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Nonlinear evolution of a terrace edge during step-flow growth

Ioana Bena, Chaouqi Misbah, and Alexandre Valance*
Institut Laue-Langevin, Boîte Postale 156X, 38042 Grenoble CEDEX, France
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The nonlinear evolution of a terrace edge during growth in the step-flow configuration is investigated. Starting from the constitutive growth equations of Burton, Cabrera, and Frank [Philos. Trans. R. Soc. London Ser. A **243**, 299 (1951)], we show that the dynamics of an isolated terrace edge, i.e., in the absence of any kind of step-step interaction, is described, close enough to the morphological instability threshold found by Bales and Zangwill [Phys. Rev. B **41**, 5500 (1990)], by a nonlinear partial differential equation. This equation, although it emerges physically in a somewhat disguised manner, can be simply interpreted as representing the growth with an effective negative line tension, a stabilizing line diffusionlike process along the edge, and a quadratic nonlinearity restoring the Galilean invariance. This nonlinear equation, despite its apparent simplicity, manifests a variety of dynamics, ranging from simple steady-cellular structures to spatiotemporal chaos. When the system size is large enough, chaotic dynamics always prevails. We argue here that when the step-step interaction is important, however, one expects "regular" solutions, such as steady-cellular, broken-parity-traveling, and oscillatory modes, to be stable over a finite range of control parameters.

I. INTRODUCTION

The feasibility of microscopic resolution imaging of solid surfaces, such as those provided by tunneling microscope and electron microscopy, has stimulated increasing interest in structural and dynamical features of surfaces.[1-14] In particular, the study of equilibrium step fluctuations of vicinal surfaces is a branch of investigation where these techniques can bring to bear their specific advantages. Another important ambitious issue where microscopic techniques should constitute crucial tools pertains to nonequilibrium features during the growth of solids from their vapor phase. Growth by molecular-beam epitaxy, for example, is a topic of much current technological and fundamental interest. On the one hand, the ability to grow single-crystal materials with essentially monolayer compositional control is of paramount importance in fabricating atomically abrupt interfaces for specific applications. On the other hand, this problem provides us with a fascinating and rich example where one is often faced with both statistical properties and out-of-equilibrium features, inherent to the growth process.

The most obvious feature of the steps is their meandering (or waviness). The meandering has often been thought of as being associated with equilibrium properties, such as, for example, thermal fluctuations. There is experimental evidence, however, where manifestly nonequilibrium features are observed.[15-17] There the steps seem to exhibit roughness of a decidely nonrandom nature. Recently, Bales and Zanbwill[18] showed from a linear stability analysis that a terrace edge growing from the vapor phase—in the so-called step-flow model—undergoes a morphological instability if the growth rate exceeds a certain critical value. This instability is intrinsic; it is driven by diffusion of atoms along the terrace. The stabilizing effect is due to the line tension of the step, which prevents short-wavelength deformations. The instability can thus take place only if the characteristic diffusion length (to be defined below) becomes smaller than the typical "chemical" atomistic length determined by the ratio of the line tension to the thermal excitation energy.

The linear stability analysis is the first step towards the understanding of the mechanisms by which a new pattern can be formed. The main benefits of a linear stability analysis are the determination of the critical condition of the onset of the instability, and the range of wave numbers of those perturbations that are likely to grow first. However, if the long-time behavior of a growing instability is to be ascertained and/or the final stage (if any) of the dynamics to be determined, then a nonlinear analysis is necessary. At arbitrary distance above the instability, threshold dynamics might involve highly nonlinear effects that make the theoretical analysis rather difficult. The analysis, however, is often facilitated by concentrating on the situation close to the instability point. We can thus hope to extract from a set of rather complicated dynamical equations only that part that is relevant to the dynamics close to the critical region. Our main objective in this paper is to develop a nonlinear theory for the terrace edge evolution during step flow. An important point in our analysis is that, close enough to the instability threshold, the most dangerous fluctuations found from the linear stability analysis of Bales and Zangwill[18] correspond to the long-wavelength (on the scale of the diffusion length) domain. We have taken advantage of this fact and shown that the growth dynamics are described by a "simple" nonlinear partial differential equation for the terrace edge. In our treatment, although we have in mind a uniform vicinal surface topology, we will confine ourselves in a first level to a situation where a single step is advancing, thus disregarding all kinds of step interactions. In this limit the equation describing the step dynamics can be set in the following (canonical) form:

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

where h represents a dimensionless position of the terrace edge, which is a function of dimensionless space x and time t variables. This equation is free of any parameter. Equation (1) intervenes in many contexts in physical and chemical phenomena;[21] it is known as the Kuramoto-Sivashinsky (KS) (Refs. 22 and 23) equation. The striking feature of this equation—a feature which still attracts extensive work—is that, despite its apparent simplicity (the only nonlinearity is quadratic), it manifests exotic dynamics ranging from ordered cellularlike structures to spatiotemporal chaos. When the system size (e.g., the lateral step extent) is large enough, spatiotemporal chaos always prevails. In that situation the terrace edge would exhibit an erratic dynamics of a completely *deterministic* origin.

Of course a single step advancing without any interaction with its neighbors is far from being realistic. We therefore intend to discuss how the step dynamics would be modified in the presence of interactions. We restrict ourselves here to a phenomenological picture where the interaction is modeled by adding a stabilizing linear term to Eq. (1), which mimics a repulsive interaction. The presence of this term is very important: the terrace edge should develop a periodic cellular steady structure which may be stable even for very extended lateral dimensions. When the importance of this term is reduced, the cellular structure becomes unstable. We will refer to these instabilities as *secondary* instabilities (we use the primary instability denomination by taking the straight step as reference). The loss of stability leads to the creation of new patterns: broken-parity-traveling cells, vascillating-breathing modes where the width of each cell oscillates in phase in opposition with its neighbors, or a complex mixture of these modes. When the step interaction is negligibly small, we recover KS dynamics.

We would like to mention here that our theory is, by its very nature, expected to be valid close enough to the instability threshold. It turns out, however, from preliminary lattice-gas simulations,[19] which handle in principle situations far from the threshold, that our treatment captures all the essential features.

The strategy of this paper is as follows. In Sec. II we write down the basic version of the step-flow model of Burton Cabrera, and Frank,[20] and comment on them briefly. We begin with an isolated step. For completeness, we reconsider in Sec. III the linear stability analysis of Bales and Zangwill.[18] Section IV is devoted to the derivation of the nonlinear evolution equation for the terrace edge. We shall then give a brief report on the basic properties of this equation in Sec. V. In Sec. VI we attempt to include phenomenologically the step-step interaction and present the overall picture of the terrace edge dynamics. In Sec. VII we sum up our results.

