# Asymmetric cell in directional solidification

Wouter-Jan Rappel
*Laboratoire de Physique Statistique, Ecole Normale Supérieure, 24 rue Lhomond, 75231 Paris CEDEX 05, France*
(Received 14 April 1993)

An asymmetric cell is obtained numerically in the symmetric model of directional solidification. It bifurcates off the symmetric cell branch and has a zero transversal velocity. The bifurcation point is characterized by a parity breaking and a period doubling. The bifurcation diagram around the codimension-two point found in previous work [Phys. Rev. A **45**, 846 (1991)] is completed.

PACS number(s): 68.45.−v, 47.20.Hw, 68.10.−m

Numerous works have recently been devoted to the understanding of spatial and spatiotemporal patterns. In particular, quasi-one-dimensional systems have attracted a lot of interest. One such system is the directional solidification of alloys or liquid crystals. Here, one pulls the material with a fixed velocity $v$ through an externally imposed temperature gradient. In particular the liquid-crystal system has been shown to exhibit a rich variety of patterns and transitions [1,2]. Most experimentally observed patterns have been reproduced in numerical investigations [3–6].

In this paper, we will present an asymmetric cellular interface obtained numerically in the symmetric model of directional solidification. Such a cell shape has recently been observed experimentally in the directional growth of succinonitrile [7] (where the one-sided model is applicable) and numerically in eutectic growth [8] as well as a dendrite in a channel [9], but not (yet?) in the directional solidification of liquid crystals.

The basic equations for the concentration field $c$ in the standard model of directional solidification have been presented in detail elsewhere [10]. For the symmetric model (equal diffusion constants in the liquid and the solid) these are (in the frame moving in the $y$ direction with the pulling velocity $v$, and lengths rescaled by the diffusion length $\frac{2D}{v}$, and time rescaled by $\frac{4D}{v^2}$)

$$\nabla^2 c + 2\frac{\partial c}{\partial y} = \partial_t c \ \text{(liquid, solid)}, \tag{1}$$

with boundary conditions at the liquid-solid interface:

$$(\hat{\mathbf{n}}\cdot\nabla c)_L - (\hat{\mathbf{n}}\cdot\nabla c)_S = -2(1-k)c_L\hat{\mathbf{n}}_y, \tag{2}$$

$$c_L = \frac{c_S}{k} = \frac{-y}{\xi} - \gamma\kappa. \tag{3}$$

Here $k$ is the partition coefficient, $\gamma$ is a rescaled capillary length, $\xi^{-1}$ is the rescaled temperature gradient, and $\kappa$ is the curvature. In the rescaled units the pulling velocity equals 2. Since we are interested in stationary interfaces we will set $\partial_t c = 0$.

We will now briefly summarize our numerical method. It consists of rewriting the diffusion equation and its boundary conditions into an integrodifferential equation. The resulting equation takes the form

$$c_L(s) = \int (1-k)c_L(s')\hat{\mathbf{n}}'\cdot\nabla' G(s,s')ds' + 1, \tag{4}$$

where the integration is over the unknown interface. $c_L$ is the value of the concentration field at the liquid side of the interface and can be expressed in terms of the interface and material parameters using (3). Here $G$, the Green's function, is a solution of

$$\nabla^2 G + 2\frac{\partial G}{\partial y} = -\delta(\mathbf{x}-\mathbf{x}') \tag{5}$$

and can be written as

$$G = \sum_n \frac{1}{2\pi} e^{-(y-y')} K_0[\sqrt{(x-x'+2pn)^2+(y-y')^2}], \tag{6}$$

where we have used the periodicity of the cells. $K_0$ is the modified Bessel function of order zero. The important advantage of this technique is that we only need to know the value of $c$ at the interface, which is given by the boundary conditions, and not everywhere in space.

After discretizing the interface, we have a set of nonlinear equations with as unknowns the interface itself and its position in the $y$ direction. This set of equations can be solved using a Newton's solver. Details of this technique can be found in [4,6].

In [6] the parameter space, $v$ versus $\lambda$, around the codimension-two point was investigated in great detail for the material parameters of the liquid-crystal experiments in [1,2]. In this paper we will use the same values for the material constants as in [6]. At the codimension-two point both the $q$ mode and the $2q$ mode become unstable, which allowed for the comparison between the numerically obtained results with an analytical approach, based on the coupling between the $q$ and $2q$ mode [11]. The agreement between the two approaches was found to be remarkable.

A variety of branches were found in [6] which we will briefly summarize here, using the notation of [6]. First of all, there is the pure $2q$ mode denoted by $S_2$. Then there are two mixed modes, i.e., both with a $q$ and $2q$ component present, which are denoted $S_\pm$. These branches all possess a reflection symmetry around the midline of the cell. In fact, using this symmetry it is sufficient to determine just half of a cell, thus taking the width of a computational box to be $\lambda/2$.

The linear stability of these symmetric cells was examined numerically. One of the mixed modes ($S_+$) was

 

found to lose its stability to a traveling wave (TW), a cell with a nonzero transversal velocity. This bifurcation constitutes a parity breaking, since the TW no longer has a reflection symmetry around the midpoint of the cell. By slightly adopting the numerical technique described above it is possible to obtain numerically the TW cells [5]. The main point is that if we go into a frame moving with velocity $v$ *and* a velocity in the transverse direction the mode is stationary again.

