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Planar solidification from an undercooled melt: Asymptotic solutions to a continuum model with interfacial kinetics

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Planar growth of a solid germ from an undercooled melt is considered within the continuum model, accounting for kinetic effects at the interface. The paper extends previous studies of this problem by (i) analyzing not only the moving fronts but also the temperature fields in each phase and (ii) accounting for the temperature dependence of the latent heat. Explicit analytic solutions are developed both for short and long times. It is shown that, in the case of critical undercooling (at the crossover from diffusion to kinetics-dominated regimes), the nonuniformity of the solid temperature and the variation of the latent heat with temperature significantly affect the long-time (t) behavior $R=\gamma t^{2/3}$ of the phase-change fronts R. Emergence of this law is related to the entropy production at the interface. Peculiarities of the temperature field, derived for the critical undercooling case, are clarified.

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I. INTRODUCTION

The unconstrained planar solidification of a pure substance from an undercooled melt is one of the key problems in crystal-growth theory. Dynamics of this process depends on the initial undercooling, parametrized by the Stefan number $\mathrm{St}=c_L(T^*-T_i)/L^*$. Here T^* and T_i denote the equilibrium and the initial temperatures, respectively, L^* is the latent heat at the temperature $T=T^*$, and c_L is the liquid specific heat, assumed to be constant. The classical Stefan formulation of a diffusion-controlled growth with sharp fronts $x=R(t)$, held at $T_c=T^*$, implies $R(t)\sim t^{1/2}$ for $\mathrm{St}<1$ [1]. It is no longer adequate for rapid solidification ($\mathrm{St}\geq 1$), due to the departure from equilibrium at the interface. This difficulty is resolved within the phase-field approach. It allows for a finite thickness of the front by introducing an order-parameter field that is coupled to the temperature. The phase-field models [2–6] predict that for $\mathrm{St}<1$ the fronts of long-time asymptotic states advance as $t^{1/2}$, as in the Stefan solutions. At $\mathrm{St}=1$ two scenarios are possible, depending on the diffusion rates of the order parameter and of the heat. One scenario yields a solution , the front of which propagates with a constant velocity, defined by the microscopic considerations. The second scenario yields fronts advancing at long times as $R\sim t^{2/3}$. In the case $\mathrm{St}>1$ (hypercooling), this approach yields traveling-wave-type solutions with a constant front velocity.

Recently, it has been pointed out [6] that the long-time asymptotic states found in the phase-field models could be derived using simpler continuum models with sharp fronts by incorporating the effects of linear interfacial kinetics [7–16]. Such models are states as follows:

$$T_{S,t}=\alpha_S T_{S,xx}, \quad x<R(T),$$
$$T_{L,t}=\alpha_L T_{L,xx}, \quad R(t)<x<\infty \ , \tag{1}$$

$$T_L|_{x=R}=T_S|_{x=R}=T_c=T^*-aR_t' \ ,$$
$$\rho L R_t'=k_S T_{s,x}-k_L T_{L,x}, \quad x=R(t) \ . \tag{2}$$

Here the subscripts L and S refer to the liquid and to the solid, respectively, k is the thermal conductivity, $\rho_S=\rho_L=\rho$ is the density, $\alpha=k/\rho c$ is the thermal diffusivity, and a is the kinetic coefficient. For $c_L\neq c_S$, L is a function of T_c, reflecting for the entropy production at the interface. As long as $aR_t'/T^*\ll 1$, the rate of entropy production at the interface σ and the corresponding value of L are given by [10–12]