II. BASIC EQUATIONS

Since the step-flow model has been described elsewhere,[18] we shall keep its discussion brief. We consider the following situation: a single atomic step on a close-packed face of a crystal is advancing in the z direction at the expense of the gas atmosphere. An adatom impinges from a gas atmosphere with a frequency F, migrates on the facet with a diffusion constant D, and evaporates with a lifetime τ. It is widely admitted[18] that most of the adatoms that contribute to the growth process come from the terrace that is ahead (the lower one in Fig. 1) of the advancing step. Without loss of generality we will consider the following extreme limit: we assume that the growth is governed only by adatoms on that terrace. This corresponds to the so-called *one-sided* model. The density c of adatoms obeys the mass conservation law, which takes, for all practical purposes,[24] the following form:

$$D\nabla^2 c - c/\tau + F = 0 \ . \tag{2}$$

The amount by which each point on the step advances in the normal direction (see Fig. 1) is determined by the concentration gradient in the same direction. More precisely the normal velocity v_n is proportional to the normal derivative of c

$$v_n \Delta c_s = D\mathbf{n}\cdot\nabla c \ , \tag{3}$$

where Δc_s is the difference between the areal density of atoms in the solid phase and the corresponding quantity on the terrace immediately adjacent to the step. Since the density of atoms in the solid phase is much higher than in the gas atmosphere, we can write $\Delta c_s \simeq 1/\Omega$, where Ω is the atomic area of the solid. $\mathbf{n}$ is a unit normal out of the solid perpendicular to the step riser (see Fig. 1). Since the basic state will be a straight step advancing at a constant velocity V_0 in the z direction, we find it convenient to measure the z coordinate in the frame of reference moving at velocity V_0, undetermined for the moment. Let $z = \zeta(x,t)$ denote the instantaneous position of the step. The normal velocity takes the form

$$v_n = [V_0 + \dot{\zeta}(x,t)]/[1+\zeta_x^2]^{1/2} \ . \tag{4}$$

According to von Neuman theorem, Eq. (2) with the normal derivative of c given by (3) has solutions in a steady-state situation, for arbitrary boundary shapes $\zeta(x)$. However, not all the solutions will be physical. Indeed there is another physical condition to be satisfied at the boundary. While Eq. (3) guarantees mass conservation across the step, we do not yet know how the mass current at the step is related to the chemical potential difference between the growing solid and gas, $\Delta\mu$. In other words we should specify how *dissipation* at the step proceeds. This question is handled by making use of the second principle of thermodynamics. In a picture like that of Onsager, the

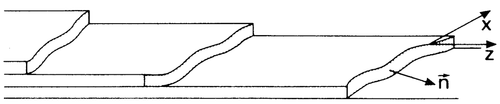

FIG. 1. Schematic view of a vicinal surface during step flow. **n** designates the normal to the step riser.

normal velocity (or equivalently the mass current) is proportional to $\Delta\mu$. We will assume there that the step is rough, in that it acts as a continuous capture center for adatoms. More specifically, we consider the extreme situation where kinetics are fast enough on all time scales of interest, so that the step can be viewed at instantaneous chemical equilibrium. The "kinetic" relation then simply reduces to $\Delta\mu=0$. By expanding $\Delta\mu$ about a reference point, we show in the Appendix that this condition amounts to [at $z=\zeta(x,t)$]

$$c=c_{\text{eq}}[1+\tilde{\Gamma}\kappa] , \tag{5}$$

where $\tilde{\Gamma}=\gamma\Omega/k_B T$ (which has a dimension of a length), with γ the line tension, k_B the Boltzmann constant, T the (fixed) temperature, c_{eq} the equilibrium concentration, and κ the step curvature counted to be positive for a convex profile:

$$\kappa=-\zeta_{xx}/[1+\zeta_x^2]^{3/2} . \tag{6}$$

We should mention that crystalline anisotropy (which can enter the diffusion coefficient and the line tension) is not accounted for in this work.

Finally far away from the step—that is, for a distance larger than the diffusion length to be introduced below—the density c should reach a constant value c_∞, given by

$$c(z\to\infty)\equiv c_\infty=\tau F . \tag{7}$$

This condition simply expresses the equilibrium between deposition and evaporation.

The set of equations (2)–(7) completely describes the growth dynamics. This set supports a simple solution: a straight step (which we take at $z=0$) moving at a constant speed V_0. Let $c_0(z)$ denote the corresponding concentration profile. Equation (2) is solved by

$$c_0=A_0\exp[-z/(D\tau)^{1/2}]+\tau F , \tag{8}$$

where A_0 is an integration factor. The concentration profile extends over a characteristic distance $x_s\equiv\sqrt{D\tau}$, which we usually call the diffusion length. Inserting expression (8) into (5), we obtain $A_0=c_{\text{eq}}-\tau F$. The use of Eq. (3) determines the growth speed as a function of the control parameter,

$$V_0=\Omega x_s(F-F_{\text{eq}}) , \tag{9}$$

where $F_{\text{eq}}=\tau c_{\text{eq}}$. The quantity $F-F_{\text{eq}}$ measures the distance from equilibrium; it is the driving force for the growth process.

III. LINEAR STABILITY ANALYSIS

In this section we briefly consider, for completeness, the linear stability analysis of Bales and Zangwill[18] and present some preliminaries to prepare the nonlinear analysis. We study the regression of fluctuations by setting

$$c(x,z,t)=c_0(z)+c_1(z)e^{iqx+\omega t}+\text{c.c.} , \tag{10}$$

$$\zeta(x,t)=\zeta_1 e^{iqx+\omega t}+\text{c.c.} , \tag{11}$$

where c.c. stands for complex conjugate, q is the wave number, ω is the amplification (or attenuation) rate that we wish to determine, and c_1 and ζ_1 are small quantities. Note that because the Fourier modes do not couple in the linear problem, it suffices to consider one Fourier component. The scheme consists now in inserting Eqs. (10) and (11) into the three basic equations (2), (3), and (5) and the boundary condition (7) and linearize the resulting equations in c_1 and z_1. The diffusion equation is linear, and c_1 is easily found to be

$$c_1=A_1e^{-\Lambda_q z}, \quad \Lambda_q\equiv\sqrt{q^2+1/x_s^2} , \tag{12}$$

where use has been made of Eq. (7). A_1 is an integration factor. Inserting (12) into the linearized version of Eqs. (3)–(5), we obtain two algebraic equations for (A_1,ζ_1),

$$A_1-[\tilde{\Gamma}c_{\text{eq}}q^2-\tau\Delta F/x_s]\zeta_1=0 , \tag{13}$$

$$\Lambda_q A_1+[\omega D\Omega+\tau\Delta F/x_s^2]\zeta_1=0 , \tag{14}$$

where $\Delta F=F-F_{\text{eq}}$. The condition that this system have a nontrivial solution leads to the dispersion relation

$$\omega=\Omega D[-q^2\tilde{\Gamma}c_{\text{eq}}\Lambda_q+(\tau\Delta F/x_s)(\Lambda_q-1/x_s)] . \tag{15}$$

This is the dispersion relation derived by Bales and Zangwill.[18] The first term inside the brackets, which is always negative, expresses the stabilizing effect due to line tension, while the second one is always positive and represents the destabilization of diffusion. The straight step is stable if $\omega<0$ for all q's. Conversely it is unstable if there exists at least one wave number q for which $\omega>0$. The critical condition is attained when $\omega=0$ for a particular value of q, say q_c, while it is negative for all other q's. This occurs when the two conditions

$$w(q_c)=0, \quad [\partial w/\partial q]_{q=q_c}=0 \tag{16}$$

are met. These two equations determine the critical condition on the onset of instability and the wave number of the bifurcating mode. By using the dispersion relation (15), we obtain from (16) that the bifurcation is characterized by

$$q_c=0, \quad \xi\equiv\frac{\tilde{\Gamma}c_{\text{eq}}}{x_s\tau\Delta F}=\frac{1}{2} . \tag{17}$$

