PHYSICAL REVIEW E VOLUME 53, NUMBER 6 JUNE 1996

Weakly nonlinear morphological instability of a cylindrical crystal growing from a pure undercooled melt

Partha P. Debroy
Department of Physics, The Cooper Union, New York, New York 10003

Robert F. Sekerka
Department of Physics and Department of Mathematics, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213
(Received 27 December 1995)

We develop a weakly nonlinear morphological stability analysis for an infinitely long right circular cylinder growing from its pure undercooled melt. For a cylinder perturbed by a specific planform consisting of sinusoids, we perform an expansion in the planform amplitude A to calculate the nonlinear critical radius (above which the chosen planform will be unstable for finite A), to the lowest order in A, by setting the normal velocity corresponding to the fundamental perturbing mode to zero. We study the nonlinear critical radius as a function of the amplitude to identify the various bifurcations, which for the chosen sinusoidal planforms are subcritical or supercritical (requiring an expansion to third order in A), since the shapes for positive and negative amplitudes are related by rotation and translation. We find that the bifurcations are mostly subcritical. For the special case of axially symmetric perturbations, we encounter a generalization of the Rayleigh varicosity instability. [S1063-651X(96)01706-4]

PACS number(s): 64.70.Dv, 81.10.Aj, 02.30.Mv

I. INTRODUCTION

We consider the free growth of a segment of an infinitely long right circular cylinder from its pure undercooled melt, subject to the quasistationary approximation. The process of crystallization from the melt is a first order phase transformation involving the release of latent heat which has to be conducted away both into the solid and the liquid as the process continues. For simplicity we assume isotropy of all crystalline properties including surface tension. The basic physics and the mathematical description of the problem is very similar to that for a spherical crystal, discussed in [1]. The details of the calculation will, however, be quite different since the cylinder, unlike the sphere, is not a shape of minimum area for a fixed volume. Indeed infinitesimal perturbations can lead to principle curvatures of opposite signs, resulting in an instability that is closely related to the Rayleigh varicosity instability.

II. THE UNDERLYING PHYSICS AND THE MODEL

We consider an infinite right circular cylinder growing by means of diffusive heat transfer (convection proscribed) from its pure undercooled melt. In the quasistationary approximation [2,3], the nondimensional governing equation and boundary conditions for thermal diffusion in the solid phase and the liquid phase may be written in the form

$$\nabla^2 U_L = 0, \tag{2.1}$$

$$\nabla^2 U_S = 0, \tag{2.2}$$

in the bulk solid and liquid, respectively. At the solid-liquid interface,

$$V_N = \boldsymbol{\nabla} U \cdot \hat{\mathbf{n}}, \tag{2.3}$$

$$U_L = U_S = 1 - K, \tag{2.4}$$

where $U = -U_L + \beta U_S$, β is the ratio of thermal conductivity in the solid to that in the liquid phase and

$$U_I \to 0 \quad \text{as} \quad \rho \to \infty. \tag{2.5}$$

The choice of dimensionless variables is similar to that of previous work [4,5] where lengths are scaled by the nucleation radius, $R^* = (T_M \gamma)/[L_0(T_M - T_\infty)]$, and time by $\tau = [(R^*)^2]/(\alpha_L S)$ where γ is the surface tension, T_M is the melting temperature, T_∞ is the far field temperature, L_0 is the latent heat, and α_L is the thermal diffusivity in the liquid phase. Note that R^* pertains to a cylindrical nucleus and is therefore a factor of two smaller than that for a sphere. The dimensionless curvature $K = R^*(1/R_1 + 1/R_2)$ where R_1 and R_2 are the dimensional principal radii of curvature. The dimensionless undercooling $S = \rho_L C_L (T_M - T_\infty)/L_0$ and we also use the dimensionless temperature fields $U_{S,L} = (T_{S,L} - T_\infty)/(T_M - T_\infty)$, where ρ_L and C_L are the density and specific heat of the liquid phase, respectively, and $T_{S,L}$ are the respective temperature fields in the solid and liquid.

III. PERTURBATION EXPANSION

We follow a procedure similar to that of [1] except in cylindrical coordinates (ρ,ϕ,z); we reproduce here only a brief outline to introduce our notation. At a particular instant of time, we study an interface of the form

$$\rho = g(z,\phi) \equiv \overline{\rho} + A \cos(kz)\cos(\nu\phi), \tag{3.1}$$

with ν being an integer, and to third order

$$\overline{\rho} = \rho_0 + \epsilon\rho_1 + \epsilon^2\rho_2 + \epsilon^3\rho_3, \tag{3.2}$$

$$A=\epsilon A_1+\epsilon^2 A_2+\epsilon^3 A_3 . \tag{3.3}$$

If we define

$$Z_i(z,\phi)=\rho_i+A_i\cos(\nu\phi)\cos(kz) \quad \text{for} \quad i=1,2,3, \tag{3.4}$$

then

$$g(z,\phi)=\rho_0+\epsilon Z_1(z,\phi)+\epsilon^2 Z_2(\phi,z)+\epsilon^3 Z_3(\phi,z). \tag{3.5}$$

Similarly we expand the temperature fields (which depend on ρ, ϕ, z)

$$U_L=U_{L0}+\epsilon U_{L1}+\epsilon^2 U_{L2}+\epsilon^3 U_{L3}, \tag{3.6}$$

$$U_S=U_{S0}+\epsilon U_{S1}+\epsilon^2 U_{S2}+\epsilon^3 U_{S3}, \tag{3.7}$$

the curvature,

$$K=K_0+\epsilon K_1+\epsilon^2 K_2+\epsilon^3 K_3 \tag{3.8}$$

and the normal growth speed

$$V_N=V_{N0}+\epsilon V_{N1}+\epsilon^2 V_{N2}+\epsilon^3 V_{N3}. \tag{3.9}$$

An explicit expression for the K_i in the curvature expansion is given in Appendix A. The differential equations and the interfacial boundary condition at each order of ϵ then appear as follows.

Order ϵ^0. The differential equations are

$$\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\,\frac{\partial U_{L0}}{\partial\rho}\right)=0, \tag{3.10}$$

$$\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\,\frac{\partial U_{S0}}{\partial\rho}\right)=0, \tag{3.11}$$

and the interface boundary conditions are

$$U_{L0}=1-\frac{1}{\rho_0}, \tag{3.12}$$

$$U_{S0}=1-\frac{1}{\rho_0}, \tag{3.13}$$

$$V_{N0}=\frac{\partial U_0}{\partial\rho}. \tag{3.14}$$

Order ϵ^n, $n=1,2,3$. The differential equations are

$$\nabla^2 U_{Ln}=0, \tag{3.15}$$

$$\nabla^2 U_{Sn}=0, \tag{3.16}$$

and the interface boundary conditions are

$$U_{Ln}+Z_n\,\frac{dU_{L0}}{d\rho}-\frac{Z_n}{\rho_0^2}-\frac{Z_{n\phi\phi}}{\rho_0^2}-Z_{nzz}=I_{Ln}, \tag{3.17}$$

$$U_{Sn}+Z_n\,\frac{dU_{S0}}{d\rho}-\frac{Z_n}{\rho_0^2}-\frac{Z_{n\phi\phi}}{\rho_0^2}-Z_{nzz}=I_{Sn}, \tag{3.18}$$

$$-V_{Nn}+\frac{\partial U_n}{\partial\rho}+Z_n\,\frac{\partial^2 U_0}{\partial\rho^2}=I_{Un}, \tag{3.19}$$

where the I's on the right hand sides denote inhomogeneous terms that are zero for $n=1$ but otherwise complicated functions of the solutions at a lower order. The relevant inhomogeneous terms are given in Appendix A. As discussed in [1], we can set $A_1=1$ and $A_2=A_3=0$ in Eq. (3.3), without any loss of generality. This amounts to renormalization and identification of $\epsilon=A$.

