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# Vicinal surfaces: growth structures close to the instability threshold and far beyond

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**Abstract.** We introduce a new numerical approach to step flow growth, making use of its analogies to dendritic growth. Concentrating on the situation close to the instability threshold of step growth, nonlinear evolutionary equations for the steps on a vicinal surface can be derived in a multiple-scale analysis. This approach retains the relevant nonlinearities sufficiently close to the threshold. Our simulations recover and visualize these findings. However, on the basis of our simulations we further report results on the behaviour far from the threshold. Step propagation is treated as a moving-boundary problem based on the Burton–Cabrera–Frank (Burton W K, Cabrera N and Frank F C 1951 *Phil. Trans. R. Soc.* A **243** 299) model. Our method handles the problem in a fully dynamical manner without any quasistatic approximations. Furthermore, it allows for overhangs.

## 1. Introduction

The ability to control atomistically the growth of metal and semiconductor crystal surfaces is one of the central topics of research both from a technological [1, 2] and from a fundamental theoretical [3] point of view. One of the focuses at the forefront of research is on the creation of new epitaxial architectures by careful manipulation of deposition processes [4]. Among the existing growth techniques, molecular beam epitaxy (MBE) has been established as a particularly important method for efficiently growing substrates in a well defined layer-by-layer mode. For example, atomically abrupt and smooth interfaces can be obtained on vicinal surfaces by keeping the MBE conditions appropriate for *step flow* growth. Step flow has long been recognized as an alternative to two-dimensional nucleation [5]. A necessary condition for 'good' materials is that the flowing step train is stable. One of the undesirable effects which eventually interferes with stable growth is faceting [6]. Destabilization mechanisms which may disturb step flow growth can be of either extrinsic or intrinsic origin. An example of the former case is provided by impurities acting as pinning centres [7]. The latter case has recently been addressed by Bales and Zangwill [8] in a linear stability analysis, showing a morphological instability driven by diffusion of atoms along the terrace. The results are the critical condition of the onset of instability and a band of active modes growing exponentially in time. Bena *et al* [9] showed that, close to the threshold, nonlinear terms can be found to saturate the linear amplification, leading to Kuramoto–Sivashinsky-like [10, 11] behaviour of the system. Higher-order nonlinear terms can change this behaviour into another regime of morphological instability, which will be illustrated by pictures derived from simulations in the following. Our simulations also reveal that crystal anisotropy can strongly influence the morphology selection.

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## 2. The numerical method

A version of the step flow model together with an explanation of the choice of its equations has already been given in [8]. At this point we merely rewrite the equations in a dimensionless form:

$$\partial_t u = \vec{\nabla} u^2 - u \tag{1}$$

$$v_n = (\partial_n u)_+ - (\partial_n u)_- \tag{2}$$

$$\pm(\partial_n u)_\pm = \overline{\nu}_\pm (u|_I - u_{eq}) \tag{3}$$

$$u_{eq} = -\Delta + d_0 \overline{\kappa} \tag{4}$$

($\partial_n$ denotes the spatial derivative in the direction normal to the interface, $v_n$ is the normal velocity at the step, $\partial_t$ the partial derivative with respect to time). The equation field $u$ is related to the original adatom concentration $c$ of the Burton–Cabrera–Frank (BCF) approach by

$$u = \Omega(c - \tau F) \tag{5}$$

where $\Omega$ is the atomic area of the solid and $F$ the frequency of the rate of impingement of adatoms from the gas atmosphere. Such adatoms diffuse and evaporate with lifetime $\tau$, so their global dynamics away from the step edge is governed by the diffusion–relaxation equation (1). In case the where it reaches the step edge before evaporation, equations (2) to (4) provide the boundary conditions, which govern its incorporation in the step edge. This incorporation process leads to the step advancing with velocity $v_n$, thereby setting the new boundary for equation (1). The parameters used are given by

$$\Delta = -\Omega(c_{eq} - \tau F) \tag{6}$$

with $c_{eq}$ equal to the equilibrium concentration at the step and $d_0 = \Omega c_{eq} \Gamma$ ($\Gamma \sim$ line tension). Finally, $\nu_\pm$ are attachment coefficients for the incorporation of adatoms at the upper and lower step edges, respectively. Figure 1 gives a sketch of the physics underlying this mathematical formulation.