It can be checked that $\omega<0$ for all q's if $\xi>\frac{1}{2}$. If $\xi<\frac{1}{2}$, however, there exists a band of wave numbers corresponding to unstable modes. This band, Δq, is given close to criticality by

$$\Delta q\simeq(1/x_s)[4(1-2\xi)/3]^{1/2} , \tag{18}$$

a relation which is obtained by expanding $\omega(q)$ to leading order about the critical point given by (17). Figure 2 shows $w(q)$ in the stable and unstable regimes. The linear stability analysis tells us then that if $\xi<\frac{1}{2}$, there is a band of active modes that grow exponentially in the course of time and that therefore the linear approximation will cease to be valid. The nonlinear terms will then intervene to saturate the linear amplification, and drive the system to a finite amplitude deformation. If the dis-

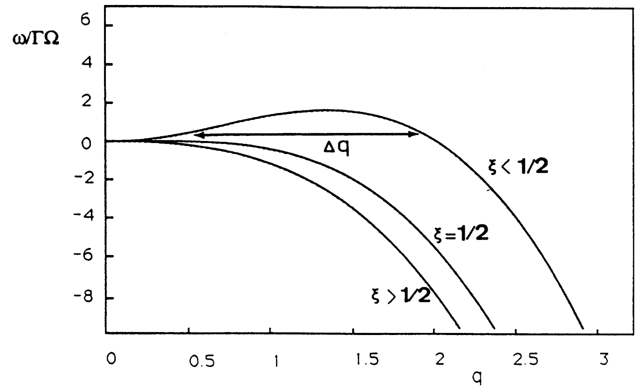

FIG. 2. The growth rate vs the wave number in the stable ($\xi > \frac{1}{2}$), critical ($\xi = \frac{1}{2}$), and the unstable ($\xi < \frac{1}{2}$) regimes. Δq designates the band of active modes.

tance from the threshold of instability is finite, then a resort to a numerical study seems unavoidable. If this distance is small enough, however, we can carry out an analytical treatment that permits us to get important information about the nonlinear evolution.

IV. NONLINEAR EVOLUTION EQUATION

Before developing our program, we would like first to rewrite the basic equations in a dimensionless form. For that purpose, we take x_s and τ as space and time units, and introduce $u = (c - \tau F)$ as a new concentration function. Equation (2) takes the form

$$\Delta u - u = 0 \,, \tag{19}$$

subject to mass conservation [Eq. (3)] at the step [$z = \zeta(x,t)$],

$$\Gamma/\xi + \dot{\zeta} = \Omega[u_z - \zeta_x u_x] \,, \tag{20}$$

to the chemical equilibrium condition [Eq. (5)]

$$u = -\Gamma/\xi - \Gamma\zeta_{xx}/[1+\zeta_x^2]^{3/2} \,, \tag{21}$$

and to condition at infinity

$$u(z \to \infty) = 0 \,, \tag{22}$$

where $\Gamma = \tilde{\Gamma} c_{\rm eq}/x_s$ is a quantity having the dimension of $c_{\rm eq}$. Note that we have kept for the spatial (ζ and x) and temporal (t) variables the same notations. This should not be confusing since we will use exclusively, from now on, only dimensionless variables.

The linear stability analysis is the first step in any stability theory. Moreover it is a natural starting point for the definition of the linear problem. In particular the linear dispersion relation serves to determine the characteristic length and time scales for the evolution of the perturbation. We first rewrite the dispersion relation (15) in a dimensionless form (and keep the same notation),

$$\omega = \frac{\Gamma\Omega}{\xi}[\sqrt{1+q^2} - 1] - \Omega\Gamma q^2\sqrt{1+q^2} \,. \tag{23}$$

Since the bifurcation occurs for $\xi = \frac{1}{2}$ and we are interested in a situation close enough to the onset of instability, we introduce a small parameter ϵ defined as

$$\epsilon = \tfrac{1}{2} - \xi \,, \tag{24}$$

which measures the distance from the threshold. Then it is easy to show that the cutoff wave number $q_{\rm cut}$ [defined by $\omega(q_{\rm cut}) = 0$] is given by

$$q_{\rm cut} \simeq (8\epsilon/3)^{1/2} \,. \tag{25}$$

This means that the band of wave numbers corresponding to the active modes scales as $\epsilon^{1/2}$. The corresponding length scale for modulations of the structure in real space is of the order of $\epsilon^{-1/2}$. We therefore introduce a new variable X related to x by

$$X = \epsilon^{1/2} x \,. \tag{26}$$

In the new variable X the structure will evolve on the scale of unity. The advantage of introducing X is simply that ϵ will appear explicitly in the governing equations and that therefore our perturbation expansion can be made systematic. Now in order to determine the time scale for the dynamics, we consider a typical wave number of order $\epsilon^{1/2}$, and expand the resulting growth rate (23) to leading order in the small parameter. The result is

$$\omega \simeq \Gamma\Omega[2\epsilon q^2 - \tfrac{3}{4}q^4] \,, \tag{27}$$

where q^4 should be retained in the expansion since $q \sim \epsilon^{1/2}$. It follows from Eq. (27) that ω scales as ϵ^2. This entails that the dynamics in the vicinity of the threshold evolves on a time scale of the order of ϵ^{-2}. As we did for the spatial dependence, we also introduce a slow temporal variable T defined by

$$T = \epsilon^2 t \,. \tag{28}$$

Now we are to understand that the step position depends only on the slow variables (X, T), while the diffusion field depends both on these slow variables and on the fast variable z. In order to somehow keep homogeneous notations, we will use Z instead of z for this variable. Finally since we expect the step deformation to be of the order of the distance from the threshold, we find it convenient—as we did it above for x and t—to introduce a new function $H(X,T)$ defined as

$$\zeta(X,T) = \epsilon H(X,T) \,, \tag{29}$$

and which is of order unity. We then expand u and H in power series of ϵ,

$$u = u_0 + \epsilon u_1 + \epsilon^2 u_2 + \ldots, \tag{30}$$

$$H = H_0 + \epsilon H_1 + \epsilon^2 H_2 + \cdots \,. \tag{31}$$

In terms of the new variables (X, T, Z, H), Eq. (2) takes the form

$$\epsilon u_{XX} + u_{ZZ} - u = 0 \,, \tag{32}$$

subject to the condition

$$u(Z \to \infty) = 0 \,, \tag{33}$$

while at $Z=\epsilon H$ we have [see Eqs. (20) and (21)]

$$\frac{\Gamma}{1/2-\epsilon}+\epsilon^3 H_T=\Omega(u_Z-\epsilon^2 H_X u_X) , \tag{34}$$

$$u=\frac{\Gamma}{\epsilon-1/2}-\epsilon^2\Gamma\frac{H_{XX}}{[1+\epsilon^3 H_X^2]^{3/2}} . \tag{35}$$

The scheme now is to insert (31) into the basic equations (32)–(35) and use the resulting equations to deduce successively higher-order contributions in powers of ϵ.

Order ϵ^0

To this order, Eq. (32) reads

$$u_{0ZZ}-u_0=0 , \tag{36}$$

which is solved by $u_0=A_0 e^{-Z}$. Making use of the boundary conditions at $Z=\epsilon H$, we obtain

$$u_0=-2\Gamma e^{-Z} . \tag{37}$$

This solution is nothing but the straight step solution [see Eq. (8)] expressed in the new variables.

