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Analytical treatment for parity breaking in eutectic growth

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Using an ansatz for an asymmetric profile of the solidification front in lamellar eutectic growth, the von Neuman problem is solved. Imposing the Gibbs-Thomson condition, we derive a general expression for the tilt angle as a function of the control parameters. We find that the front undergoes a supercritical parity-breaking bifurcation at a critical value of $\sigma = d_0 l/\lambda^2$ (d_0 is the capillary length, l the diffusion length, and λ the wavelength of the pattern). We further find that parity-breaking causes a reduction of the average front undercooling. All these features agree with previous numerical results.

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During the past few years considerable experimental and theoretical efforts [1] have been devoted to the understanding of secondary instabilities of pattern-forming systems. A particularly interesting example is the so-called "solitary mode." This is an inclusion of asymmetric cells drifting along the interface. Soon after the discovery of this growth mode by Simon, Bechhoefer, and Libchaber [2], it became clear that this phenomenon is a robust feature of a large variety of one-dimensional patterns [2–6]. Coullet, Goldstein, and Gunaratne [7] put forward the idea that this mode results from a loss of stability of the initially symmetric state against antisymmetric fluctuations. They built a phenomenological model that captures some interesting features seen in experiments. It was shown later for both eutectic and liquid-crystal systems [8,9] that the "microscopic" models of growth indeed support parity-broken solutions that move transversely to the front.

Besides the case where interface dynamics can be mapped onto that of two interacting resonant modes—a situation which holds close to a codimension two bifurcation of the structureless (e.g., planar) state [10,11]—all the progress came from numerical calculations. Given the large number of material and control parameters in the eutectic system, and in the hope of guiding further theoretical and experimental investigations, it is strongly desirable to have analytical results at our disposal. The aim of this Rapid Communication is to develop an analytical treatment for parity-breaking bifurcation in eutectic growth. Let us first present the main lines that motivate our strategy.

First we recall that the Jackson and Hunt [12] theory has dealt with the existence of steady symmetric solutions. The crux of their theory was to solve the diffusion equation in the liquid phase subject to mass balance at the interface by assuming it to be planar. Their next step is to impose the Gibbs-Thomson condition consistent with the diffusion field calculated for the planar front. Emerging from this analysis is a relationship between the periodicity λ and the front undercooling $\Delta(\lambda)$; there thus exists a continuous family of solutions parametrized by the periodicity.

More recently numerical analysis [8,13] of the growth equations of lamellar eutectic structures has revealed three important results: (i) the existence, for a *given* wavelength λ, of a discrete set of symmetric solutions, and not a unique solution as found by Jackson and Hunt [12]; this means that there exist many branches $\Delta(\lambda)$, (ii) as λ increases the branches coalesce by pairs to form fold singularities where symmetric solutions cease to exist; and (iii) slightly before the fold singularity takes place, a branch of parity-broken solutions admix as a forward bifurcation. An important result [13,14] that emerged from that analysis is that the Jackson and Hunt [12] theory (which considers only the problem of symmetric growth), although it misses the discrete set and the fold singularity, describes remarkably well the lower branch of symmetric solutions found numerically [14]. In particular this also holds close to the critical point for the parity-breaking bifurcation. These results give a strong hint that the simplest next step analysis beyond that of Jackson and Hunt [12], which involves including an antisymmetric component in the front profile, should capture the essential features of the parity-breaking transition.

In order not to unnecessarily complicate the presentation in this Rapid Communication, we will use the following simplifications. (i) We will confine ourselves to a eutectic system with the two solid phases having exactly the same physical properties; (ii) we will assume that the liquid far ahead of the solidification front is maintained at its eutectic composition; and (iii) we will consider that the system evolves in an isothermal environment. Since in standard directional solidification experiments the thermal length is much larger than the wavelength of the pattern the last assumption is, beyond any doubt, justified. It is easy to convince oneself that relaxing all these assumptions would pose no specific challenge, and we are planning to report on the general situation in the future.