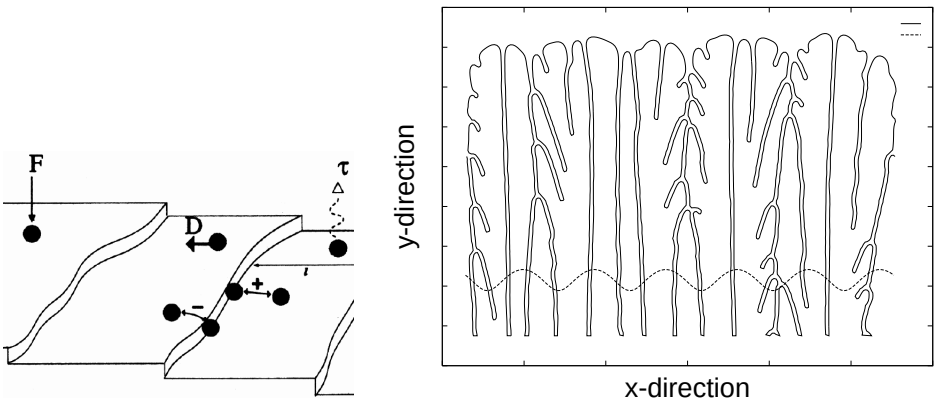


**Figure 1.** Left: the basic physics of step flow growth. Adatoms are impinging on a terrace with frequency $F$ and desorbing after a lifetime $\tau$. On reaching the step edges, their incorporation is governed by different attachment coefficients $\nu_{pm}$ for attachment from above or below, respectively. Right: contours of a single step advancing in the $y$-direction. Note that the frame of reference of the simulation moves with the structures' tip velocity. The dotted line gives the initial conditions at $t = 0$ and is only superposed.

Equations (1) to (4) formulate a moving-boundary problem, since $v_n$ is the velocity of the interface acting on the diffusion field through changing its boundary every time step. Such problems have been dealt with in various contexts—amongst others, for the problem of free diffusive growth from an undercooled melt [12]. We adopt the numerical method used in reference [12] with its special features of eliminating additional numerical anisotropy by using sandwiched grids and solving the full dynamical problem. The latter is necessary if Hopf bifurcations, which could in principle arise in such a system [20], are detected.

## 3. Morphologies beyond the threshold

The threshold of instability for step flow growth can be found in a linear stability analysis [8]. Concentrating on the situation close to the threshold, nonlinear terms which are relevant for the evolution of the system can be derived in a multiple-scale analysis. Up to third order, such an analysis was carried out in [9] leading to the following canonical-form equation (Kuramoto–Sivashinsky (KS)):

$$h_t = -h_{xx} - h_{xxxx} + \frac{1}{2}h_x^2 \tag{7}$$

(here partial derivatives are denoted by subscripts). The first term leads to an amplification of the long-wavelength modes, while the second stabilizes short-wavelength modes. The nonlinear term obviously serves to saturate the unstable modes, resulting for example in cellular structures or viscoelastic behaviour [15].

A little further away from the threshold, the systems' behaviour can already be characterized by the evolution of deep holes in an initially weakly modulated interface (figure 1, right). This behaviour can be related to a term $h_{xx}^2$ found in a multiple-scale analysis up to fourth order. This analysis can be carried out as in [13], and in the same manner it is possible to record a kinetic phase diagram for the '$d_0$–$\Delta$' plane (figure 2).

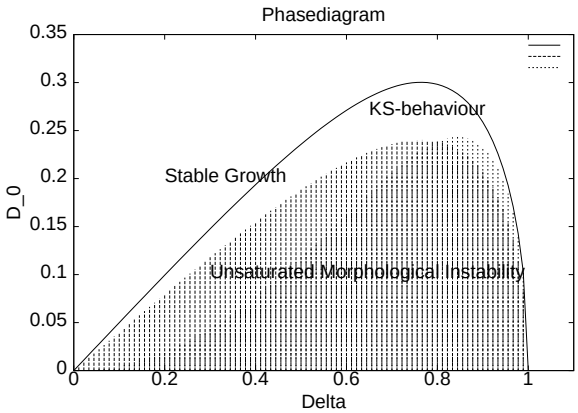


**Figure 2.** The phase diagram in the '$d_0$–$\Delta$' plane. The precise contour of the KS regime can only be estimated from the analytical limits $\epsilon \ll 1$ and $\epsilon/(1-\Delta) \ll 1$ together with simulation experience.