Order ϵ^1

To this order Eq. (32) becomes

$$u_{1ZZ}-u_1=0 , \tag{38}$$

whose solution is given by

$$u_1=A_1 e^{-Z} , \tag{39}$$

where A_1 is an integration factor which should be treated as a function of the slow variables (X,T), since Eqs. (32)–(35) are partial differential equations. Equations (34) and (35) expanded up to order ϵ give

$$u_{1Z}+H_0 u_{0ZZ}=4\Gamma , \tag{40}$$

$$u_1+H_0 u_{0Z}=-4\Gamma . \tag{41}$$

From now on all the functions entering the boundary conditions at the step are understood to be evaluated at $Z=0$. Using the zeroth-order solution, we find that the above two equations are identical. The factor A_1 is found to be given by

$$A_1=-4\Gamma-2\Gamma H_0(X,T) . \tag{42}$$

As stated above, A_1 is indeed a function of the slow variables. At this stage H_0 is an unknown quantity.

Order ϵ^2

To order ϵ^2, u_2 obeys an inhomogeneous equation

$$u_{2ZZ}-u_2=-A_{1XX}e^{-Z} , \tag{43}$$

whose solution consists of the sum of a homogeneous solution and a particular solution

$$u_2=A_2 e^{-Z}+\frac{ZA_{1XX}}{2}e^{-Z} , \tag{44}$$

where A_2 is an integration factor. To this order the chemical equilibrium equation (35) provides

$$u_2+H_0 u_{1Z}+H_1 u_{0Z}+\tfrac{1}{2}u_{0ZZ}H_0^2=-\Gamma(H_{0XX}-8) , \tag{45}$$

while the mass conservation equation (34) yields

$$u_{2ZZ}+H_0 u_{1XX}+H_1 u_{0ZZ}+\tfrac{1}{2}H_0^2 u_{0ZZZ} . \tag{46}$$

Exploiting the solutions of order ϵ^0 and ϵ, we obtain the expression for A_2:

$$A_2/\Gamma=-4H_0-H_0^2-H_{0XX}-2H_1-8 . \tag{47}$$

We can mention that, as in the previous order, the two equations (45) and (46) give identical results. Still at this order neither H_0 nor H_1 are determined. The really interesting result emerges at next order where a solvability-like condition emerges as a constraint on H_0. This constraint is nothing but the evolution equation that we wish to determine.

Order ϵ^3

To this order u_3 obeys

$$u_{3ZZ}-u_3=-\left[A_{2XX}+\frac{ZA_{1XXXX}}{2}\right]e^{-Z} , \tag{48}$$

the solution of which can be written as

$$u_3=A_3 e^{-Z}+\frac{A_{2XX}Z}{2}e^{-Z}+\frac{A_{1XXXX}}{8}(Z^2+Z)e^{-Z} , \tag{49}$$

where A_3 is an integration factor. Using the boundary condition (34), we find

$$\begin{aligned}16\Gamma+\Omega^{-1}H_{0T}=&-H_{0XX}u_{1X}+u_{3Z}+H_0 u_{2ZZ}+H_1 u_{1ZZ}\\&+H_2 u_{0ZZ}+\tfrac{1}{2}H_0^2 u_{1ZZZ}\\&+H_0 H_1 u_{0ZZZ}+\tfrac{1}{6}H_0^3 u_{0ZZZZ} .\end{aligned} \tag{50}$$

Finally Eq. (35) reads at this order

$$\begin{aligned}u_3+H_0 u_{2Z}+H_1 u_{1Z}+H_2 u_{0Z}+\tfrac{1}{2}H_0^2 u_{1ZZ}+H_0 H_1 u_{0ZZ}&\\+\frac{1}{6}H_0^3 u_{0ZZZ}=-16\Gamma-\Gamma H_{1XX} .&\end{aligned} \tag{51}$$

After having used the previous orders solutions, both with (49), one discovers that the above two boundary equations have many terms in common; in particular, this holds for all the terms containing H_1 and H_2. A simple algebraic manipulation of the two equations leads to the cancellation of these terms. The remaining part is the sought-after evolution equation

$$(\Omega\Gamma)^{-1}H_{0T}=-2H_{0XX}-\tfrac{3}{4}H_{0XXXX}+H_{0X}^2 . \tag{52}$$

The second piece of information that we can get from the set of two boundary equations is the determination of A_3 as a function of $H_0, H_1 \ldots$. Since we are only interested in the extraction of the evolution equation to leading order, there is not need to go further in the ϵ expansion.

Note that if the nonlinear term is neglected, and if we set $H_0 \sim e^{\omega\epsilon^2 t+x\sqrt{\epsilon}}$ (with x and t being the original fast variables), we immediately recover the linear dispersion relation (27). Let us now go back to the original dispersion relation (23) and make some comments. The first term on the right-hand side (rhs) represents the effect of the diffusion (we can easily check that the ratio Γ/ξ is independent of line tension), while the second one represents the line-tension effect. The expansion, close to the critical point, of ω [Eq. (27)] can be rewritten as $\omega \simeq \Gamma\Omega[(1-2\xi)q^2-(3/4)q^4]$, in view of the definition of ϵ [see Eq. (24)]. The q^2 term is composed of two terms: a stabilizing term $(-2\xi q^2)$ coming from the line-tension contribution in the original dispersion relation, and a destabilizing one $(+q^2)$ which originates from terrace diffusion. We can thus say that the diffusion causes a renormalization of the line-tension effect. We can speak of an "effective" line tension whose sign is determined by that of $2\xi-1$. Below the instability threshold the effective line tension is positive (since $\xi > \frac{1}{2}$) and it becomes negative when $\xi < \frac{1}{2}$, which is the condition of instability met before. Since in our derivation we were interested in the situation above the threshold (by assuming $\epsilon > 0$), we got a negative effective line tension signaled by the presence of a negative sign in front of H_{XX} in Eq. (52). Had we let ϵ have an arbitrary sign, we would then have obtained a step function multiplying H_{XX}. The fourth derivative in Eq. (52) is stabilizing. The q^4 term in the linear dispersion relation can be interpreted as representing an effective diffusion along the step. Indeed assume that there is a lack of homogeneity along the step in the distribution of gas atoms. If that inhomogeneity were the only effect that causes the growth, then the growth velocity $(\sim H_T)$ would be simply proportional to the divergence of the mass current along the step, due to mass conservation. Since the mass current is proportional to the derivative of the chemical potential (Onsager's law), the velocity is proportional to the second derivative (for a one-dimensional system, and when nonlinear effects, which enters through the tangential vector, are neglected) with respect to X, $H_T \sim \mu_{XX}$, μ being the chemical potential. The chemical potential is a function of the deformation H. For a slowly varying deformation the chemical potential can be expanded as a function of the derivatives of H. Due to symmetry H_X is absent. The first term (up to an additive constant which can be absorbed in H_T) is $\mu \sim H_{XX}$, which entails that $H_T \sim \mu_{XX} \sim H_{XXXX}$.

We can attribute to the presence of the nonlinear term in Eq. (52) a simple meaning. First of all it is clear in order to saturate the linear amplification of the perturbations that the nonlinearity should be taken into account. What type of nonlinearity could we expect *a priori?* Two other candidates could have been H_0^2 or $H_{0X}H$, but then we would break the translational symmetry: the invariance of the governing equation under the transformation $H_0 \to H_0 + a$, where a is a real constant. This breaking is not physical since the straight step position can be taken anywhere; as a consequence these two candidates are immediately disqualified.

