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On the Long-Wave Nonlinear Instability in Directional Solidification

Jin-Yuan Hsieh and Chi-Chuan Hwang
Department of Mechanical Engineering, Chung Yuan Christian University, Chung-Li, Taiwan 320, R. O. C.

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In the present work, we undertake a fully nonlinear analysis of an evolution equation which describes the behavior of a solid-liquid interface in the directional solidification of a dilute binary mixture. The steady-state solutions, together with their stability analyses, and the locations of regular turning points in cases of subcritical bifurcation are evaluated. In the supercritical bifurcation, the stable finite amplitude to which a small perturbation to the planar interface will grow is also obtained. Some 2-D cellular patterns are also shown.

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I. INTRODUCTION

The general problem of solidification is quite difficult because it is a free boundary problem with nonlinear couplings among heat, mass, and momentum transfer. However, to obtain close-form solutions for qualitative analyses, the whole system may be reduced to simplified mathematical models [1]. The long-wave morphological instabilities in directional solidification have been studied extensively. One reason is that the simplified model can be totally governed by an evolution equation in the form of a partial differential equation.

Sivashinsky [2] derived a weakly nonlinear evolution equation applicable in the limit of an asymptotically small segregation coefficient. His evolution equation has subsequently been modified by including other considerations, e.g., see Novick-Cohen and Sivashinsky [3]. Weakly nonlinear analysis, including phase dispersion, was undertaken by Dee and Mathur [4], Wheeler [5] and Caroli, Caroli, and Roulet [6]. Brattkus and Davis [7] considering the absolute stability limit, have obtained a strongly nonlinear long-wave evolution equation. Riley and Davis [8] further examined the nonlinear development of long-wave disturbances when the segregation coefficient is small and the surface energy is large, and obtained another strongly nonlinear evolution equation. Recently, a strongly nonlinear evolution equation which contains the equation mentioned above [2,7,8] has been devived by Hsieh and Hwang [9,10].

In each limit, as the evolution equation is accomplished, weakly nonlinear instabilities can then be analyzed. The method follows the analyses of Wollkind and Segel [11] and of Sriranganathan, Wollkind, and Oulton [12]. The weakly nonlinear analysis of Sivashinsky's equation reveals that the corresponding bifurcation structure is subcritical. Supercritical bifurcation, instead, can be shown from the analysis of Brattkus and Davis's equation. These two limits could be connected when Riley and Davis's analysis showed that the bifurcation structure is supercritical (subcritical) if the morphological parameter is larger (smaller) than 9/4.

But, due to the simplicity of each equation mentioned above, there exists a good opportunity for carrying out in detail a fully nonlinear analysis. Langer [1] had suggested that the spatially periodic disturbance of the solid-liquid interface should be approximated by a Fourier series, then a set of coupled nonlinear equations can be solved numerically. Here we intend to follow Langer's suggestion to undertake a fully nonlinear analysis of Riley and Davis's equation. The reason is that this equation is simple, and from it both supercritical and subcritical bifurication can be observed. A two-dimensional disturbance to the planar state is considered, although Riley and Davis had pointed out that two dimensional solutions are unstable to a three-dimensional disturbance. We can do this only if three-dimensional disturbances can be suppressed. Nevertheless, the two-dimensional solutions will provide the basic states for the three-dimensional disturbances, and a three-dimensional analysis will be undertaken further in future.

Let us write down Riley and Davis's equation in advance:

$$\begin{aligned} h_t - M\Gamma\nabla^2 h_t + (M - 1 - kM\Gamma)\nabla^2 h + M\Gamma\nabla^4 h + kh \\ = \nabla\cdot(h\nabla h) - M\Gamma\nabla\cdot(\nabla^2 h\nabla h), \end{aligned} \tag{1}$$

where M, Γ, and k are three parameters during directional solidification, namely, the morphological parameter, the scaled surface energy, and the segregation coefficient, and h denotes the disturbance's amplitude. A linear stability analysis of (1) shows that the critical wave number of h is

$$a_c = \left(\frac{M^{\frac{1}{2}} - 1)^2}{M\Gamma}\right)^{\frac{1}{2}}, \tag{2}$$