An illustration of our simulations of the viscoelastic KS-type behaviour is found in figure 3. We show a train of three steps advancing in the $y$-direction. The lines give the contours of the step edges seen from above. The grey-levels represent the adatom density, which is lowest actually at the step edges. We do *not* show the monatomic height difference between the steps (compare figure 1 (left)).

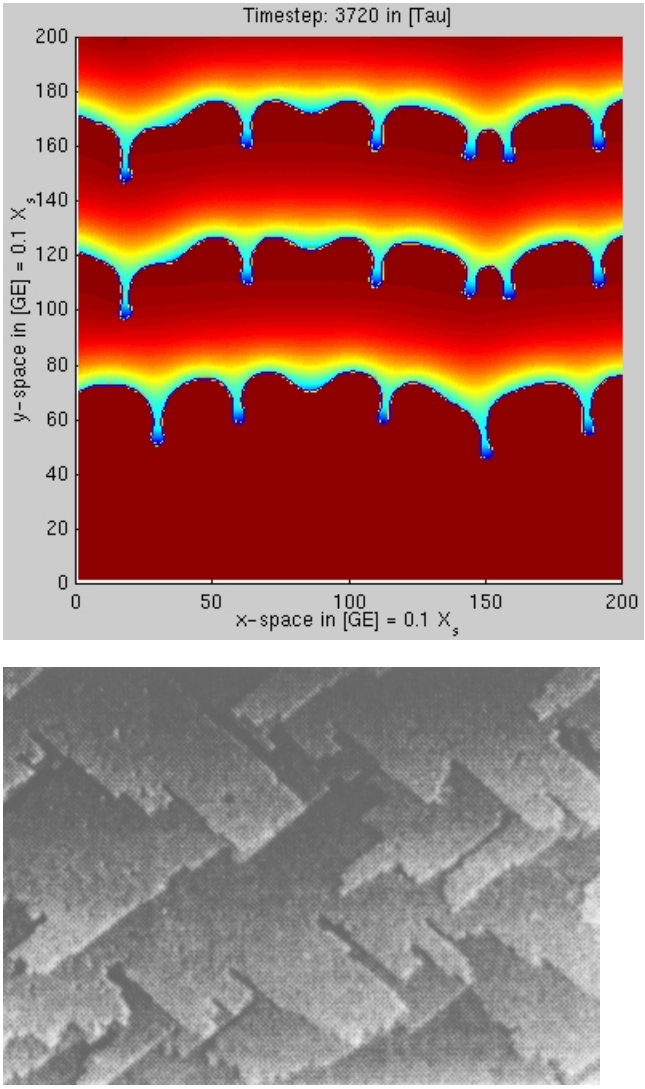


**Figure 3.** Top: a train of three steps simulated with our numerical approach slightly below the threshold, yielding KS-type behaviour. Bottom: a STM of a vicinal Si(001) surface with a miscut in the ⟨100⟩ direction after 500 seconds of deposition at a rate of 0.02 monolayers per second [16]. (This figure can be viewed in colour in the electronic version of the article; see `www.iop.org`)

In the following, we would like to comment on the meaning of this KS regime for experimental step flow growth at vicinal surfaces. The viscoelastic behaviour of the KS equation is known to be a manifestation of the coupling of an Eckhaus instability with long-wavelength Galilean distortions [19]. However, an Eckhaus instability can only be excited if the wavenumber of the initial perturbation of the interface falls into the interval

$$q_I \in \{q_e; q_c\} \tag{8}$$

where $q_e$ is the wavenumber of the onset of the Eckhaus instability and $q_c$ is the wavenumber corresponding to $\lambda_c$, the shortest wavelength to yield linearly unstable growth. A linear stability

analysis taking into account the propagation of the step yields

$$q_c^2 = 4\left(\frac{2}{\Delta} - 1\right)^2 (3 - \Delta)^{-1}\epsilon \tag{9}$$