There is in fact a more enlightening way—and a more robust condition—from which the nonlinear term in Eq. (52) follows. Let us first rewrite that equation in a canonical form,

$$h_t = -h_{xx} - h_{xxxx} + \tfrac{1}{2}h_x^2 \,, \tag{53}$$

where the new variables (h,x,t) are related to (H_0,X,T) by

$$h = H_0, \quad x = X\sqrt{\tfrac{8}{3}}, \quad t = 16\Gamma\Omega T/3 \,. \tag{54}$$

The evolution equation thus reduces to a *universal* one in the sense that all the parameters scale out. This feature is more general: if we multiply all the terms in Eq. (52) by different constants, it is always possible to find a transformation of the type (54) such that the resulting equation becomes free of any parameter.

Let us now pursue our remark on the nonlinear term in Eq. (52). For that purpose we find it convenient to set $h_X = v$ and write the equation for v:

$$v_t - v v_x = -v_{xx} - v_{xxxx} \,, \tag{55}$$

an equation that is directly obtained from (52) upon differentiation with respect to x. Equation (55) is reminiscent of the Navier-Stokes equation with a negative viscosity and with higher-order "viscous" terms (the term proportional to v_{xxxx}). Note that the presence of a minus sign in front of the nonlinear term is unimportant, since that sign can be changed upon the transformation $v \to -v$. As is the case with the Navier-Stokes equation, Eq. (55) is invariant under the transformation

$$v' = v + v_0, \quad x' = x - v_0 t, \quad t' = t \tag{56}$$

where v_0 is a constant. This is nothing but a Galilean transformation. In order to check this invariance it suffices to realize that under this transformation v_t transforms into $v_{t'} + v_0 v_{x'}$, and $v v_x$ transforms into $v' v_{x'} + v_0 v_{x'}$, while the other terms remain invariant. We see thus that the nonlinear term is necessary to restore the Galilean invariance that would be broken by the v_t— term in the linear regime.

It should be added here that v, by its definition, is a *parallel transported* quantity, in the sense that it is everywhere tangential to the real step profile, therefore making the similarity between the Kuramoto-Sivashinsky equation and the "Navier-Stokes" equation apparent.

We will see in the next section that the Galilean invariance, obvious as it may appear, has nevertheless a strong consequence on dynamics.

V. A SHORT REVIEW OF THE PROPERTIES OF THE EVOLUTION EQUATION

It would be extremely ambitious to review in this short paragraph all the properties of the KS equation. We will simply present briefly only those properties that are relevant for our purposes.

First, our experience of patterning instabilities, such as those encountered in Rayleigh-Benard and Taylor systems, or interfacial instabilities in crystal growth (which is a closer system to the one we are concerned with), may lead one to think that above the instability threshold, the system will develop a cellularlike structure, which will eventually become more and more complex, ultimately giving rise to long fingers or even to dendrites. As a first step, it is natural to ask whether the above evolution equation (52) may admit periodic-cellular structures, and if so are these structures stable, and under which conditions? First, it is easy to see that in the frame of reference moving with the straight step, a frame in which Eq. (52) is written, a periodically deformed step cannot be stationary. Indeed integration of Eq. (53) over a period (and due to periodic boundary conditions) leads to

$$\langle h_t \rangle = \tfrac{1}{2}\langle h_x^2 \rangle > 0 \,, \tag{57}$$

where $\langle \cdots \rangle$ signifies the mean value over a period. Inequality (57) means that the mean step position is permanently drifting. This implies that steady-state periodic solutions, if they are to exist, move with an additional constant speed v in the z — direction, given by

$$v = \tfrac{1}{2}\langle h_{0x}^2 \rangle \,, \tag{58}$$

where $h_0(x)$ designates the steady solution obeying

$$v = -h_{0xx} - h_{0xxxx} + \tfrac{1}{2} h_{0x}^2 \,. \tag{59}$$

Equation (59) has been solved by many authors.[25] Before presenting the results, some preliminary discussion is necessary. The linear dispersion relation extracted from Eq. (53) (by taking $h \sim e^{iqx+\omega t}$) is given by $\omega = q^2 - q^4$. The cutoff wave number is now given by $q_0 = 1$ [obtained by setting $\omega(q_0) = 0$]. Let us consider a periodic solution with a basic wave number 9. It is convenient to introduce a parameter $\mathcal{A}$ defined by $\mathcal{A} = \lambda/\lambda_0$, where $\lambda = 2\pi/q$ and $\lambda_0 = 2\pi/q_0$. $\mathcal{A}$ can be thought of as an *aspect ratio,* measuring the ratio of the actual periodicity to the characteristic "stability" length. It is interesting to plot ω as a function of $\mathcal{A}$ (Fig. 3). If $\mathcal{A} < 1$ ($\lambda < \lambda_0$), it is clear that the perturbations about $h = 0$ are linearly stable, and the only solution found is the straight solution. When $\mathcal{A} > 1$ a solution with a finite amplitude merges. Close to $\mathcal{A} = 1 \equiv \mathcal{A}_0$, the deformation amplitude behaves as $\sqrt{\mathcal{A} - \mathcal{A}_0}$, as in a usual Landau-Ginzburg-like transition. This steady solution is called the cellular solution [Fig. 4(a)]. As $\mathcal{A}$ approaches the value 2, the original cellular solution develops a tip splitting and, approximately, in the interval $\mathcal{A} \in [2,3]$ the steady solution consists in 2-cells of the type shown in Fig. 4(a). On further increase of $\mathcal{A}$, a similar scenario occurs; that is (approximately) in the interval [3,4] a 3-cells solution appears. An interesting result occurs when $\mathcal{A}$ is slightly larger than 4: a new solution merges, a solution which has been called a strange solution. Figure 4(b) displays this solution. On increasing $\mathcal{A}$ a 4-cells solution takes place, and so on. If the system size L is large in comparison to the basic periodicity λ, then we would see no difference in morphology between 2-cells, 3-cells, etc., solutions (since each "entity" is the same), but only between the strange and the cellular solution.

Assume that the system size is a quantity that we can vary as we wish in a certain physical situation. More precisely, assume that the system size is exactly the periodicity λ. If the size is smaller than the stability length, then the only solution is the straight step one. By increasing the size (which amounts to increasing $\mathcal{A}$, as done above), we would see that our system is composed in total of one cell at small size; then the number of cells will increase as

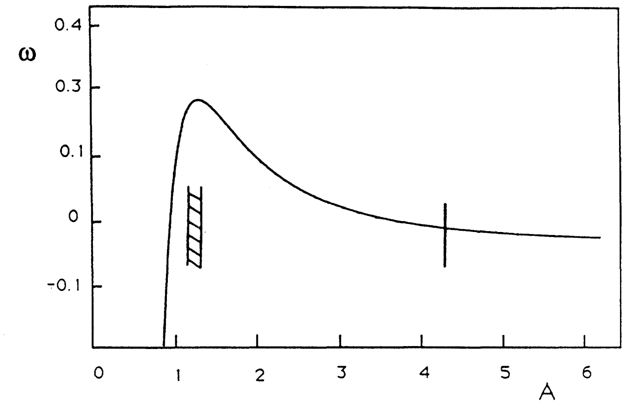

FIG. 3. The growth rate as a function of the aspect ratio $\mathcal{A}$. The hatched part to the left refers to the domain of stability of the cellular solution, while the one to the right (which is so narrow that it appears as a thick line) represents the stability interval of the strange solution (see later in the text).