IV. PERTURBATION ALONG THE z AND ϕ DIRECTIONS

We consider a perturbation of the form

$$Z_i(\phi,z)=\rho_i+\epsilon\,\cos(kz)\cos(\nu\phi). \tag{4.1}$$

A. Zero order solution

We solve Eqs. (3.10) and (3.11) subject to the boundary conditions, Eqs. (3.12)–(3.14), to get

$$U_{S0}=\frac{\rho_0-1}{\rho_0}, \tag{4.2}$$

$$U_{L0}(\rho)=\frac{1-\rho_0}{\rho_0\ln(\rho_\infty/\rho_0)}\,\ln(\rho/\rho_\infty), \tag{4.3}$$

$$V_{N0}=\frac{\rho_0-1}{\rho_0^2\ln(\rho_\infty/\rho_0)}. \tag{4.4}$$

In cylindrical coordinates we need a finite cutoff ρ_∞ for the solution to remain finite. Following Coriell and Parker [6] we write $\rho_\infty=\rho_0/(\gamma\lambda)$, where $\ln\gamma^2=0.5572$ (Euler's constant) and for supercooling $S\ll 1$, λ is a solution of the equation

$$\lambda^2\ln(\gamma^2\lambda^2)+S=0.$$

With this choice of ρ_∞, the growth rate given by Eq. (4.4) is the same, to lowest order in S, as the growth rate calculated by using the fully time-dependent diffusion equation. For later use, we introduce the notation

$$\mathscr{A}_\lambda=\ln(\rho_\infty/\rho_0)=S/(2\lambda^2).$$

B. First order solution

The trial solutions to Eqs. (3.15) and (3.16), subject to the boundary conditions, Eqs. (3.17)–(3.19), can be written in the form

$$U_{L1}(\rho,z,\phi)=\alpha_{L1}^{(0)}\ln(\rho/\rho_\infty)+\alpha_{L1}^{(1)}[K_\nu(k\rho)-C_\nu I_\nu(k\rho)]\times\cos(kz)\cos(\nu\phi), \tag{4.5}$$

$$U_{S1}(\rho,z,\phi)=\alpha_{S1}^{(0)}+\alpha_{S1}^{(1)}I_\nu(k\rho)\cos(kz)\cos(\nu\phi), \tag{4.6}$$

$$V_{N1}(z,\phi)=V_{N1}^{(0)}+V_{N1}^{(1)}\cos(kz)\cos(\nu\phi), \tag{4.7}$$

$$Z_1(z,\phi)=\rho_1+\cos(kz)\cos(\nu\phi), \tag{4.8}$$

where K_ν and I_ν are the modified Bessel functions. The quantities, C_ν, $\alpha_{L1}^{(0)}$, $\alpha_{L1}^{(1)}$, $\alpha_{S1}^{(0)}$, $\alpha_{S1}^{(1)}$, $V_{N1}^{(0)}$, $V_{N1}^{(1)}$, and ρ_1, obtained by substituting the trial solutions into Eqs. (3.17)–(3.19), are given in Appendix B. We reproduce the expression for $V_{N1}^{(1)}$ for later reference

$$V_{N1}^{(1)}=\frac{1}{\rho_0^3\mathscr{A}_\lambda}\{(\rho_0-1)[\mathscr{J}_\nu(x)-1] -\mathscr{A}_\lambda(\nu^2+x^2-1)[\mathscr{J}_\nu(x)+\beta\mathscr{H}_\nu(x)]\}, \quad (4.9)$$

where

$$\mathscr{J}_\nu(x)=x\,\frac{-K_\nu'(x)+C_\nu I_\nu'(x)}{K_\nu(x)-C_\nu I_\nu(x)},$$

$$\mathscr{H}_\nu(x)=x\,\frac{I_\nu'(x)}{I_\nu(x)},$$

$$C_\nu=\frac{K_\nu(xe^{\mathscr{A}_\lambda})}{I_\nu(xe^{\mathscr{A}_\lambda})},$$

$$x=k\rho_0.$$

At the onset of instability, the normal velocity corresponding to the perturbing mode must vanish to all orders. For the first order, this amounts to setting

$$V_{N1}^{(1)}=0$$

in Eq. (4.9). For $\mathscr{J}_\nu(x)\neq 1$ this gives the critical radius for instability in terms of k, ν, and β. It is easier to first express ρ_0 in terms of x, ν, and β as

$$\rho_0(x,\nu,\beta)=1+\mathscr{A}_\lambda\left[\frac{(\nu^2+x^2-1)[\mathscr{J}_\nu(x)+\beta\mathscr{H}_\nu(x)]}{\mathscr{J}_\nu(x)-1}\right] \quad (4.10)$$

and later to divide x by ρ_0 to obtain the corresponding value of k. The critical radius as a function of k for various ν is shown in Fig. 1.

To see how a certain perturbing mode grows, we imagine a line k=const in either of the plots in Fig. 1. If the constant is zero, the vertical line cuts each curve at a single point, implying that the system starts out stable and then goes unstable with respect to a given mode as ρ increases past the corresponding critical value. For any other value of the constant, up to some maximum corresponding to the knee of the curve, the vertical line can intersect each curve at two points, which means that the system is linearly stable for ρ less than that given by the lower branch of the curve, goes unstable above it but stabilizes again for ρ greater than the upper branch of the curve. Thus the region enclosed by each curve and the ρ_0 axis constitutes an unstable zone, while the rest of the plane is a stable zone for the corresponding perturbing mode. We postpone the discussion of the case ν=0 (which corresponds to a perturbation in the z direction only) to the next subsection because it has some unique features.

Another case that needs special mention is perturbation with k=0 and ν=1. This corresponds to a perturbation by cos ϕ along the ϕ direction and no perturbation along the axis of the cylinder. In that limit

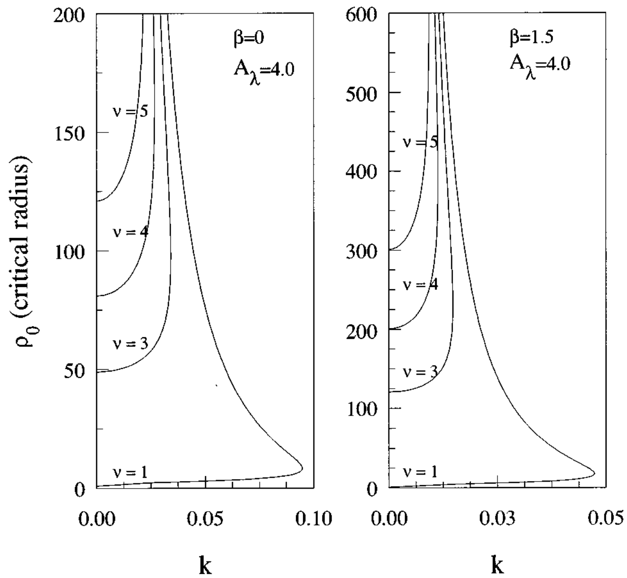

FIG. 1. The critical radius as a function of the z-direction wave number k for various ν. The thermal conductivity ratio β is 0.0 for the left plot and 1.5 for the right. The supersaturation is such that $\mathscr{A}_\lambda$=4.0. For each of the plots, ν=1,3,4,5 from bottom to top. As we approach a given critical curve from below, the system is linearly stable for any radius less than that given by the lower branch of the curve, goes unstable above it, but restabilizes as we cross the upper branch of the curve.

