L. M. Sander, Europhys. Lett. **27**, 611 (1994); M. Rost and J. Krug, Phys. Rev. E **55**, 3952 (1997).
[18] P. Nozières, in *Solids Far From Equilibrium*, edited by C. Godrèche (Cambridge University Press, Cambridge, 1991), p. 1.
[19] M. Uwaha and Y. Saito, Phys. Rev. Lett. **68**, 224 (1992).
[20] Y. Saito and M. Uwaha, Phys. Rev. B **49**, 10 677 (1994).
[21] T. Salditt and H. Spohn, Phys. Rev. E **47**, 3524 (1993).
[22] A quantitative comparison with the continuum version requires an analysis of the out-of phase analog of Eq. (5).
[23] P. Politi, J. Phys. I (France) **7**, 797 (1997).
[24] Z. Zhang, J. Detch, and H. Metiu, Phys. Rev. B **48**, 4972 (1993); P. Politi and J. Villain, Phys. Rev. B **54**, 5114 (1996).
[25] J. Krug, J. Stat. Phys. **87**, 505 (1997).