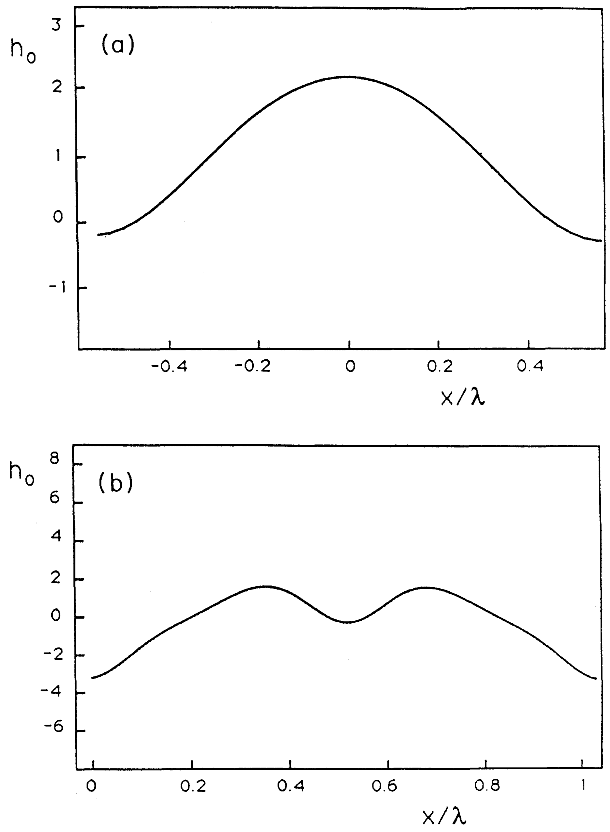

FIG. 4. The cellular solution (a) and the strange one (b).

described above, the development of a strange solution will occur, and so on. It is worth mentioning that since we are considering only steady-state solutions, we can neither say that those solutions are everywhere stable (and therefore admissible in a real situation), nor that solutions may exist other than the steady ones. Hyman, Nicolaenko, and Zaleski[26] have integrated the full dynamical equation by considering moderate aspect ratios going from $\mathcal{A}=1$ to $\mathcal{A}=9$, and have found, besides the solutions described above, other small intervals where rather complex dynamics take place. These unsteady solutions result from the loss of stability of the steady solutions.

If one believes—as suggested by experimental observations—that the width of each cell is small in comparison to the system size, one should then in principle integrate Eq. (53) over a size such that $\mathcal{A} \gg 1$. Let us as a first stage consider the following question. Are the above steady solutions stable? Under which circumstances? If a solution is stable against all infinitesimal perturbations, then we can expect *a priori* that this solution would be realizable. If, on the contrary, the solution becomes unstable, then the first question to ask is about the nature of the instability. Then in order to investigate the subsequent development of the instability, a full integration of the evolution equation is necessary. The linear stability analysis is a very important step informing us that something is happening in a certain region (e.g., in a certain interval of $\mathcal{A}$). We will consider here that the system size is large enough so that all perturbation wavelengths are permissible. The study of the stability of the steady solutions can be achieved by linearizing the full equation (53) about the steady solution $h_0(x)$. This results in a linear equation with periodic coefficients, due to the periodicity of h_0. The resulting eigenvalue problem is solved by making use of the Floquet-Bloch theorem.[25] The result that emerges from this analysis[25] shows that the intervals of stability of the cellular and strange solution are extremely narrow. More precisely the cellular solution is stable in the interval $\mathcal{A} \in [1.195, 1.305]$, while the strange solution is stable in a much narrower interval $\mathcal{A} \in [4.22, 4.23]$. These stability domains are delimited by long-wavelength oscillatory instabilities. The nature of this instability can be traced back to symmetry properties. Indeed, if $h_0(x)$ is a solution, then $h_0(x+\phi)$, where ϕ is a constant, is also a solution for an infinite system. This means that the translational (Goldstone) mode is a neutral mode. This "perturbation" is of infinite wavelength. If one wants to know whether such a perturbation would really be dangerous, one should check what happens to the spectrum if the wavelength of the perturbation is not infinite but very large (exactly as if we were doing a linear-response theory to calculate, for example, the susceptibility of a given system). The usual result that emerges is that the instability that takes place is of the Eckhaus[27] type. This is a phase instability which manifests itself as a long-wavelength modulation of the periodicity of the structure, a modulation which induces a phase diffusion (expressed by an Onsager-like law), and which will ultimately lead to the creation or destruction of a cell according to whether the original (unstable) state has a large or a small basic periodicity. If the only symmetry were the translational invariance along the x direction, we would then be simply faced with this diffusive instability. There is, however, another group of symmetry: the Galilean invariance evoked in the preceding section. As is the case with the Eckhaus instability, long-wavelength Galilean distortions are dangerous and also of diffusive type. Since the two dangerous modes are coexisting and coupled to each other, the diffusive character of the bare perturbation can transform into a propagative one, which explains[28] the possibility for the creation of long-wavelength oscillatory instabilities in the KS equation. Of course if the system size is small (say $\mathcal{A}$ of the order of a few unities), these instabilities would not manifest themselves, and would be suppressed by boundary conditions. As a consequence, the dynamics would be relatively simple. Indeed, we would get stable steady solutions, and other regular dynamics (as simple oscillations).[26] For large sizes, however, and because the domains of stability of the steady solutions are extremely narrow, any small perturbation will easily "push" the system in the unstable region. Even if the system turns out to have locally a periodicity in the stable regime, the propagative character of the perturbations will easily cause a contamination of local order. What would we then observe? Numerical solution[21] of the KS equation has shown that even for moderate aspect ratios (say $\mathcal{A} \sim 10$), and for larger values, the dynamics are always chaotic in the time and space point of view.

In concluding this section, we remark that in a system described by a KS equation with a lateral extent approximately ten times (or larger) the stability length, obtained from a linear theory, spatiotemporal chaos will always prevail.

Finally as our treatment is expected to be valid close enough to the threshold obtained from the linear theory, it is important to ask whether this conclusion survives even far from the threshold or not. Preliminary results due to Saito and Uwaha,[19] who investigate the step-flow growth by means of lattice-gas simulation, tend to confirm this picture even at finite distances from the threshold.

VI. A NAIVE ATTEMPT FOR STEP-STEP INTERACTION

In this attempt, we try to include some aspect of the step-step interaction. We consider that the step is surrounded by steps of the same sign, as in a vicinal surface topology. We consider, in a first level, that the steps are moving altogether in the straight regime by a velocity V_0. Our main assumption here is that we assume that the steps surrounding the step reference are rigid, in that during the motion of this step about its original position in the steady growth, the surrounding steps remain in the same state. We should mention that in the linear theory of Bales and Zangwill,[18] the steps are supposed to move in a synchronized way. Of course, in reality there is always a certain incoherence between the step motion, and there is no reason that all the steps move together as a whole. In this situation, each step will see permanently its distance from the neighboring steps either increasing or decreasing. As a consequence, the step reference will be subject to a net repulsion (in one sense or another) which tends to reduce its excursion. As an extreme limit we assume that only the step reference is subject to fluctuations about its mean position. This is not thought of as being realistic, but simply as modelization to understand the role of repulsion. The interactions can be either mediated by diffusion of adatoms, or of other types such as elastic interaction.