and the critical value of the segregation coefficient is

$$k_c = \frac{(M^{\frac{1}{2}} - 1)^2}{M\Gamma}. \tag{3}$$

If k is chosen as the bifurcation parameter, a weakly nonlinear analysis predicts that the equilibrium amplitude is

$$h^e = \left(\frac{k_c - k}{\beta}\right)^{\frac{1}{2}}, \tag{4}$$

where $\beta = (2M^{\frac{1}{2}} - 3)/18M\Gamma$. In subcritical bifurcation (M < $9/4$), h^e is the unstable threshold amplitude, while in supercritical bifurcation, h^e becomes the stable amplitude to which a small disturbance will gradually grow.

A fully two-dimensional nonlinear analysis of Eq. (1), (i.e., h is assumed to be independent of y), begins with letting

$$h(x, t) = \sum_{m=0}^{N-1} b_m(t) \cos(max), \tag{5}$$

if $h(x,t)$ is spatially periodic. In expansion (5), N denotes the number of expansion terms and a is the fundamental wave number of $h(x,t)$. It should be mentioned here that, in the following calculations, $b_0(0) = 0$ is always chosen and, in addition, the fundamental wave number is set to be the critical one for the reduction of parameters. The Fourier series of $h(x,t)$ is then substituted into (1). Consequently, a set of N coupled nonlinear equations is obtained which are expressed by

$$\begin{aligned}\dot{b}_m = {} & \frac{(M-1-kM\Gamma)m^2a_c^2 - M\Gamma m^4 a_c^4 - k}{1+m^2a_c^2M\Gamma} b_m \\ & - \sum_{n=0}^{m} \frac{mna_c^2[1 + n(m-n)M\Gamma a_c^2]}{2(1+m^2a_c^2M\Gamma)} b_n b_{m-n} \\ & - \sum_{n=0}^{N-1-m} \frac{m^2a_c^2[1 - n(m+n)M\Gamma a_c^2]}{2(1+m^2a_c^2M\Gamma)} b_n b_{m+n}, \\ & \text{for } m = 0, 1, 2, \cdots, N-1,\end{aligned} \tag{6}$$

where the dot symbolizes d/dt. The equilibrium solutions for $\{b_m\}$ can be obtained numerically by taking vanishing $\{\dot{b}_m\}$ and choosing some specified N. Here we define the norm of the steady-state h by

$$||h||_N = \left(\sum_{m=0}^{N-1} b_m^2\right)^{\frac{1}{2}}. \tag{7}$$

Obviously, choices of N depend on the accuracy and on the three parameters, M, Γ, and k. Fig. 1 shows the $||h||_N$ *vs.* N results for some M's under $k = 2.0 \times 10^{-4}$ and $\Gamma = 100$. From these results, if accuracy $|||h||_{N+1} - ||h||_N| < 10^{-6}$ is required, we suggest that N = 36 should be chosen for $M = 1.4$, for example.

The bifurcation analysis of (6) begins with fixing the values of M and Γ and varying k *as* the bifurcation parameter. For convenience and for the reduction of parameters, $\Gamma = 100$ is chosen for all of the following calculations. If $M < 9/4$, i.e., if subcritical

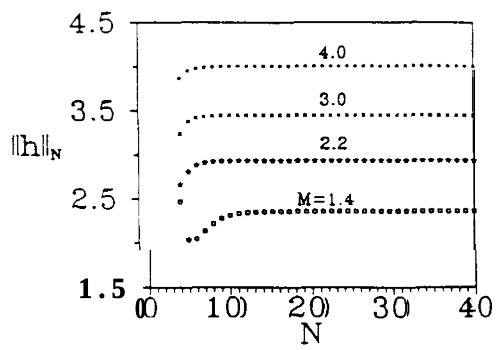

FIG. 1. Variations of $||h||_N$ with N. $k = 2.0 \times 10^{-4}$, I ' = 100.

bifurcation is considered, a hysteresis loop appears for each M. Such a hysteresis loop indicates that, besides k_c, there also exists another critical value of k which corresponds to the regular turning point on the loop and can not be evaluated from a weakly nonlinear analysis. Similarly, as far as supercritical bifurcation (M > $9/4$) is concerned, we will have a parabola-like curve for each M from the linear unstable region to $h = 0$.