$$\mathscr{H}_1(0)=1,$$

$$\mathscr{J}_1(0)=\frac{\rho_\infty^2+\rho_0^2}{\rho_\infty^2-\rho_0^2}.$$

If $\mathscr{J}_1\to 1$, the denominator of Eq. (4.10) tends to zero and Eq. (4.9) becomes

$$V_{N1}^{(1)}=\frac{1}{\rho_0^3\mathscr{A}_\lambda}\,\frac{2\rho_0^2(\rho_0-1)}{\rho_\infty^2-\rho_0^2}. \quad (4.11)$$

For $\rho_\infty \gg \rho_0$, we see that $V_{N1}^{(1)}\to 0$, so this mode is nearly neutrally stable. This arises because perturbation by cos ϕ represents a translation to first order in ϵ; however, the finite cutoff ρ_∞ used to avoid the singularity at infinity to solutions of the Laplace equation, spoils this precise symmetry. The corresponding case for the sphere was treated in [1] in which case no finite cutoff was necessary.

Perturbation in the z direction only

The results for a z perturbation alone (axial symmetry) can be obtained by setting ν=0 in the general expressions. Equation (4.10) yields

$$\rho_0(x,\beta)=1+\mathscr{A}_\lambda\left[\frac{(x^2-1)[\mathscr{J}_0(x)+\beta\mathscr{H}_0(x)]}{\mathscr{J}_0(x)-1}\right]. \quad (4.12)$$

A plot of ρ_0 as a function of k (the z-direction wave number) is shown in Fig. 2. The broken line represents the hyperbola $k\rho_0$=1. The normal velocity as a function of the radius ρ_0 for two different values of k is shown in Fig. 3. To understand

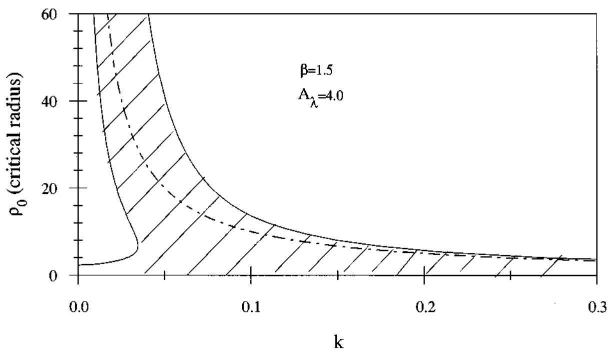

FIG. 2. Critical radius as a function of the wave number k for the perturbation in the z direction only ($\nu=0$). The unstable region has been hatched. The long wavelength instability due to the Rayleigh condition, represented by the region $k\rho_0<1$ (the broken line represents $k\rho_0=1$) is a characteristic of this class of perturbations.

how a perturbation grows, we imagine a vertical line $k=0.02$ in Fig. 2 in conjunction with the first plot in Fig. 3. For small ρ_0, the normal velocity is positive, so the system is unstable. As ρ_0 increases past the lower branch of the curve, the normal velocity changes sign and the system becomes stable. However, further increase of ρ_0 beyond the upper branch of the curve renders the system unstable again. For $k=0.2$, on the other hand, we see that the system is unstable at first, but then becomes stable as ρ_0 crosses the critical value. Therefore the curves in Fig. 2 divide the plane into a stable (extreme left), an unstable (middle), and a stable (right) region, respectively. From considerations of surface free energy alone, the region $k\rho_0<1$ bounded by the dotted line, would have been unstable while the region to the right would have been stable. This is the well known Rayleigh varicosity instability, and is a result of the fact that sinusoidal perturbations of wavelengths greater than $2\pi\rho_0$ lower the surface to volume ratio of a cylinder. Thus the class of perturbations with $\nu=0$ has a long wavelength instability not seen in the general case when $\nu\neq 0$. Similar conclusions were discussed by Coriell and Parker [6] in terms of the variables ρ_0, x, in which case the Rayleigh varicosity instability would correspond to a vertical line $x=1$. McFadden, Coriell, and Murray [7] considered the related case of a cylinder that is not growing, made possible by imposing a fixed temperature at an inner cutoff radius as well as an outer cutoff radius. They found that a positive temperature gradient in the liquid tends to oppose or even suppress the Rayleigh instability, whereas our negative temperature gradient in the liquid, needed for the unperturbed cylinder to grow, enhances the Rayleigh instability.

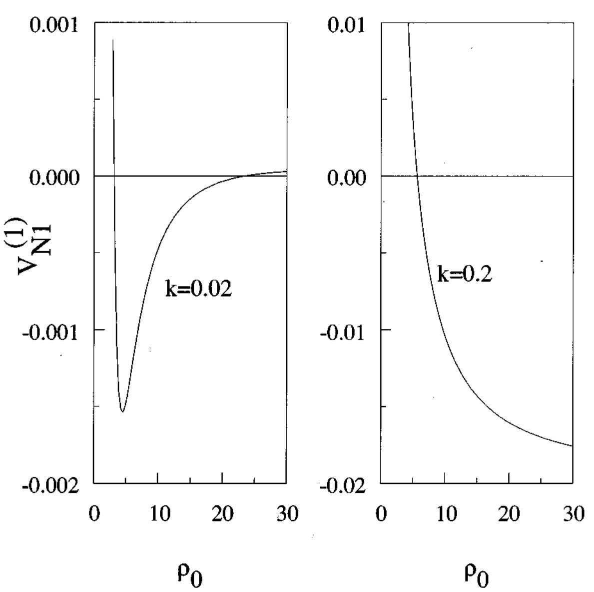

FIG. 3. The normal velocity component proportional to the perturbing mode at first order. The left plot is for $k=0.02$ and the right plot is for $k=0.2$.

C. Second order solution

The inhomogeneous terms in Eqs. (3.17)–(3.19) can be expressed formally as

$$I=I^{(A)}+\rho_1 I^{(B)}\cos(kz)\cos(\nu\phi)+I^{(C)}\cos(2kz)\cos(2\nu\phi)+I^{(D)}\cos(2kz)+I^{(E)}\cos(2\nu\phi),$$

where all the coefficients are known and appear in Appendix B. This suggests the following form for the second order trial solutions:

$$U_{L2}(\rho,z,\phi)=\alpha_{L2}^{(0)}\ln(\rho/\rho_\infty)+\alpha_{L2}^{(1)}K_\nu(k\rho)\cos(kz)\cos(\nu\phi)+\alpha_{L2}^{(2)}[K_{2\nu}(2k\rho)-C_{2\nu}I_{2\nu}(2k\rho)]\cos(2kz)\times\cos(2\nu\phi)+\alpha_{L2}^{(3)}\times[K_0(2k\rho)-C_0I_0(2k\rho)]\cos(2kz)+\alpha_{L2}^{(4)}\times\left[\left(\frac{\rho_\infty}{\rho}\right)^{2\nu}-\left(\frac{\rho}{\rho_\infty}\right)^{2\nu}\right]\cos(2\nu\phi), \quad (4.13)$$

$$U_{S2}(\rho,z,\phi)=\alpha_{S2}^{(0)}+\alpha_{S2}^{(1)}I_\nu(k\rho)\cos(kz)\cos(\nu\phi)+\alpha_{S2}^{(2)}I_{2\nu}(2k\rho)\cos(2kz)\cos(2\nu\phi)+\alpha_{S2}^{(3)}I_0(2k\rho)\cos(2kz)+\alpha_{S2}^{(4)}\rho^{2\nu}\cos(2\nu\phi), \quad (4.14)$$