$$q_e^2 = \frac{1}{3}q_c^2 \tag{10}$$

where $\epsilon$ measures the distance from the threshold. On the other hand, an experimental vicinal surface is finite; thus the only possible wavenumbers of an initial perturbation which are solutions to the KS equation are a discrete rather than a continuous set:

$$q_n = \frac{2\pi}{L}n \qquad n = 1, 2, \ldots \tag{11}$$

where $L$ is the width of the vicinal surface perpendicular to the direction of step propagation. Since the KS regime is limited to a very restricted set of $\epsilon$-values (figure 2), even in extended experimental systems it might not be possible to prepare an initial state with a wavenumber obeying equations (8) to (11) for certain $d_0/\Delta$ ratios.

There is just as good physical evidence that long-range Galilean distortions are unlikely to occur in an experimental system. The reason is that the approach described above completely neglects the influence of crystal anisotropy. From dendritic growth analysis, however, anisotropy is known to have a great impact on the selection of growth morphologies. This point was already mentioned in [18], where the authors found that crystal anisotropy stabilizes the step advancing in the direction of minimal step stiffness. Now, due to dimerization effects, the direction of minimal step stiffness does not need to be aligned with the direction of global step propagation (figure 3, bottom). We can take account of that in our mathematical formulation by changing equation (4) to

$$u_{eq} = -\Delta + d_0\overline{\kappa}[1 - \epsilon_m \cos(m(\theta - \theta_0))] \tag{12}$$

where $\theta_0$ is the angle between the directions of minimal step stiffness and global propagation ($\epsilon_m$ denotes the crystal anisotropy). Simulation of equations (1), (2), (3) and (12) with $\epsilon_4 = 0.1$ and $\theta_0 = 25^\circ$ gives rise to travelling waves, as shown in figure 4. In this physical context, no Galilean distortions are to be expected. However, in long-time evolution it is possible to detect wavelength modulations (figure 4, bottom) as a result of a purely diffusive instability due to the translational Goldstone mode which is a neutral mode. We are currently in the process of building up an analytical description of this phenomenon via a coupled set of amplitude and phase equations, and will comment on its bifurcation scenario elsewhere [21].

## 4. Summary

Analysis carried out to extend the Bales–Zangwill stability analysis to the weakly nonlinear regime [9] is valuable for showing that linearly unstable step flow growth can be saturated, giving rise to further physical scenarios. However, its relevance to experimental systems remains unclear, and the experimental proof of a step meandering in chaotic fluctuations rather than statistical ones remains to be given. In the meantime, it is worthwhile to study the influence of further experimental variables via our numerical approach, to increase our understanding of the mechanisms of step flow growth as revealed in experiments. We believe crystal anisotropy to constitute quite an important factor in this context. First simulations have revealed that, in particular, especially unsaturated instabilities, such as those shown in figure 1 (right), can be changed drastically by crystal anisotropy as well as by the anisotropy of the attachment kinetics. We will report our results which model the formation of vicinal facets, as found experimentally in [17], in a further article [22].