The model equations are by now standard [14]. Let $u = (c - c_\infty)/\Delta c$ denote the dimensionless concentration, where $c_\infty = c(z \rightarrow \infty) \equiv c_e$, c_e being the eutectic concentration and Δc the usual concentration gap between the two solid phases at the eutectic temperature. In the frame of reference where the pattern is at rest, u obeys

$$\nabla^2 u + 2u_z + 2\tan(\phi) u_x = 0\,, \tag{1}$$

subject to the conservation and the Gibbs-Thomson conditions at the interface $[z = \zeta(x)]$

$$u_z - \zeta_x u_x = \mp [1 + \zeta_x \tan(\phi)]\,, \tag{2}$$

$$u = \pm(\Delta - d_0 \kappa)\,. \tag{3}$$

Lengths are reduced by the diffusion length $l = 2D/V$, D being the diffusion constant, d_0 the capillary length, $\kappa = -\zeta_{xx}/(1+\zeta_x^2)^{3/2}$ the interface curvature, and $\Delta = (T_e - T_0)/m\Delta c$ the dimensionless supercooling, where T_e is the eutectic temperature, T_0 the temperature of the environment, and m the absolute value of the liquidus slope (in directional growth Δ is a measure of the average front position). Finally ϕ is the (unknown) tilt angle. Note that the upper (lower) sign in Eqs. (2) and (3) refers to the solid phase with the lower (higher) composition. Equations (1)-(3) should be supplemented by the mechanical equilibrium conditions at the triple points:

$$\zeta_x(0) = \tan(\theta - \phi),\quad \zeta_x(\lambda/2) = -\tan(\theta + \phi)\,, \tag{4}$$

where θ is the contact angle at the triple point.

Let us recall that in order to calculate the diffusion field Jackson and Hunt [12] assume a planar front. That is they solve Eq. (1) subject to condition (2) at $\zeta = 0$, $u_z(0) = \mp 1$. The simplest treatment that involves parity-broken solutions consists of solving the von Neuman problem by considering a small asymmetric deviation from the planar interface. The ansatz consists of assuming that the front consists of two straight segments defined by

$$\zeta(x) = \begin{cases} x\tan(\theta - \phi), & 0 \le x \le x_0 \quad (5)\\ (\lambda/2 - x)\tan(\theta + \phi), & x_0 \le x \le \lambda/2\,, \quad (6)\end{cases}$$

where x_0, the intersection point, is given by

$$x_0 = \lambda \tan(\theta + \phi)/2[\tan(\theta + \phi) + \tan(\theta - \phi)]\,.$$

To leading order in the asymmetric front deviation, and in the (standard) small Péclet number limit the diffusion field is found at length to be given by

$$u = \sum_{n=1}^{\infty} B_{2n-1} \sin\left[\frac{2\pi}{\lambda}(2n-1)x\right] + \sum_{n=1}^{\infty} D_{2n-1}\cos\left[\frac{2\pi}{\lambda}(2n-1)x\right]\,, \tag{7}$$

where

$$B_{2n-1} = \frac{2\lambda}{\pi^2(2n-1)^2} + \frac{\lambda\tan(\phi)}{\pi^2(2n-1)^2}\left\{H^{(-)} + H^{(+)}\cos\left[\frac{2\pi}{\lambda}x_0(2n-1)\right]\right\}$$
$$+\frac{\lambda - 2x_0}{\pi(2n-1)}\tan(\theta+\phi) - 2\lambda H^{(+)}\sum_{m=1}^{\infty}\frac{1-\cos[(4\pi/\lambda)x_0 m]}{\pi^3 m[4m^2-(2n-1)^2]} + \lambda H^{(+)}\sum_{m=1}^{n-1}\frac{1-\cos[(4\pi/\lambda)x_0 m]}{\pi^3 m^2[2m-(2n-1)]}\,, \tag{8}$$

$$D_{2n-1} = -\lambda H^{(+)}\tan(\phi)\frac{\sin[(2\pi/\lambda)x_0(2n-1)]}{\pi^2(2n-1)^2} - \lambda H^{(+)}\sum_{m=1}^{\infty}\frac{(2n-1)\sin[(4\pi/\lambda)x_0 m]}{\pi^3 m^2[4m^2-(2n-1)^2]}$$
$$-\lambda H^{(+)}\sum_{m=1}^{n-1}\frac{\sin[(4\pi/\lambda)x_0 m]}{\pi^3 m^2[2m-(2n-1)]}\,. \tag{9}$$

Here $H^{(\pm)} = \tan(\theta+\phi) \pm \tan(\theta - \phi)$. It can be checked that B_{2n-1} and D_{2n-1} are even and odd analytic functions of ϕ respectively.