$$\sigma=a\rho L^* R_t'^2/T^*, \quad L=L^*-a(c_L-c_S)R_t' \ . \tag{3}$$

Such models, indeed, were utilized in studies of planar fronts. The exact traveling-wave-type solutions were found for $\mathrm{St}>1$ [7,13,14]. The $\mathrm{St}=1$ case was first considered in [14], analyzing the growth of a semi-infinite solid into a uniformly undercooled melt. It was found that for long times $R\sim t^{2/3}$. In [15] the long-time asymptotics of the fronts was addressed within the one-phase formulation ($k_S=0$). Again, $R\sim t^{2/3}$ was obtained for a uniform initial undercooling with $\mathrm{St}=1$. The physics of the diffusion and the kinetics-dominated growth of a solid germ from a supercooled melt has been recently clarified by Oswald [16]. Using the overall heat balance and making several *ad hoc* assumptions concerning the temperature field, he reached conclusions supporting the main results of the phase-field models for $\mathrm{St}<1$ and $\mathrm{St}>1$ and yielding $R\sim t^{2/3}$ for the critical case $\mathrm{St}=1$ at the crossover from diffusion to kinetics-dominated growths.

The above models, concerned with the long-time behavior of the fronts, did not develop explicit analytic expressions for the temperature field and often ignored the heat diffusion in the emerging solid. Derivation of such expressions would provide a more detailed picture of the process. It might also affect some quantitative results. Most of the above works (with the notable exception of [7]) disregarded the temperature dependence of the latent heat. As shown in [7] for $\mathrm{St}>1$ the latter dependence, indeed, affects advance of the fronts and therefore should be incorporated in the analysis of the problem with an arbitrary initial undercooling.

The present paper addresses these issues for a planar growth of a solid germ. This growth is treated essentially as a two-phase problem, stated by Eqs. (1)–(3), accounting for the heat diffusion in each phase, and for unequal thermal conductivities and specific heats of solid and liquid. (The latter implies a temperature-dependent latent heat.)

II. FORMULATION OF THE PROBLEM

We assume that the solid germ of infinitesimal thickness has nucleated at $x=0$ in a uniformly undercooled melt with $T_L(0,x)=T_\infty$. The initial temperature of the germ, $T_S(0,0)$, is assumed to be equal to T_∞. Since the growth is symmetric with respect to the $x=0$ plane, our analysis is restricted to the semi-infinite strip occupied by the solid $[0<x<R(t)]$, and by the melt $[R(t)<x<1]$. It is assumed that the heat flux vanishes at infinity. Thus the initial-boundary data are stated as follows:

$$\begin{aligned} &T_L(0,x)=T_i=T_S(0,0)=T_L(t,\infty)=T_\infty\ ,\\ &T_{S,x}|_{x=0}=T_{L,x}|_{x=0}=0\ ,\\ &R(0)=0\ . \end{aligned} \tag{4}$$

Our choice of the germ-growth problem, rather than that for the growth of a semi-infinite solid [2–6,14], is suggested by the previous works [5] and [16]. Yet the data (4) differ from those of [5], where growth of a localized germ was studied within phase-field models: In [5] the germ had a finite initial width and its initial temperature was well above that of the melt, whereas in Eq. (4) the initial germ temperature is equal to T_∞, and the width of the germ at $t=0$ is negligibly small. The data (4) are of the same nature as those of [9], stated for a slow ($\mathrm{St}<1$) radial growth, accounting for interfacial kinetics and neglecting the Gibbs-Thompson effect. We attempt to mimic the latter problem within a planar setup for $\mathrm{St}<1$, and to treat also the case $\mathrm{St}\geq 1$.

The key element of our analysis is the global enthalpy balance. It is obtained by integrating Eqs. (1) with respect to x and t and using Eqs. (2)–(4):

$$c_S\int_0^R(T_S-T^*)dx+c_L\int_R^\infty(T_L-T_\infty)dx+c_L(T^*-T_\infty)R=L^*R\ . \tag{5}$$