Since the parity is broken, we now have to calculate the whole cell shape and we have to take $\lambda$ as the width of our computational box. However, compared to the new cells to be discussed below, the wavelength of the cell is the same as the wavelength of the symmetric cell.

Of course, more complicated cellular solutions cannot be ruled out. One such possibility is an asymmetric cell with zero velocity in the transverse direction. There is evidence for the existence of such a cell in the directional solidification of succinonitrile [7]. Furthermore, such a cell has been found in the numerical investigation of eutectic growth [8] and of a dendrite in a channel [9]. It would therefore be logical to ask whether such a cell exists in the symmetric model of directional solidification.

In fact, the existence of a TW has helped us to find such a solution. This solution can be thought of as a series of domains of alternating left and right going traveling waves. The resulting cell will now have a wavelength *twice* that of a TW cell. Consequently, we have to change our Green's function which now becomes

$$G = \sum_n \frac{1}{2\pi} e^{-(y-y')} K_0[\sqrt{(x-x'+4pn)^2+(y-y')^2}] \tag{7}$$

since our periodicity has doubled. As an initial guess we have taken the interface shape of a TW and its reflection around $x = \lambda$. In this way we have indeed found a different cellular structure. Two typical asymmetric (AS) cells are displayed in Fig. 1, one for each part of the branch (see below), along with the TW for the same computational box. We see that the asymmetric cell has twice the period of the traveling-wave cell. It should be noted that the full cell is reflection symmetric and thus stationary.

The new branch, which we call AS for asymmetric, branches off the $S_+$ branch close to but not at the bifurcation point for the TW. Note that this bifurcation means a period doubling, since the effective wavelength becomes $2\lambda$, and a parity breaking. The natural bifurcation amplitude for this branch is the height difference $h$ of the cell at $x = \lambda/2$, which corresponds to the end point of our computational box, and $x = 0$. In Fig. 2 we have plotted $h$ as a function of the wave number. We see that the bifurcation from the $S_+$ branch is supercritical and that the branch has a fold and returns to a branch where we again have symmetric cells (the dashed curve denoted $S_{3/2}$ in Fig. 2). What actually happens at the end of the AS branch (the point where the solid curve in Fig. 2 joins the dashed curve) can be seen most easily from Fig. 1. As we increase $q$ the midpoint of the cell at $x = \lambda/2$ (in Fig. 1 this corresponds to the point $x = 91.25$ $\mu$m) continues to increase until it has reached the same value as

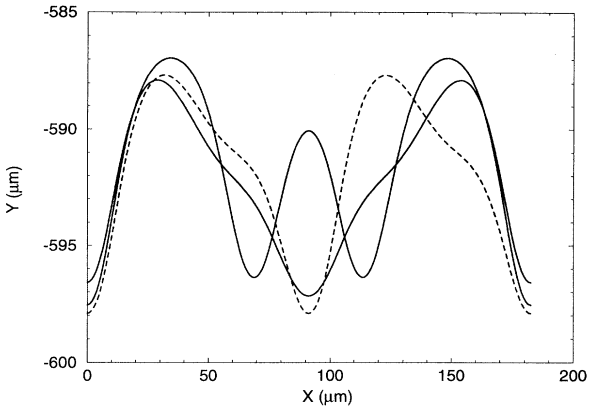


FIG. 1. Cell shapes for an AS cell for the two parts of the branch (solid line) and a TW cell for $v = 18.5$ $\mu$m/s. For both cells the computational box has the width 91.25 $\mu$m but for the TW branch one complete cell is in the box while for the AS branch only half of the cell is in the box.

the other local extremum of the cell. Since we then have one-and-a-half symmetric pure $2q$ cells in our computational box, we will denote this branch by $S_{3/2}$.

In Fig. 3 we have presented a full bifurcation diagram of all the cellular solutions found. For this diagram we have chosen to plot the total amplitude $A$ of the cell as a function of the wave number. The bifurcation diagram is quite complicated and merits an explanation. The thin lines are the branches already found in previous work. First of all, the pure $2q$ mode $S_2$ branches off the planar ($A = 0$) interface. The critical wave number for this bifurcation is determined by the linear stability analysis for the planar interface. The mixed modes $S_\pm$ both merge with the $S_2$ branch. Finally, the traveling-wave branch TW connects the $S_-$ and the $S_+$ branch.