Note that the diffusion field given by Eq. (7) corresponds to the solution of Eq. (1) subject to condition (2) where the front profile $\zeta(x)$ is given by Eqs. (5) and (6). We may mention, if need be, that once the geometry [i.e., $\zeta(x)$] is fixed the von Neuman theorem ensures the unicity of u.

Hitherto, we have made no assumption on ϕ, so that the present problem seems to be solved for each value of ϕ. An arbitrary value of ϕ, however, will not in general ensure that the two ends of a given lamella are at the same height, as they should be. We therefore have to impose this condition, which leads generically to isolated values of the tilt angle ϕ for a given value of the control parameters. In order to solve for the actual front profile, which is compatible with the calculated field, we should insert (7) into (3). The determination of the profile then amounts to solving a nonlinear differential equation [15] for $\zeta(x)$, subject to mechanical boundary conditions (4). This is of course a much simpler problem than the usual one where one has to solve the full integrodifferential equation [8]. We can in fact go much farther in the analytical analysis if we are only interested in determining the bifurcation equation. Indeed Eq. (3) can be integrated once over x [from 0 to x; where we make use of the mechanical equilibrium condition at $x = 0$ in Eq. (4)]. A second integration from $x = 0$ to $x = \lambda/2$, taking into account the second condition in Eq. (4), provides, after imposing that the two ends of a lamella be at the same height, the following condition

$$F(\phi,\mu) \equiv \int_0^{\lambda/2} \frac{f(x)}{(1-f^2)^{1/2}}dx = 0\,, \tag{10}$$

where $f = \sin(\theta - \phi) + \int_0^x (u - \Delta)/d_0 dx$ and μ stands for the material and control parameters (e.g., θ, λ). Equation (10) is a general expression for the tilt angle as a function of the other parameters. We can mention at this level that since Eq. (10) is formally an algebraic equation for the unknown ϕ, it can be satisfied, for a given μ, by a discrete set of ϕ values. Before exploiting it, let us first derive the equation that relates the undercooling to the other parameters. For that purpose we average (3) over a period (actually over a half period because the two solid phases are identical). The result is

$$\Delta=\frac{2d_0}{\lambda}\{\sin(\theta+\phi)+\sin(\theta-\phi)\}+2\sum_{n=1}^{\infty}\frac{B_{2n-1}}{\pi(2n-1)}. \quad (11)$$

Equations (10) and (11) are general expressions which determine ϕ and Δ as a function of the other parameters. It can be checked that $F(-\phi)=-F(\phi)$ and $\Delta(-\phi)=\Delta(\phi)$, as they should be. In order to investigate the possibility for the front to undergo a parity-breaking instability, and if so, to determine the nature (supercritical or subcritical) of the bifurcation, it suffices to expand F up to third order in ϕ. This can be done in general. However, in order to avoid long formulas in this paper, we confine ourselves to small contact angles, where the expressions turn out to be simple. The calculation involves evaluations of numerical series, some of them can be evaluated exactly, otherwise they are computed numerically. Equation (10) yields

$$\pi^4(\sigma_c-\sigma)\phi-\frac{\phi^3}{4\theta^2}+O(\phi^5)=0, \quad (12)$$

with

$$\sigma\equiv\frac{\tilde{d}_0\tilde{l}}{\tilde{\lambda}^2},\quad \sigma_c\simeq 0.2/\pi^4, \quad (13)$$

where the variables with tildes refer to the physical ones. Equation (12) shows that there exists a nontrivial solution given by

$$\phi=\pm 2\pi^2\theta(\sigma_c-\sigma)^{1/2}, \quad (14)$$

subject to the condition $\sigma<\sigma_c$. This means that the parity symmetry spontaneously breaks supercritically at $\sigma=\sigma_c$. Using the definition of σ (13) we can restate this result as follows: For a given growth velocity the initially symmetric solution loses its stability against parity-broken fluctuations for $\tilde{\lambda}>\tilde{\lambda}_c=(2\tilde{d}_0D/V\sigma_c)^{1/2}$. Now we expand Eq. (11) up to order ϕ^2 to obtain