The calculated steady solutions, however, need a further local stability analysis. This kind of analysis can easily be done by adding small perturbations to the steady-state solutions of $\{b_m\}$ and putting them into (6) for the linearization procedure. The procedure is typical so we have not included it but show the results directly.

An appropriate N has been chosen for each calculation. Fig. 2 shows the hysteresis loop in the $(k, ||h||_N)$ plane for $M = 1.4$. In this diagram, k_{cn} denotes the other critical value of k, which corresponds to the turning point. Local stability analysis will clearly determine whether the steady-state solution is stable. The solid lines in Fig. 2 represent the stable steady states, while the dashed lines are the unstable ones. When $k > k_{cn}$, there is only one solid line, $h = 0$, thus, any small pertubation will decay and the solid-liquid interface remains planar during solidification. This region, $k > k_{cn}$, then is the unconditional stable region. When $k_c < k < k_{cn}$, there are two solid lines and one dashed line, which is the threshold branch. This indicates that any initial disturbance with an amplitude larger (smaller) than the threshold amplitude will jump (decay) to a finite amplitude $(h = 0)$. Thus, the region $k_c < k < k_{cn}$, is the conditionally stable region for $h = 0$. Finally, as $k < k_c$, the line $h = 0$ is unstable and there is another solid line, the stable finite amplitude line. In this region, any small perturbation will grow to a finite amplitude and a cellular structure will be found. This region, $k < k_c$, then is the unstable region for $h = 0$. However, k_c, as well as k_{cn}, varies with M, if Γ is fixed. The variations of k_c and k_{cn} with M are

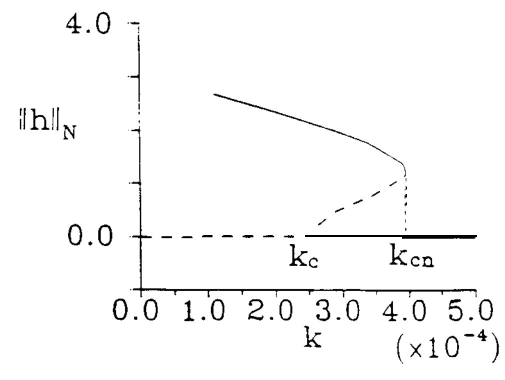

FIG. 2. Subcritical bifurcation diagram with M = 1.4, Γ = 100, and a = a,.

shown in Fig. 3. In this figure, the solid line represents the k_{cn} curve and the dashed line is the k_c curve. Thus, from this figure we can easily read that the domain above the solid line is the unconditionally stable domain, the domain between these lines represents the conditionally stable domain. The last domain is the unstable one.

In the case of supercritical bifurcation, the bifurcation diagram can be constructed by numerical calculation of (6). We show in Fig. 4 the supercritical bifurcation diagram for $M=$ 4.0. In the region where $k>k_c$, any small perturbation will decay and the solid-liquid interface remains planar during the solidification process, while in the region where $k<k_c$,

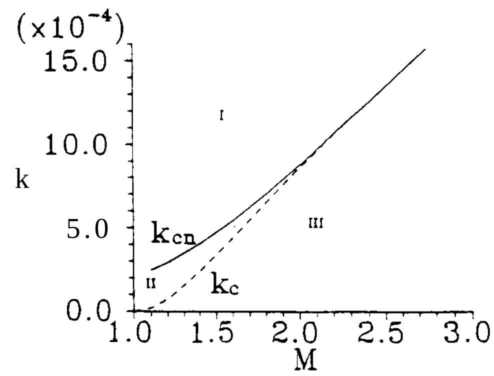

FIG. 3. Variations of k_c and k_{cn} with M where Γ = 100. As $M > 9/4$, supercritical bifurcation can be found and $k_{cn}=k$,. As $M < 9/4$ (subcritical bifurcation) the system is unconditionally stable if $k > k_{cn}$ (region I), is conditionally stable if $k_c<k<k_{cn}$ (region II), and is linear unstable if $k<k_c$ (region III).