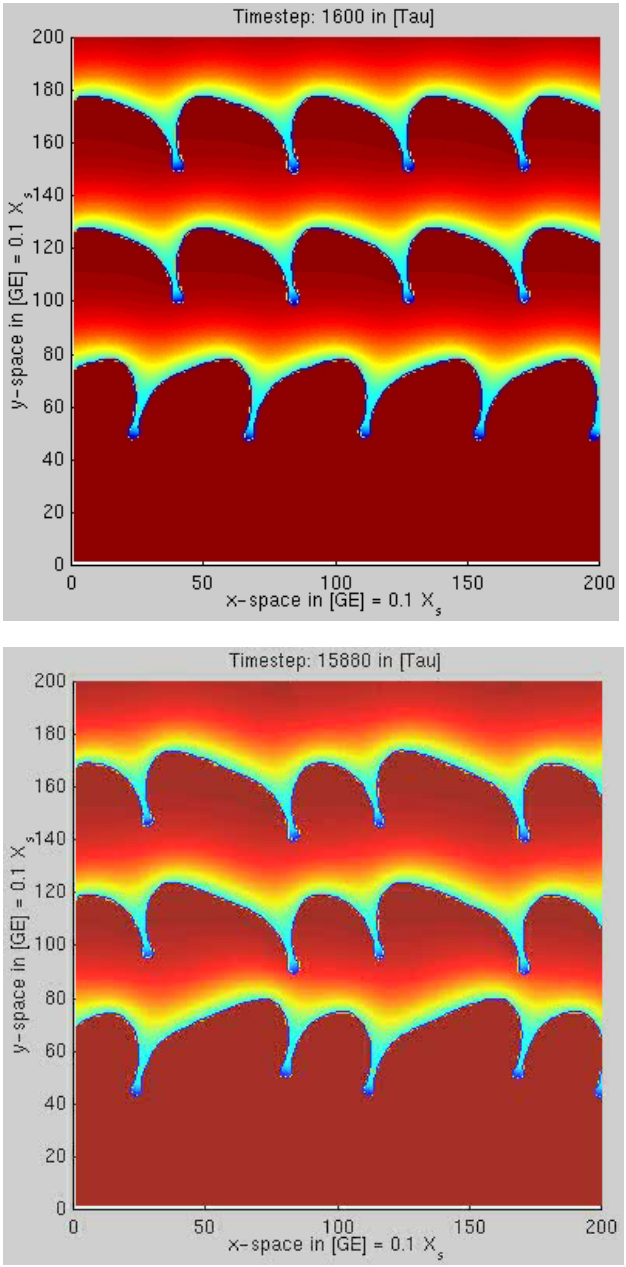


**Figure 4.** Top: a simulation using the same parameters as in figure 3 (top); however, equation (12) with $\epsilon_4 = 0.1$ and $\theta_0 = 25^\circ$ is used instead of equation (4). Bottom: the same simulation as at the top, but roughly $1.4 \times 10^4$ time steps later.

(This figure can be viewed in colour in the electronic version of the article; see `www.iop.org`)

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## References


[1] Ben-Naim E, Krapivsky P L and Redner S 1994 *Phys. Rev.* E **50** 822
[2] Eaglesham D J 1995 *J. Appl. Phys.* **77** 3597
[3] Barabasi A L and Stanley H E 1995 *Fractal Concepts in Surface Growth* (Cambridge: Cambridge University Press)
[4] Bauer E G 1999 *J. Mater. Res.* to be published
[5] Rosenberger F 1982 *Interfacial Aspects of Phase Transformations* ed B Mutaftschiev (Dordrecht: Reidel)
[6] Hirayama H and Asahi H 1994 MBE with gaseous source *Handbook of Crystal Growth 3* ed D T J Hurdle
[7] Frank C 1958 *Growth and Perfection of Crystals* ed R Doremus *et al* (New York: Wiley)
[8] Bales G S and Zangwill A 1990 *Phys. Rev.* B **41** 5500
[9] Bena I, Misbah C and Valance A 1993 *Phys. Rev.* B **47** 7408
[10] Sivashinsky G I 1983 *Physica* D **8** 243
[11] Kuramoto Y and Tsuzuki T 1976 *Prog. Theor. Phys.* **55** 356
[12] Ihle T and Mueller-Krumbhaar H 1995 *Phys. Rev.* E **51** 4751
[13] Ihle T and Mueller-Krumbhaar H 1996 *J. Physique* I **6** 949
[14] Akamatsu S, Faivre G and Ihle T 1994 *Phys. Rev.* E **49** 2972
[15] Manneville P 1988 *Propagations in Systems far from Equilibrium* ed J E Wesfreid *et al* (Berlin: Springer)
[16] Wu F, Jaloviar S G, Savage D E and Lagally M G 1993 *Phys. Rev. Lett.* **71** 4190
[17] Schwenger L, Folkerts R L and Ernst H J 1997 *Phys. Rev.* B **55** R7406
[18] Saito Y and Uwaha M 1994 *Phys. Rev.* B **49** 10 677
[19] Cross M C and Hohenberg P C 1993 *Rev. Mod. Phys.* **65** 851
[20] Emmerich H, Schleussner D, Kassner K and Ihle T 1999 submitted
[21] Igarashi A and Emmerich H 1999 *Minisymposium, MPIPKS, Dresden*
[22] Emmerich H, unpublished