$$V_{N2}(z,\phi)=V_{N2}^{(0)}+V_{N2}^{(1)}\cos(kz)\cos(\nu\phi)+V_{N2}^{(2)}\cos(2kz)\cos(2\nu\phi)+V_{N3}^{(3)}\cos(2kz)+V_{N2}^{(4)}\cos(2\nu\phi), \quad (4.15)$$

$$Z_2=\rho_2, \quad (4.16)$$

where

$$C_{2\nu}=\frac{K_{2\nu}(2k\rho_\infty)}{I_{2\nu}(2k\rho_\infty)},$$

$$C_0=\frac{K_0(2\kappa\rho_\infty)}{I_0(2k\rho_\infty)}.$$

The quantities $\alpha_{L2}^i,\alpha_{S2}^i,V_{N2}^i$ for $i=0,1,2,3,4$, found by substituting the trial solutions into the interface boundary conditions, Eqs. (3.17)–(3.19), are given in Appendix B. The marginal stability condition at second order requires $V_{N2}^{(1)}$ to vanish, providing us with

$$\rho_1\left[I_{U2}^{(B)}+I_{L2}^{(B)}\frac{x}{\rho_0}\frac{K_\nu'(x)}{K_\nu(x)}-I_{S2}^{(B)}\beta\frac{x}{\rho_0}\frac{I_\nu'(x)}{I_\nu(x)}\right]=0. \quad (4.17)$$

Since the terms within the square brackets in Eq. (4.17) can be shown to be nonzero, we must have

$$\rho_1=0, \quad (4.18)$$

forcing

$$\alpha_{L2}^{(1)}=\alpha_{S2}^{(1)}=0.$$

This leaves ρ_2 as the only free parameter, which will be determined at the next order.

D. Third order solution

The third order inhomogeneous terms in Eqs. (3.17)–(3.19) can be expressed as

$$I_{L3}=I_{L3}^{(B)}\cos(kz)\cos(\nu\phi)+\cdots,$$

$$I_{S3}=I_{S3}^{(B)}\cos(kz)\cos(\nu\phi)+\cdots,$$

$$I_{U3}=I_{U3}^{(B)}\cos(kz)\cos(\nu\phi)+\cdots,$$

where we show only the terms necessary to determine ρ_2. Therefore the trial solutions to Eqs. (3.15) and (3.16) will look like

$$U_{L3}(\rho,z,\phi)=\alpha_{L3}^{(1)}[K_\nu(k\rho)-C_\nu I_\nu(k\rho)]\cos(kz)\cos(\nu\phi)+\cdots, \quad (4.19)$$

$$U_{S3}(\rho,z,\phi)=\alpha_{S3}^{(1)}I_\nu(k\rho)\cos(kz)\cos(\nu\phi)+\cdots, \quad (4.20)$$

$$V_{N3}(z,\phi)=V_{N3}^{(1)}\cos(kz)\cos(\nu\phi)+\cdots, \quad (4.21)$$

$$Z_3=\rho_3. \quad (4.22)$$

These terms will suffice in finding an expression for ρ_2, which had remained undetermined at the end of the second order. We substitute the trial solutions in the boundary condition Eqs. (3.17)–(3.19) to find the quantities $\alpha_{L3}^{(1)}$, $\alpha_{S3}^{(1)}$, and $V_{N3}^{(1)}$, which appear in Appendix B. In particular, the normal velocity coefficient proportional to the fundamental perturbing mode is

$$V_{N3}^{(1)}=-I_{L3}^{(B)}\frac{x}{\rho_0}\frac{K_\nu'(x)-C_\nu I_\nu'(x)}{K_\nu(x)-C_\nu I_\nu(x)}+I_{S3}^{(B)}\beta\frac{x}{\rho_0}\frac{I_\nu'(x)}{I_\nu(x)}-I_{U3}^{(B)}. \quad (4.23)$$

At the onset of instability we must have

$$V_{N3}^{(1)}=0$$

in Eq. (4.23), giving us an expression for ρ_2 as a function of x, β, ν. The explicit expression appears in Appendix B and a

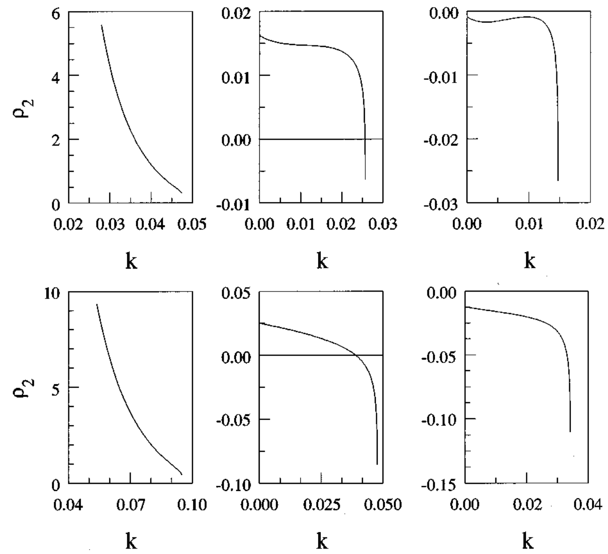

FIG. 4. ρ_2 as a function of the z-direction wave number k for various ν values. The thermal conductivity ratio β is 0.0 in the lower plots and 1.5 in the upper plots. The supersaturation is such that $\mathscr{A}_\lambda=4.0$. $\nu=1,2,3$ from left to right. For all plots the system is linearly stable beyond the maximum value of k as given by Fig. 1 for each of the curves. Positive ρ_2 implies supercritical bifurcation while negative values imply subcritical bifurcation.

plot of ρ_2 as a function of k for various values of ν and β is shown in Fig. 4. If we go back to Eq. (3.2) we have, since $\rho_1=0$,

$$\rho(x,\nu,\beta)=\rho_0(x,\nu,\beta)+A^2\rho_2(x,\nu,\beta). \quad (4.24)$$

The plot of ρ as a function of A will be a parabola and the bifurcation will be subcritical if ρ_2 is negative and supercritical otherwise. Thus, in contrast to the case of the sphere [1], the bifurcations are never transcritical for these planforms on a cylinder. This happens because, for the chosen planform, the positive and the negative amplitude perturbations are related by rotation (for the ϕ axis) and translation (for the z axis) and do not constitute any distinct physical states. Consequently the critical ρ is independent of the sign of the amplitude A.

V. CONCLUSIONS

An expansion in the perturbation amplitude A is performed and the critical radius to the lowest order in A is found by setting the normal velocity corresponding to the fundamental perturbing mode to zero. Depending on the symmetry of the perturbing mode we found the following results.

(1) $\nu=0$ and arbitrary k: The critical radius for this axially symmetric perturbation is given by Eq. (4.12). Perturbations of this form are subject to a long wavelength instability related to the Rayleigh varicosity instability, which occurs because sinusoidal perturbations with a wavelength greater than the circumference of a cylinder can lower its surface to the volume ratio.

(2) $\nu=1$ and $k=0$: This case corresponds to a perturbation by $\cos(\phi)$ along the ϕ direction and no perturbation along the axis of the cylinder. To the first order in the per-

turbation amplitude, this amounts to a translation of the cylinder without any shape change. If such a cylinder were in an infinite medium, this perturbation would be neutrally stable. For the case of a finite cutoff radius, [see Eq. (4.11)] the normal growth speed of a perturbation tends to zero for $\rho_\infty \gg \rho_0$.