Assume that the step reference moves slightly towards the (supposed rigid) step that is ahead of it. The elastic repulsion, for example, will tend to act against such a tendency. The same will happen if the step goes towards the one which is behind it. The simple way to model this repulsion consists in introducing a dampinglike term that stabilizes all perturbations, and, in particular, a homogeneous perturbation. To do so we simply introduce a linear stabilizing term in the original equation (53),

$$h_t = -\alpha h - h_{xx} - h_{xxxx} + \tfrac{1}{2} h_x^2 \,, \tag{60}$$

where α is a phenomenological positive (in order to be stabilizing) parameter. Note that if we wish the repulsive interaction to act also on a straight step, this repulsion should necessarily give a linear contribution in the evolution equation. It can be easily checked that now there is no scale transformation which allows us to scale the parameter α out of the equation.

The linear dispersion relation that follows from Eq. (61) is given by

$$\omega = -\alpha + q^2 - q^4 \,. \tag{61}$$

The bifurcation is now characterized by [see Eq. (16) for the definition of the bifurcation]

$$\alpha \equiv \alpha_c = \tfrac{1}{4}, \quad q \equiv q_c = 1/\sqrt{2} \,, \tag{62}$$

α_c is the critical value below which the $h = 0$ solution becomes unstable. Shown in Fig. 5 is the neutral curve (defined by $\omega = 0$) in the (α, q) plane inside which the basic solution is unstable. The maximum of the curve is nothing but the bifurcation point expressed by (62). The situation now is very different from the one where $\alpha = 0$. Indeed slightly above the instability threshold ($\alpha < \alpha_c$), there is a small band, when compared to the bifurcation wave number q_c, of active modes. We have seen that when $\alpha = 0$, the bifurcation wave number is exactly zero. This means that close above the threshold (say 10% or 20%), if one takes a small enough wave number (in comparison to the cutoff wave number) inside the instability band, then the higher harmonics ($2q, 3q, \ldots$) will be active also. This increases quickly the number of degrees of freedom, thus easily driving the system towards complex dynamics, which turns out to be chaotic. In the presence of the α term the situation is quite different. Indeed slightly above the threshold the only active mode is the one with wave number q_c, while the other harmonics are highly damped. The α term serves as a "filter" for perturbations. We then expect the dynamics to be simple close enough to the threshold and become more and more complex as α is reduced. Finally in the limit of a vanishing α, we should recover the spatially and temporally chaotic solution.

We have made a long study of Eq. (60), both numerically and analytically. We intend to devote a separate publication to this study. Here we will simply summarize the results. This is done in Fig. 6. We observe there that

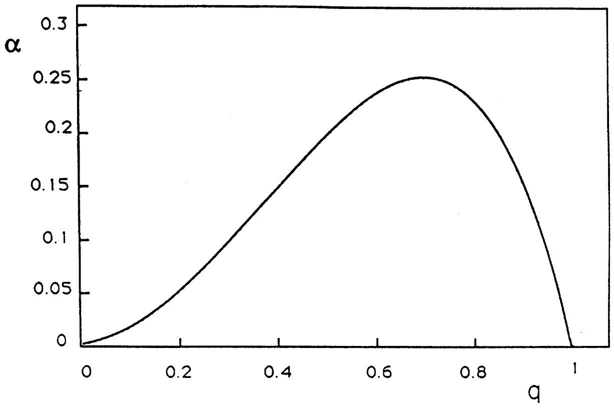

FIG. 5. The neutral curve in the $\alpha - q$ plane, inside which the solution $h = 0$ is unstable.

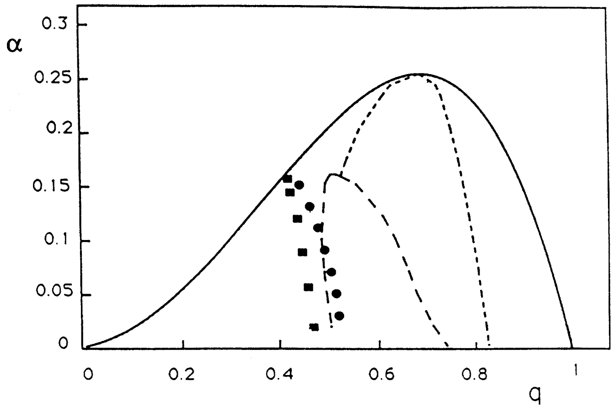

FIG. 6. An overall picture of the stability domains. Full line, neutral curve; short-dashed line, Eckhaus boundaries, outside which the cellular solution is unstable against long-wavelength phase modulations. The long-dashed line delimits the domain of stability of the cellular solution against oscillatory modes. Close enough to the maximum of the curve which delimits the oscillatory instability, the most dangerous fluctuations have a periodicity which is double of the basic one. Far below from the maximum, the periodicity of the perturbation is generically "irrationally" related to the basic one (see the text). The squares represent the boundary below which the steady-cellular solution with the basic wave number q ceases to exist and merges with a solution having $2q$ as a basic wave number (tip-splitting instability). Before this occurs, a branch of parity-broken traveling solutions appears below the line represented by the circles.

a steady-cellular solution of the type shown in Fig. 4(a) is stable over a wide range of parameters. This domain of stability is delimited by complex boundaries which we will present here by avoiding many details. First the dashed part represents the Eckhaus boundary, mentioned in the preceding section. In the long-wavelength range in Fig. 6, the only instability is the Eckhaus one. In the short wave-number range, however, at least two types of dynamics appear, resulting from the loss of stability of the cellular solution. The first one is a homogeneous instability characterized by the loss of the parity symmetry, and as a result of this symmetry breaking the structure travels sideways. We call this mode of growth the *broken-parity-traveling mode.* This mode was identified in other physical contexts, both experimentally[29] and theoretically.[30] Another mode of growth is the oscillatory one where, in the simple situation, each cell width oscillates in phase opposition with its neighbors. This is referred to as the *vacillating-breathing* mode. Figure 7 shows these two modes of growth. One sees there that in the oscillatory case the spatial periodicity has doubled. This feature is very similar to the dimerization problem resulting from Peierls instability in one-dimensional conductors. The difference is that the present instability is oscillatory, while the Peielrs one is steady. We have also found other dynamics where the spatial periodicity of the oscillating mode is *irrationally* related to that of the basic one (note that for the *vacillating-breathing* mode the ratio of the periodicity to that of the basic one is equal to 2). Other interesting dynamics discovered in this equation consist in complex mixtures of all these modes.

VII. CONCLUSION

We have treated the nonlinear evolution of a terrace edge during step flow growth. In a first level we have considered a single step. We have shown that the dynamics are described close to the threshold by a Kuramoto-Sivashinsky equation. For large lateral sizes this equation produces spatiotemporal chaos. In contrast to the situation dominated by thermal fluctuations, the step in this idealized limit would exhibit an erratic motion of a completely *deterministic* origin. It is an important task for future investigation to see what would happen if these two features coexisted.

The overall picture that we have drawn from analytical extraction of the nonlinear evolution equation is expected to be valid close enough to the threshold. It turns out, however, from preliminary lattice-gas simulations,[19] that this picture seems to persist even at a finite distance above the instability threshold.