The thick line in Fig. 3 is the newly found asymmetric cell branch (AS). It branches off the $S_+$ solution and the

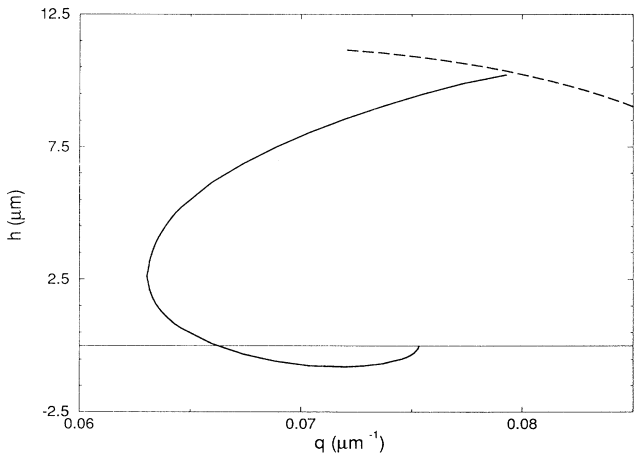


FIG. 2. The height difference $h$ between the cell at $x = \lambda/2$ and $x = 0$ as a function of the wave number $q$ for the AS cell ($v = 18.5$ $\mu$m/s). The thick solid line corresponds to the asymmetric cell shapes and the dashed line corresponds to the solution branch for the symmetric pure $2q$ mode.

cell undergoes at this point a parity breaking and a period doubling. The actual wave number of this branch should therefore be read as *half* the wave number on the axis of Fig. 3. Since the bifurcation is supercritical we expect the $S_+$ branch to lose its stability to the new AS branch. Since the $S_+$ branch is already unstable to the TW we expect the AS to be unstable to the TW as well. As we decrease the wave number, we encounter a fold in the AS branch. The branch turns around and merges with the symmetric $S_{3/2}$ mode branch. In our computational box (which is *twice* as big as the computational box used for the $S_2$ branch) we find $\frac{3}{2}$ of a pure $2q$ cell, so the dashed branch $S_{3/2}$ in Fig. 3 is nothing but the $S_2$ branch scaled by a factor of 4/3. Note that the self-crossing of the AS branch is an artifact of the way of presenting the bifurcation diagram. The amplitude $A$ of the cells at the crosspoint is the same on the two parts of the branches, but, as we can see from Fig. 2, the cells have a different $h$ and have thus a different shape.

We summarize this paper as follows: We have found numerically an asymmetric cell in the symmetric model for directional solidification. These cells bifurcate off the symmetric cell branch resulting in a bifurcation which breaks the parity and doubles the period.

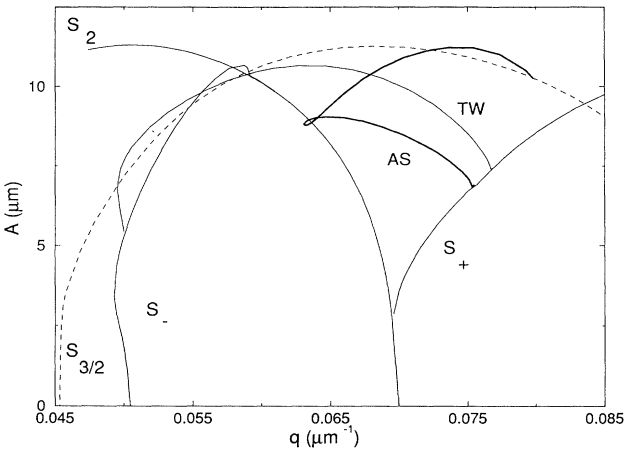


FIG. 3. The complete bifurcation diagram for $v = 18.5\mu$m/s. The thin lines correspond to previously obtained results and the dashed branch $S_{3/2}$ is the $S_2$ branch scaled by a factor of 4/3. The thick line is the new AS branch found in this paper.

I would like to thank C. Caroli and G. Faivre for suggesting this problem and for helpful discussions. I would also like to thank E. Brener for many fruitful discussions. This work was supported in part under an EC Human Capital and Mobility program. The Laboratoire de Physique Statistique, Ecole Normale Supérieure is "associé aux Universités Paris VI and VII."

[1] A. Simon, J. Bechhoefer, and A. Libchaber, Phys. Rev. Lett. **61**, 2574 (1988).
[2] J.-M. Flesselles, A. Simon, and A. Libchaber, Adv. Phys. **40**, 1 (1991).
[3] D.A. Kessler, J. Koplik, and H. Levine, Adv. Phys. **37**, 255 (1988).
[4] D.A. Kessler and H. Levine, Phys. Rev. A **39**, 3041 (1989).
[5] H. Levine and W.-J.Rappel, Phys. Rev. A **42**, 7475 (1991).
[6] W.-J. Rappel and H. Riecke, Phys. Rev. A **45**, 846 (1991).
[7] H. Jamgotchian, R. Trivedi, and B. Billia, Phys. Rev. E **47**, 4313 (1993).
[8] K. Kassner, A. Valance, C. Misbah, and D. Temkin, Phys. Rev. E **48**, 1091 (1993).
[9] E. Brener, H. Müller-Krumbhaar, Y. Saito, and D. Temkin, Phys. Rev. E **47**, 1151 (1993).
[10] For a review, see, e.g., J. Langer, in *Chance and Matter*, edited by J. Soulette, J. Vannimenus, and R. Stora (North-Holland, Amsterdam, 1987).
[11] C.A. Jones and M.R.E. Proctor, Phys. Lett. **121A**, 224 (1987); M.R.E. Proctor and C.A. Jones, J. Fluid Mech. **188**, 301 (1988).