(3) $\nu \neq 0$ and arbitrary k: The critical radius for a perturbation of finite amplitude A is found by carrying the perturbation expansion in A to the third order and is given by Eq. (4.24). The bifurcation is supercritical if $\rho_2(x,\nu,\beta)>0$, and subcritical if $\rho_2(x,\nu,\beta)<0$. In this case, the shapes generated by the positive and negative amplitude are related by rotation (in the ϕ direction) and translation (in the z direction). The governing equations are therefore independent of the sign of A.

In three dimensions, the nature of the bifurcations depend on the symmetry of the planform under consideration. For the perturbed sphere [1], we found that such bifurcations could be transcritical, subcritical, or supercritical depending on the particular spherical harmonic under consideration. Moreover, capillarity was always a stabilizing force. This behavior for a sphere arose because an unperturbed sphere has two positive and equal principal radii of curvature. Any perturbations of such a body at a fixed volume tend to increase its surface area, and all directions along the surface of the unperturbed sphere are equivalent. For the unperturbed circular cylinder, on the other hand, one principal radius of curvature is positive and the other is infinite. Thus small perturbations along the z direction can lead to regions of negative curvature for sufficiently long wavelengths, the origin of the Rayleigh instability. Moreover, directions along the surface of the perturbed cylinder are not equivalent and perturbations of the form $A\cos(\nu\phi)$ and $A\cos(kz)$ display symmetries of the form $A \to -A$ when, respectively, $\nu \to \nu + 2\pi$ and $z \to z + 2\pi/k$. Therefore there are no transcritical bifurcations for the cylinder.

The origin of another feature of our analysis of the cylinder, namely, the upper branches in Fig. 1 that correspond to restabilization, is worth mentioning. These branches arise because we chose to discuss the problem at fixed ν and fixed k. By fixing ν, one fixes the number of nodes in the ϕ direction, so the wavelengths associated with ϕ perturbations are ρ_0/ν, i.e., they scale with ρ_0. But the wavelengths associated with the z perturbations are $2\pi/k$, independent of ρ_0. Therefore, as ρ_0 increases and the gradient effect that gives rise to instability is weakened, the capillary stabilization due to z perturbations with a *fixed* wavelength becomes stabilizing. This can also be seen by writing Eq. (4.10) in the form

$$\rho_0 = 1 + C(x)(\nu^2 + k^2\rho_0^2 - 1).$$

To the degree that $C(x)$ is not strongly dependent on x, we see approximately how the $k^2\rho_0^2$ term on the right hand side becomes important as ρ_0 increases. This restabilizing behavior would be alleviated if one discussed the problem in terms of fixed ν and fixed x, so that both wavelengths would scale with ρ_0. This was done by Coriell and Parker [6] whose variable k_z is the same as our x, and makes perfectly good sense for a linear stability analysis. On the other hand, in an initial value problem for a perturbation corresponding to given ν and k, one would not be able to hold x constant as ρ_0 increases, which makes x an inconvenient parameter for a nonlinear analysis. As mentioned in our previous paper in connection with the sphere, these weakly nonlinear results constitute a nontrivial test for the development of numerical algorithms for three dimensional problems. It would also be interesting to explore the effect of anisotropies in surface free energy, especially those that would couple with the ν perturbations in a manner that might shed light on the crystallographically directed sidebranches of dendrites.

ACKNOWLEDGMENTS

This work was supported by the Division of Materials Research of the NSF under Grant Nos. DMR89-12752 and DMR 92-11276. The authors would also like to thank S. R. Coriell and G. B. McFadden for valuable discussions.

APPENDIX A

The expression for the curvature expansion terms in Eq. (3.8) are as follows:

$$K_0 = \frac{1}{\rho_0},$$

$$K_1 = -\frac{Z_1}{\rho_0^2} - \frac{Z_{1\phi\phi}}{\rho_0^2} - Z_{1zz},$$

$$K_2 = -\frac{Z_2}{\rho_0^2} - \frac{Z_{2\phi\phi}}{\rho_0^2} - Z_{2zz} + \frac{Z_1^2}{\rho_0^3} + \frac{2Z_1Z_{1\phi\phi}}{\rho_0^3} + \frac{Z_{1\phi}^2}{2\rho_0^3} - \frac{Z_{1z}^2}{2\rho_0},$$

$$K_3 = -\frac{Z_3}{\rho_0^2} - \frac{Z_{3\phi\phi}}{\rho_0^2} - Z_{3zz} + \frac{2Z_1Z_2}{\rho_0^3} + \frac{2Z_1Z_{2\phi\phi}}{\rho_0^3} + \frac{2Z_2Z_{1\phi\phi}}{\rho_0^3} + \frac{Z_{1\phi}Z_{2\phi}}{\rho_0^3} - \frac{Z_{1z}Z_{2z}}{\rho_0} - \frac{Z_1^3}{\rho_0^4} - 3\frac{Z_1^2Z_{1\phi\phi}}{\rho_0^4} + \frac{Z_1Z_{1z}^2}{2\rho_0^2} + \frac{Z_{1\phi\phi}Z_{1z}^2}{2\rho_0^2} + \frac{Z_{1zz}Z_{1\phi}^2}{2\rho_0^2} - \frac{3}{2}\frac{Z_1Z_{1\phi}^2}{\rho_0^4} + \frac{3}{2}Z_{1zz}Z_{1z}^2 + 2\frac{Z_{1\phi}Z_{1z}Z_{1\phi z}}{\rho_0^2} + \frac{3}{2}\frac{Z_{1\phi}^2Z_{1\phi\phi}}{\rho_0^4}.$$

In the above expressions and subsequent text, the subscripts ϕ and z denote the respective partial derivatives.

The inhomogeneous terms for Sec. III are:

$$I_{L2} = -\frac{1}{2}\left(\frac{\partial^2 U_{L0}}{\partial\rho^2}\right)Z_1^2 - \left(\frac{\partial U_{L1}}{\partial\rho}\right)Z_1 - \frac{Z_1^2}{\rho_0^3} - \frac{2Z_1Z_{1\phi\phi}}{\rho_0^3} - \frac{Z_{1\phi}^2}{2\rho_0^3} + \frac{Z_{1z}^2}{2\rho_0},$$

$$I_{S2} = -\frac{1}{2}\left(\frac{\partial !^2 U_{S0}}{\partial\rho^2}\right)Z_1^2 - \left(\frac{\partial U_{S1}}{\partial\rho}\right)Z_1 - \frac{Z_1^2}{\rho_0^3} - \frac{2Z_1Z_{1\phi\phi}}{\rho_0^3} - \frac{Z_{1\phi}^2}{2\rho_0^3} + \frac{Z_{1z}^2}{2\rho_0},$$

$$I_{U2}=-\frac{1}{2}\left(\frac{\partial^3 U_0}{\partial\rho^3}\right)Z_1^2-\left(\frac{\partial^2 U_1}{\partial\rho^2}\right)Z_1+\frac{1}{\rho_0^2}\,Z_{1\phi}\,\frac{\partial U_1}{\partial\phi}+Z_{1z}\,\frac{\partial U_1}{\partial z}+\frac{1}{2}\,\frac{\partial U_0}{\partial\rho}\left[\frac{1}{\rho_0^2}\,Z_{1\phi}^2+Z_{1z}^2\right]$$