In a second stage we have attempted to include phenomenologically the interaction of the reference step with a "rigid" one. We have shown—with the proviso that our modeling of interaction is legitimate—that the step-step interaction may strongly modify the dynamics: the cellular structure can remain stable over a finite range. As there are some observations where the step meandering is rather regular, it is tempting to speculate that this fact signals the importance of step-step interaction in the growth dynamics. We are tempted to evaluate the strength of the interaction in comparison to appropriate

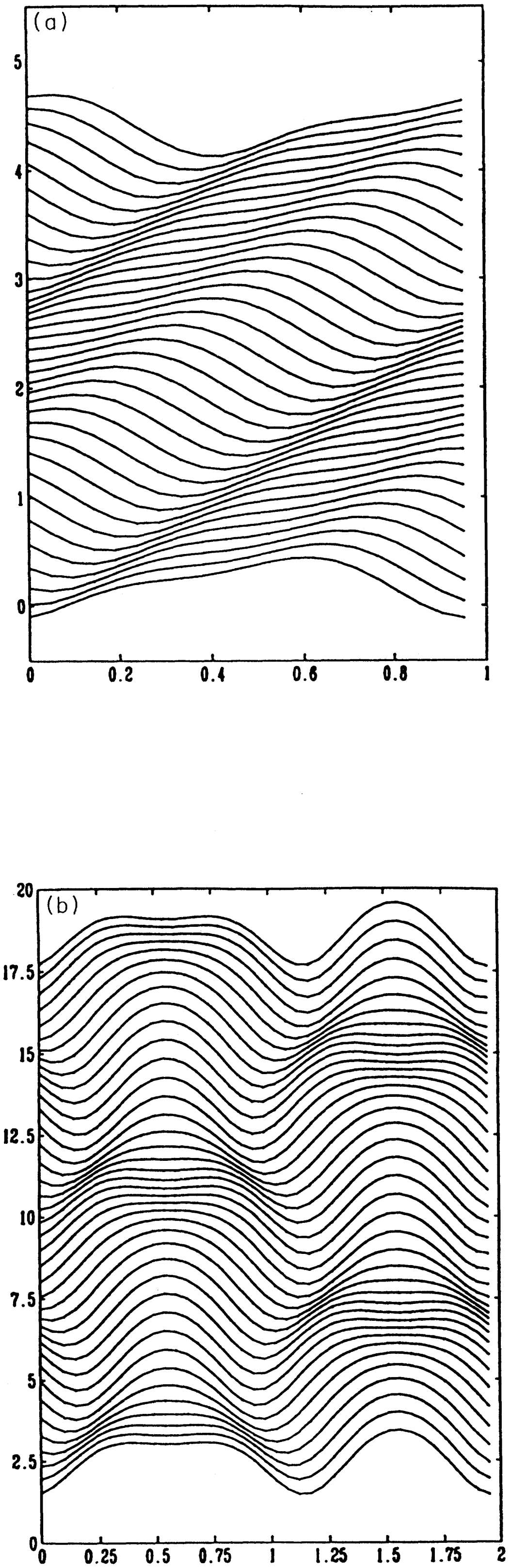

FIG. 7. Two types of growth modes: the broken-parity mode (a) and the vacillating-breathing one (b).

characteristic amplitude dynamics. This calculation should be crucial to settle the important question pertaining to the role of the step-step interaction in the process of pattern selection in a out-of-equilibrium situation. Finally it is noteworthy that our treatment can in principle be extended to situations where kinetics, crystalline anisotropy, and full step-step interaction are included.

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APPENDIX; DERIVATION OF THE SMOOTHNESS CONDITION AT THE STEP

In this appendix, we give a simple derivation of the boundary condition at the step [Eq. (25)]. In the solid phase the chemical potential μ_s is approximately given by the free energy per harmonic oscillator, and the $p_s v_s$ term (p_s is the pressure and v_s the atomic volume) does not play a crucial role, since v_s is small as compared to the volume occupied by a gas atom. The solid chemical potential is a function of p_s and temperature T, and its precise form is in fact unimportant. Since the step is assumed to evolve in an isothermal environment, we shall omit the symbol T in the arguments. The gas atoms on the terrace are well described by an ideal gas potential, $\mu(c,p_g)$, where c is the superficial density and p_g the (surface) gas pressure. Let the gas pressure be fixed by the operator to a value p_g, and denote by $c_{\rm eq}$ the equilibrium concentration for a straight step. The condition of extremely fast kinetics implies that the chemical potentials in the gas and the solid on both sides of the step are (virtually) equal,

$$\mu_s(p_s)=\mu(p_g,c)\ . \tag{A1}$$

For a small departure about the equilibrium reference point $(p_g,c_{\rm eq})$, Eq. (A1) provides to leading order

$$(p_s-p_g)(\partial\mu_s/\partial p)_{p=p_g,c=c_{\rm eq}} = (c-c_{\rm eq})(\partial\mu_g/\partial c)_{p=p_g,c=c_{\rm eq}}\ . \tag{A2}$$

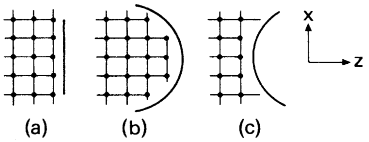

FIG. 8. Schematic view of the bond configuration for a straight step (a), a convex step (b), and a concave step (c).

The quantity $(\partial\mu_s/\partial p)=\Omega$ (recall that Ω is the atomic area of the solid), which is an exact thermodynamic equality. Note that it is Ω and not v_s which enters here, since the pressure refers to the reversible adiabatic work to change the area and not the volume. For the gas, we take the ideal situation, so that $(\partial\mu_g/\partial c)=k_BT/c_{\rm eq}$. Equation (A2) then becomes

$$c-c_{\rm eq}=\frac{c_{\rm eq}\Omega}{k_BT}(p_s-p_g)\ . \tag{A3}$$

For a curved step, mechanical equilibrium implies that the pressure difference should counterbalance the line-tension effect.[31] This is the Laplace law which takes the form $(p_s-p_g)=\gamma\kappa$, where κ is the step curvature counted positive for a convex profile. Indeed a convex profile means that we have a forward bulge pointing in the Oz direction. As a consequence the line tension will act on the bulge by exerting a normal force pointing in the negative z direction. To preserve mechanical equilibrium the solid pressure should be higher than the gas one, the difference being the line-tension effect, $\gamma\kappa$. By inserting the Laplace law into Eq. (A3), we obtain

$$c-c_{\rm eq}=\frac{c_{\rm eq}\Omega}{k_BT}\gamma\kappa\ , \tag{A4}$$

which is exactly Eq. (5).

There is an enlightening way to understand the sign of the curvature effect in Eq. (A4). Indeed assume that the step is straight. Figure 8(a) shows the configuration of the bonds in the solid phase. If the profile is convex [Fig. 8(b)], then atoms attached to the solid on the step will have broken bonds that will call for more adatoms close to the top of the bulge. As a consequence, the density of atoms in the gas will increase there to reduce the bonds deficit. This is exactly what Eq. (A4) expresses. Conversely, for a concave step [Fig. 8(c)], the number of neighboring atoms in the solid has increased, thus inducing a reduction of the areal density near the minimum.

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*Also at Groupe de Physique des Solides, Universités Paris 7 et 6, 2 place Jussieu, Tour 23, 75005, Paris, France.

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