$$\Delta=4\lambda\left[\sigma\theta+\frac{\alpha_1}{\pi^3}\right]-\frac{\alpha_2}{\pi^2}\phi^2, \quad (15)$$

where $\alpha_1\simeq 1.0$ and $\alpha_2\simeq 0.12$. For $\phi=0$ we recover the Jackson and Hunt [12] result. Equation (15) shows that parity-breaking results in a reduction of the front undercooling for a fixed velocity, or, equivalently, increases the velocity for a given undercooling. All the features following from Eqs. (12) and (15) are in qualitative agreement with numerical solutions of the full problem [8,13].

Further results follow. First it can be shown that $\tilde{\lambda}_c/\tilde{\lambda}_{\min}\sim 5/\sqrt{\theta}$, where $\tilde{\lambda}_{\min}$ is the (physical) wavelength which corresponds to the symmetric front with the minimum undercooling. This result means that the parity symmetry is lost at a wavelength larger than $\tilde{\lambda}_{\min}$, that is in a regime where the front dynamics is dominated by diffusion rather than by capillary forces. This is also a feature that agrees with previous numerical analysis [8,13]. Second, the wavelength $\tilde{\lambda}_d$ where the symmetric profile develops pockets (a wavelength which is close to the fold singularity [13]) is close to $\tilde{\lambda}_c$, and the corresponding value of σ, σ_d, scales as σ_c, that is to say they are both numbers, independent of θ, in contrast to $\sigma_{\min}$ which scales (for small θ) as $\sigma_{\min}\sim 1/\theta$. This result is comforting since $\tilde{\lambda}_{\min}$ results from a compromise between diffusion and capillarity [$\tilde{\lambda}\sim(\tilde{d}_0\theta\tilde{l})^{1/2}$; if θ is small the boundary layer of the capillary action is small; as a consequence diffusion will dominate at smaller wavelengths], while parity-breaking is a diffusion-driven instability, which should be present even for a zero-contact angle (note that parity-breaking occurs for interfaces with a single ordered phase where there is no notion of a pinning contact at all).

In summary we have presented a successful analytical treatment of parity-breaking transition in eutectic systems. Of course the restriction to the purely symmetric case is unrealistic. It is, however, of great importance to see that despite the oversimplification of the model, it still captures all the essential features. Naturally we are keeping in mind that in order to guide further experimental investigations it is necessary to extend our calculation to the general case. We intend to report on this work and on an extensive comparison with the full numerical calculations in the near future.

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[1] J.-M. Flesselles, A. J. Simon, and A. J. Libchaber, Adv. Phys. **40**, 1 (1991).
[2] A. J. Simon, J. Bechhoefer, and A. Libchaber, Phys. Rev. Lett. **61**, 2574 (1988).
[3] G. Faivre, S. de Cheveigné, C. Guthmann, and P. Kurowski, Europhys. Lett. **9**, 779 (1989).
[4] M. Rabaud, S. Michalland, and Y. Couder, Phys. Rev. Lett. **64**, 184 (1990).
[5] I. Mutabazi, J. Hegset, C. D. Andereck, and J. Wesfreid, Phys. Rev. Lett. **64**, 1729 (1990).
[6] P. Oswald, J. Phys. (France) II **1**, 571 (1991).
[7] P. Coullet, R. Goldstein, and G. H. Gunaratne, Phys. Rev. Lett. **63**, 1954 (1989).
[8] K. Kassner and C. Misbah, Phys. Rev. Lett. **65**, 1458 (1990); **66**, 522(E) (1991).
[9] H. Levine and W. J. Rappel, Phys. Rev. A **42**, 7475 (1990).
[10] B. A. Malomed and M. I. Tribelsky, Physica D **14**, 67 (1984).
[11] M. R. E. Proctor and C. A. Jones, J. Fluid Mech. **188**, 301 (1988).
[12] K. A. Jackson and J. D. Hunt, Trans. Metall. Soc. AIME **236**, 1121 (1966).
[13] K. Kassner and C. Misbah, Phys. Rev. A **44**, 6513 (1991).
[14] K. Kassner and C. Misbah, Phys. Rev. A **44**, 6533 (1991).
[15] C. Misbah and D. E. Temkin (unpublished).