and

$$\begin{aligned}I_{L3}=&-\frac{Z_1^3}{6}\,\frac{\partial^3 U_{L0}}{\partial\rho^3}-Z_1Z_2\,\frac{\partial^2 U_{L0}}{\partial\rho^2}-Z_1\,\frac{\partial U_{L2}}{\partial\rho}-Z_2\,\frac{\partial U_{L1}}{\partial\rho}\\&-\frac{Z_1^2}{2}\,\frac{\partial^2 U_{L1}}{\partial\rho^2}-\frac{2Z_1Z_2}{\rho_0^3}-\frac{2Z_1Z_{2\phi\phi}}{\rho_0^3}-\frac{2Z_2Z_{1\phi\phi}}{\rho_0^3}\\&-\frac{Z_{1\phi}Z_{2\phi}}{\rho_0^3}+\frac{Z_{1z}Z_{2z}}{\rho_0}+\frac{Z_1^3}{\rho_0^4}+\frac{3Z_1^2Z_{1\phi\phi}}{\rho_0^4}-\frac{Z_1Z_{2z}^2}{2\rho_0^2}\\&-\frac{Z_{1\phi\phi}Z_{1z}^2}{2\rho_0^2}-\frac{Z_{1zz}Z_{1\phi}^2}{2\rho_0^2}+\frac{3Z_1Z_{1\phi}^2}{2\rho_0^4}-\frac{3Z_{1zz}Z_{1z}^2}{2}\\&-\frac{2}{\rho_0^2}\,Z_{1\phi}Z_{1z}Z_{1\phi z}-\frac{3}{2\rho_0^4}\,Z_{1\phi}^2Z_{1\phi\phi},\end{aligned}$$

$$\begin{aligned}I_{S3}=&-\frac{Z_1^3}{6}\,\frac{\partial^3 U_{S0}}{\partial\rho^3}-Z_1Z_2\,\frac{\partial^2 U_{S0}}{\partial\rho^2}-Z_1\,\frac{\partial U_{S2}}{\partial\rho}-Z_2\,\frac{\partial U_{S1}}{\partial\rho}\\&-\frac{Z_1^2}{2}\,\frac{\partial^2 U_{S1}}{\partial\rho^2}-\frac{2Z_1Z_2}{\rho_0^3}-\frac{2Z_1Z_{2\phi\phi}}{\rho_0^3}-\frac{2Z_2Z_{1\phi\phi}}{\rho_0^3}\\&-\frac{Z_{1\phi}Z_{2\phi}}{\rho_0^3}+\frac{Z_{1z}Z_{2z}}{\rho_0}+\frac{Z_1^3}{\rho_0^4}+\frac{3Z_1^2Z_{1\phi\phi}}{\rho_0^4}-\frac{Z_1Z_{2z}^2}{2\rho_0^2}\\&-\frac{Z_{1\phi\phi}Z_{1z}^2}{2\rho_0^2}-\frac{Z_{1zz}Z_{1\phi}^2}{2\rho_0^2}+\frac{3Z_1Z_{1\phi}^2}{2\rho_0^4}-\frac{3Z_{1zz}Z_{1z}^2}{2}\\&-\frac{2}{\rho_0^2}\,Z_{1\phi}Z_{1z}Z_{1\phi z}-\frac{3}{2\rho_0^4}\,Z_{1\phi}^2Z_{1\phi\phi},\end{aligned}$$

$$\begin{aligned}I_{U3}=&-Z_1Z_2\,\frac{\partial^3 U_0}{\partial\rho^3}-\frac{1}{6}\,Z_1^3\,\frac{\partial^4 U_0}{\partial\rho^4}-Z_2\,\frac{\partial^2 U_1}{\partial\rho^2}-\frac{1}{2}\,Z_1^2\,\frac{\partial^3 U_1}{\partial\rho^3}\\&-Z_1\,\frac{\partial^2 U_2}{\partial\rho^2}+\frac{Z_1}{\rho_0^2}\,Z_{1\phi}\,\frac{\partial^2 U_1}{\partial\rho\partial\phi}+Z_1Z_{1z}\,\frac{\partial^2 U_1}{\partial\rho\partial z}\\&+\frac{Z_1}{2}\,\frac{\partial^2 U_0}{\partial\rho^2}\left[\frac{1}{\rho_0^2}\,Z_{1\phi}^2+Z_{1z}^2\right]-\frac{2Z_1}{\rho_0^3}\,Z_{1\phi}\,\frac{\partial U_1}{\partial\phi}\\&+\frac{1}{\rho_0^2}\,Z_{1\phi}\,\frac{\partial U_2}{\partial\phi}+Z_{1z}\,\frac{\partial U_2}{\partial z}+\frac{1}{\rho_0^2}\,Z_{2z}\,\frac{\partial U_1}{\partial\phi}+Z_{2z}\,\frac{\partial U_1}{\partial z}\\&+\frac{1}{2}\,\frac{\partial U_1}{\partial\rho}\left[\frac{1}{\rho_0^2}\,Z_{1\phi}^2+Z_{1z}^2\right]-\frac{Z_1}{\rho_0^3}\,\frac{\partial U_0}{\partial\rho}\,Z_{1\phi}^2\\&+\frac{\partial U_0}{\partial\rho}\left[\frac{1}{\rho_0^2}\,Z_{1\phi}Z_{2\phi}+Z_{1z}Z_{2z}\right].\end{aligned}$$

APPENDIX B

Terms corresponding to Sec. IV B:

$$\alpha_{L1}^{(0)}=-\rho_1(1/\rho_0^2-\alpha_{L0}/\rho_0)/\mathscr{A}_\lambda,$$

$$\alpha_{L1}^{(1)}=-\frac{\rho_0\alpha_{L0}+\nu^2+x^2-1}{\rho_0^2[K_\nu(x)-C_\nu I_\nu(x)]},$$

$$\alpha_{S1}^{(0)}=\rho_1/\rho_0^2,$$

$$\alpha_{S1}^{(1)}=-\frac{\nu^2+x^2-1}{\rho_0^2 I_\nu(x)},$$

$$V_{N1}^{(0)}=\rho_1\,\frac{\alpha_{L0}}{\rho_0^2}-\frac{\alpha_{L1}^{(0)}}{\rho_0},$$

$$V_{N1}^{(1)}=-\frac{x\alpha_{L1}^{(1)}}{\rho_0}\,\{K_\nu'(x)-C_\nu I_\nu'(x)\}+\beta\alpha_{S1}^{(1)}xI_\nu'(x)/\rho_0+\alpha_{L0}/\rho_0^2,$$

where

$$\alpha_{L0}=\frac{1-\rho_0}{\rho_0\mathscr{A}_\lambda},$$

$$C_\nu=\frac{K_\nu(xe^{\mathscr{A}_\lambda})}{I_\nu(xe^{\mathscr{A}_\lambda})},$$

$$x=k\rho_0.$$

The terms corresponding to Sec. IV C: We write the inhomogeneous terms in Eqs. (3.17)–(3.19) as

$$I=I^{(A)}+\rho_1 I^{(B)}\cos(kz)\cos(\nu\phi)+I^{(C)}\cos(2kz)\cos(2\nu\phi)+I^{(D)}\cos(2kz)+I^{(E)}\cos(2\nu\phi),$$

where

$$I_{L2}^{(A)}=\frac{\alpha_{L0}}{8\rho_0^2}-\frac{x}{4\rho_0}\,\alpha_{L1}^{(1)}[K_\nu'(x)-C_\nu I_\nu'(x)]+\frac{1}{8\rho_0^3}\,(x^2+3\nu^2-2),$$

$$I_{L2}^{(C)}=\frac{\alpha_{L0}}{8\rho_0^2}-\frac{x}{4\rho_0}\,\alpha_{L1}^{(1)}[K_\nu'(x)-C_\nu I_\nu'(x)]+\frac{1}{8\rho_0^3}\,(-x^2+5\nu^2-2),$$