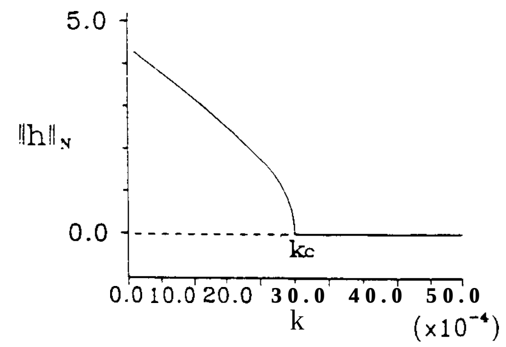

FIG. 4. Supercritical bifurcation diagram with $M = 4.0$, $\Gamma = 100$, and $a = a_c$.

an initial small disturbance will finally grow to a finite amplitude and a cellular structure will be found.

In the last figure, Fig. 5, we show some cellular patterns. These patterns are calculated with $k = 2.0 \times 10^{-4}$ and $M = 1.4$, 2.2, 3.0, and 4.0, respectively. It is shown that the amplitude increases as M increases. This phenomenon is rational because M in fact represents the constitutional supercooling during solidification and destablizes the solid-liquid interfacial disturbance.

Riley and Davis ' s twodimensional bifurcation analysis is only a weakly nonlinear one. Their results will lose accuracy when the bifurcation parmeter is far from its critical point. However, due to the simplicity of their equation, a fully nonlinear analysis is possible. In this work, we have expanded the interfacial amplitude into a Fourier series and then obtained a set of coupled nonlinear equations. A numerical method was applied to obtain

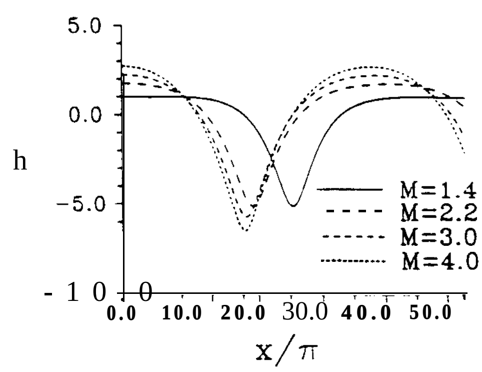

FIG. 5. Some cellular patterns of the stable steady-state h(z) with $k = 2.0 \times 10^{-4}$ and $\Gamma = 100$.

the steady-state solutions for these equations, and the strongly nonlinear behaviors of Riley and Davis's equation can then be studied. Information that can not be obtained from a weakly nonlinear analysis is clearly shown from the fully nonlinear analysis. This includes the critical value k_{cn}, a more accurate threshold amplitude, and the stable finite amplitude to which a disturbance should grow or jump to. Although Riley and Davis, after studing their equation, had declared that a two-dimensional disturbance is unstable to a three-dimensional disturbance, two-dimensional bifurcation analysis still plays an important role in directional solidification. In any case it is suggested that studies of the evolution of a three-dimensional disturbance use an approach similar to that applied in the present work.

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REFERENCES

[1] J. S. Langer, Rev. Mod. Phys. 52, 1 (1980).

[2] G. J. Sivashinsky, Phys. D8, 243 (1983).

[3] A. Novick-Cohen and G. J. Sivashinsky, Phys. D20, 237 (1986).

[4] G. Dee and R. Mathur, Phys. Rev. B27, 7073 (1983).

[5] A. A. Wheeler, SIAM J. Appl. Math. 35, 131 (1985).

[6] B. Caroli, C. Caroli, and B. Roulet, J. Crystal Growth 68, 677 (1984).

[7] K. Brattkus and S. H. Davis, Phys. Rev. B38, 11452 (1988).

[8] D. S. Riley and S. H. Davis, SIAM J. Appl. Math. 50, 420 (1990).

[9] J. Y. Hsieh and C. C. Hwang, J. Crystal Growth 126, 447 (1993).

[10] J. Y. Hsieh and C. C. Hwang, J. Appl. Phys. 76, 2765 (1994).

[11] D. J. Wollkind and L. A. Segel, Philos. Trans. Roy. Soc. London A, 268, 351 (1970).

[12] R. Sriranganathan, D. J. Wollkind, and D. B. Oulton, J. Crystal Growth 62, 265 (1983).