$$I_{L2}^{(D)}=\frac{\alpha_{L0}}{8\rho_0^2}-\frac{x}{4\rho_0}\,\alpha_{L1}^{(1)}[K_\nu'(x)-C_\nu I_\nu'(x)]+\frac{1}{8\rho_0^3}\,(-x^2+3\nu^2-2),$$

$$I_{L2}^{(E)}=\frac{\alpha_{L0}}{8\rho_0^2}-\frac{x}{4\rho_0}\,\alpha_{L1}^{(1)}[K_\nu'(x)-C_\nu I_\nu'(x)]+\frac{1}{8\rho_0^3}\,(x^2+5\nu^2-2),$$

$$I_{S2}^{(A)}=-\frac{1}{4\rho_0}\,\alpha_{S1}^{(1)}xI_\nu'(x)+\frac{1}{8\rho_0^3}\,(x^2+3\nu^2-2),$$

$$I_{S2}^{(C)}=-\frac{1}{4\rho_0}\,\alpha_{S1}^{(1)}xI_\nu'(x)+\frac{1}{8\rho_0^3}\,(-x^2+5\nu^2-2),$$

$$I_{S2}^{(D)}=-\frac{1}{4\rho_0}\,\alpha_{S1}^{(1)}xI_\nu'(x)+\frac{1}{8\rho_0^3}\,(-x^2+3\nu^2-2),$$

$$I_{S2}^{(E)}=-\frac{1}{4\rho_0}\,\alpha_{S1}^{(1)}xI_\nu'(x)+\frac{1}{8\rho_0^3}\,(x^2+5\nu^2-2),$$

$$I_{U2}^{(A)}=\frac{\alpha_{L0}}{8\rho_0^3}\,(2-x^2-\nu^2)+\frac{x^2}{4\rho_0^2}\,\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S1}^{(1)}I_\nu''(x)\}-\frac{1}{4\rho_0^2}\,(x^2+\nu^2)\{\alpha_{L1}^{(1)}[K_\nu(x)-C_\nu I_\nu(x)]-\beta\alpha_{S1}^{(1)}I_\nu(x)\},$$

$$I_{U2}^{(C)}=\frac{\alpha_{L0}}{8\rho_0^3}\,(2+x^2+\nu^2)+\frac{x^2}{4\rho_0^2}\,\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S1}^{(1)}I_\nu''(x)\}+\frac{1}{4\rho_0^2}\,(x^2+\nu^2)\{\alpha_{L1}^{(1)}[K_\nu(x)-C_\nu I_\nu(x)]-\beta\alpha_{S1}^{(1)}I_\nu(x)\},$$

$$I_{U2}^{(D)}=\frac{\alpha_{L0}}{8\rho_0^3}\,(2+x^2-\nu^2)+\frac{x^2}{4\rho_0^2}\,\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S1}^{(1)}I_\nu''(x)\}-\frac{1}{4\rho_0^2}\,(-x^2+\nu^2)\times\{\alpha_{L1}^{(1)}[K_\nu(x)-C_\nu I_\nu(x)]-\beta\alpha_{S1}^{(1)}I_\nu(x)\},$$

$$I_{U2}^{(E)}=\frac{\alpha_{L0}}{8\rho_0^3}\,(2-x^2+\nu^2)+\frac{x^2}{4\rho_0^2}\,\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S}^{(1)}l_\nu''(x)\}+\frac{1}{4\rho_0^2}\,(-x^2+\nu^2)\times\{\alpha_{L1}^{(1)}[K_\nu(x)-C_\nu I_\nu(x)]-\beta\alpha_{S1}^{(1)}I_\nu(x)\}.$$

For the second order solution:

$$\alpha_{L2}^{(0)}=-\frac{I_{L2}^{(A)}}{\mathscr{A}_\lambda}+\rho_2\,\frac{\rho_0\alpha_{L0}-1}{\rho_0^2\mathscr{A}_\lambda},$$

$$\alpha_{L2}^{(1)}=\rho_1\,\frac{I_{L2}^{(B)}}{K_\nu(x)},$$

$$\alpha_{L2}^{(2)}=\frac{I_{L2}^{(C)}}{K_{2\nu}(2x)-C_{2\nu}I_{2\nu}(2x)},$$

$$\alpha_{L2}^{(3)}=\frac{I_{L2}^{(D)}}{K_0(2x)-C_0I_0(2x)},$$

$$\alpha_{L2}^{(4)}=\frac{I_{L2}^{(E)}}{e^{2\nu\mathscr{A}_\lambda}-e^{-2\nu\mathscr{A}_\lambda}},$$

$$\alpha_{S2}^{(0)}=\rho_2\,\frac{1}{\rho_0^2}+I_{S2}^{(A)},$$

$$\alpha_{S2}^{(1)}=\rho_1\,\frac{I_{S2}^{(B)}}{I_\nu(x)},$$

$$\alpha_{S2}^{(2)}=\frac{I_{S2}^{(C)}}{I_{2\nu}(2x)},$$

$$\alpha_{S2}^{(3)}=\frac{I_{S2}^{(D)}}{I_0(2x)},$$

$$\alpha_{S2}^{(4)}=\frac{I_{S2}^{(E)}}{\rho_0^{2\nu}},$$

where $I_\nu(x),I_{2\nu}(2x),I_0(2x),K_0(2x),K_\nu(x),K_{2\nu}(2x)$ are modified Bessel functions and

$$C_{2\nu}=\frac{K_{2\nu}(2k\rho_\infty)}{I_{2\nu}(2k\rho_\infty)}.$$

The terms corresponding to Sec. IV: The third order inhomogeneous terms in Eqs. (3.17)–(3.19) can be written as

$$I=I^{(B)}\cos(k2)\cos(\nu\phi)+\cdots,$$

where

$$I_{L3}^{(B)}=\rho_2\left[\frac{\alpha_{L0}}{\rho_0^2}-\alpha_{L1}^{(1)}\,\frac{x}{\rho_0}\,[K_\nu'(x)-C_\nu I_\nu'(x)]-\frac{2}{\rho_0^3}+\frac{2\nu^2}{\rho_0^3}-\frac{\rho_0\alpha_{L0}-1}{\rho_0^3\mathscr{A}}\right]+I_{L3}^{*},$$

$$I_{S3}^{(B)}=\rho_2\left[-\alpha_{S1}^{(1)}\,\frac{xI_\nu'(x)}{\rho_0}-\frac{2}{\rho_0^3}+\frac{2\nu^2}{\rho_0^3}\right]+I_{S3}^{*},$$

$$I_{U3}^{(B)}=\rho_2\left[2\,\frac{\alpha_{L0}}{\rho_0^3}+\frac{x^2}{\rho_0^2}\,\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S1}^{(1)}I_\nu''(x)\}-\frac{1}{\mathscr{A}_\lambda\rho_0^4}\,(\rho_0\alpha_{L0}-1)\right]+I_{U3}^{*},$$

with

$$I_{L3}^{*}=-\frac{3}{16}\frac{\alpha_{L0}}{\rho_0^3}+\frac{I_{L2}^{(A)}}{\rho_0\mathscr{A}_\lambda}-\frac{x}{2\rho_0}I_{L2}^{(C)}\frac{K_{2\nu}'(2x)-C_{2\nu}I_{2\nu}'(2x)}{K_{2\nu}(2x)-C_{2\nu}I_{2\nu}(2x)}-\frac{x}{\rho_0}I_{L2}^{(D)}\frac{K_0'(2x)-C_0I_0'(2x)}{K_0(2x)-C_0I_0(2x)}+\frac{\nu}{\rho_0}I_{L2}^{(E)}\frac{e^{2\nu\mathscr{A}_\lambda}+e^{-2\nu\mathscr{A}_\lambda}}{e^{2\nu\mathscr{A}_\lambda}-e^{-2\nu\mathscr{A}_\lambda}}$$
$$-\frac{9}{32}\frac{x^2}{\rho_0^2}\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]+\frac{9}{16\rho_0^4}+\frac{\nu^2x^2}{16\rho_0^4}-\frac{45}{32}\frac{\nu^2}{\rho_0^4}+\frac{9}{32}\left(\frac{\nu^4}{\rho_0^4}+\frac{x^4}{\rho_0^4}\right),$$

$$I_{S3}^{*}=-\frac{x}{2\rho_0}I_{S2}^{(C)}\frac{I_{2\nu}'(2x)}{I_{2\nu}(2x)}-\frac{x}{\rho_0}I_{S2}^{(D)}\frac{I_0'(2x)}{I_0(2x)}-I_{S2}^{(E)}\frac{\nu}{\rho_0}-\frac{9}{32}\frac{x^2}{\rho_0^2}\alpha_{S1}^{(1)}I_\nu''(x)+\frac{9}{16\rho_0^4}-\frac{45}{32}\frac{\nu^2}{\rho_0^4}+\frac{\nu^2x^2}{16\rho_0^4}+\frac{9}{32}\left(\frac{x^4}{\rho_0^4}+\frac{\nu^4}{\rho_0^4}\right),$$

$$I_{U3}^{*}=-\frac{18}{32}\frac{\alpha_{L0}}{\rho_0^4}+\frac{9}{32}\frac{x^3}{\rho_0^3}\{\alpha_{L1}^{(1)}[K_\nu'''(x)-C_\nu I_\nu'''(x)]-\beta\alpha_{S1}^{(1)}I_\nu'''(x)\}+\frac{I_{L2}^{(A)}}{\rho_0^2\mathscr{A}_\lambda}+\frac{x^2}{\rho_0^2}\{\alpha_{L2}^{(2)}[K_{2\nu}''(2x)-C_{2\nu}I_{2\nu}''(2x)]$$
$$-\beta\alpha_{S2}^{(2)}I_{2\nu}''(2x)\}+\frac{2x^2}{\rho_0^2}\{\alpha_{L2}^{(3)}[K_0''(2x)-C_0I_0''(2x)]-\beta\alpha_{S2}^{(3)}I_0''(2x)\}+\frac{\nu}{\rho_0^2}\{\alpha_{L2}^{(4)}[(2\nu+1)e^{2\nu\mathscr{A}_\lambda}-(2\nu-1)e^{-2\nu\mathscr{A}_\lambda}]$$
$$-\beta(2\nu-1)\alpha_{S2}^{(4)}\rho_0^{2\nu}\}+\frac{9x}{32\rho_0^3}\{-\alpha_{L1}^{(1)}[K_\nu'(x)-C_\nu I_\nu'(x)]+\beta\alpha_{S1}^{(1)}I_\nu'(x)\}(\nu^2+x^2)+\frac{3\alpha_{L0}}{32\rho_0^4}(\nu^2+x^2)$$
$$+\frac{3\nu^2}{8\rho_0^3}\{\alpha_{L1}^{(1)}[K_\nu(x)-C_\nu I_\nu(x)]-\beta\alpha_{S1}^{(1)}I_\nu(x)\}+\frac{\nu^2}{\rho_0^2}(-\alpha_{L2}^{(4)}(e^{2\nu\mathscr{A}_\lambda}-e^{-2\nu\mathscr{A}_\lambda})+\beta\alpha_{S2}^{4}\rho_0^{2\nu})+\frac{x^2}{\rho_0^2}$$
$$\times\{-\alpha_{L2}^{(3)}[K_0(2x)-C_0I_0(2x)]+\beta\alpha_{S2}^{(3)}I_0(2x)\}+\frac{1}{2\rho_0^2}(\nu^2+x^2)\{-\alpha_{L2}^{(2)}[K_{2\nu}(2x)-C_{2\nu}I_{2\nu}(2x)]+\beta\alpha_{S2}^{(2)}I_{2\nu}(2x)\}$$
$$+\frac{3\nu^2}{16\rho_0^4\,\alpha_{L0}}.$$

The third order constants are

$$\alpha_{L3}^{(1)}=\frac{I_{L3}^{(B)}}{K_\nu(x)-C_\nu I_\nu(x)},$$

$$\alpha_{S3}^{(1)}=\frac{I_{S3}^{(B)}}{I_\nu(x)},$$

$$\rho_2(x,\beta,\nu)=\mathscr{B}/\mathscr{D},$$

where

$$\mathscr{B}=-\frac{xI_{L3}^{*}}{\rho_0}\left(\frac{K_\nu'(x)-C_\nu I_\nu'(x)}{K_\nu(x)-C_\nu I_\nu(x)}\right)+\beta I_{S3}^{*}\frac{x}{\rho_0}\frac{I_\nu'(x)}{I_\nu(x)}-I_{U3}^{*},$$

$$\mathscr{D}=\frac{x}{\rho_0}\left(\frac{K_\nu'(x)-C_\nu I_\nu'(x)}{K_\nu(x)-C_\nu I_\nu(x)}\right)\left(\frac{\alpha_{L0}}{\rho_0^2}-\frac{x\alpha_{L1}^{(1)}}{\rho_0}[K_\nu'(x)-C_\nu I_\nu'(x)]-\frac{2}{\rho_0^3}+\frac{2\nu^2}{\rho_0^3}-\frac{\alpha_{L0}\rho_0-1}{\rho_0^3\mathscr{A}_\lambda}\right)-\beta\frac{x}{\rho_0}\frac{I_\nu'(x)}{I_\nu(x)}$$
$$\times\left(-\frac{x}{\rho_0}\alpha_{S1}^{(1)}I_\nu'(x)-\frac{2}{\rho_0^3}+\frac{2\nu^2}{\rho_0^3}\right)+\left(\frac{2\alpha_{L0}}{\rho_0^3}+\frac{x^2}{\rho_0^2}\{\alpha_{L1}^{(1)}[K_\nu''(x)-C_\nu I_\nu''(x)]-\beta\alpha_{S1}^{(1)}I_\nu''(x)\}-\frac{\rho_0\alpha_{L0}-1}{\rho_0^4\mathscr{A}_\lambda}\right).$$

[1] P. P. Debroy and R. F. Sekerka, Phys. Rev. E **51**, 4608 (1995).
[2] W. W. Mullins and R. F. Sekerka, J. Appl. Phys. **34**, 323 (1963).
[3] R. F. Sekerka, in *Encyclopedia of Material Science and Engineering*, edited by Michael B. Bever (Pergamon, New York, 1986), pp. 3486–3493.
[4] L. N. Brush and R. F. Sekerka, J. Cryst. Growth **96**, 419 (1989).
[5] L. N. Brush, R. F. Sekerka, and G. McFadden, J. Cryst. Growth **100**, 89 (1990).
[6] S. R. Coriell and R. L. Parker, J. Appl. Phys. **36**, 632 (1965).
[7] G. B. McFadden, S. R. Coriell, and B. T. Murray, in *Variational and Free Boundary Problems*, edited by A. Friedman and J. Spruck (Springer-Verlag, Berlin, 1993), pp. 159–169.