The analysis is significantly simplified by introducing the natural space-time scales $t_0=\alpha_S a^2/(T^*-T_\infty)^2$, $x_0=(\alpha_S t_0)^{1/2}$, along with the dimensionless quantities: $\theta=(T-T^*)/(T^*-T_\infty)$, $\alpha=\alpha_L/\alpha_S$, $k=k_L/k_S$, $c=c_L/c_S$, $x'=x/x_0$, $\tau=t/t_0$, $r=R/x_0$, $r'=dr/d\tau$. It is also convenient to reformulate the problem in terms of variables $\xi=x'/r$ and τ, in which the interface is at rest. This yields

$$r^2\theta_{S,\tau}=\theta_{S,\xi\xi}+\xi rr'\theta_{S,\theta},\quad 0<\xi<1 \tag{6}$$

$$r^2\theta_{L,\tau}=\alpha\theta_{L,\xi\xi}+\xi rr'\theta_{L,\xi}\ ,\quad 0<\xi<\infty \tag{7}$$

$$\theta_S|_{\xi=1}=\theta_L|_{\xi=1}=-r'\ , \tag{8}$$

$$rr'(c/\mathrm{St})[1-r'\mathrm{St}(c-1)/c]=\theta_{S,\xi}|_{\xi=1}-k\theta_{L,\xi}|_{\xi=1}\ , \tag{9}$$

$$\theta_{S,\xi}|_{\xi=0}=\theta_{L,\xi}|_{\xi\to\infty}=0,\quad \theta_L|_{\xi\to\infty}=-1,\quad r(0)=0\ . \tag{10}$$

In terms of the dimensionless variables the overall heat balance reads

$$c\int_1^\infty(\theta_L+1)d\xi+\int_0^1\theta_S d\xi=c[(1/\mathrm{St})-1]\ . \tag{11}$$

III. SHORT-TIME ASYMPTOTIC BEHAVIOR

We now analyze the initial asymptotic behavior of the problem. The initial and boundary conditions imply that at very short times

$$r(\tau)=\tau[1-B\tau^\beta/(\beta+1)+\cdots]\ , \tag{12}$$

where β and B are still undetermined constants. This equation, along with Eqs. (8) and (10), suggests the following form of θ_L:

$$\theta_L(\xi,\tau)=-1+B\tau^\beta F(\xi,\tau)\ . \tag{13}$$

Such a profile of θ_L is compatible with a constant right-hand side of Eq. (11), if $F(\xi,\tau)$ is a function of a single variable $\nu=(\xi-1)(\tau/\alpha)^\beta$. It follows from Eq. (7) that $\beta=\frac{1}{2}$, and $F(\nu)$ is subjected to

$$2F_{,\nu\nu}+\nu F_{,\nu\nu}-F=0,\quad F|_{\nu=0}=1,\quad F|_{\nu\to\infty}=0\ . \tag{14}$$

The latter problem admits an analytic solution [1]. It yields

$$\theta_L=-1+B\sqrt{\pi\tau}\,i\,\mathrm{erfc}[(\xi-1)\sqrt{\tau/\alpha}/2]\ . \tag{15}$$

Let us consider now the sensible heat of the solid germ, applying the heat balance integral method [17,18]. Integrating Eq. (6) with respect to ξ and using the boundary conditions (8) and (10) one obtains

$$r\left[r\int_0^1\theta_S d\xi\right]_{,\tau}+rr'^2=\theta_{S,\xi}|_{\xi=1}\ . \tag{16}$$

Assuming a parabolic profile for θ_S, accounting for a zero heat flux at $\xi=0$, and for $\theta_S|_{\xi=1}=-r'$, at the front, and using Eq. (16), one obtains

$$\begin{aligned} &\theta_S(\xi,\tau)=A(\tau)(\xi^2-1)-r'\ ,\\ &A'=-3r''/2-3A/r^2-Ar'/r\ . \end{aligned} \tag{17}$$

For $r(\tau)$ given by Eq. (12) with $\beta=\frac{1}{2}$, $A(\tau)$ is of the order $t^{3/2}$, whereas the interface temperature $-r'=-1+B\tau^{1/2}$. Thus at the onset of freezing $\theta_S\approx -r'$ and the heat diffusion in the solid is negligibly small. Inserting this value of θ_S, and Eq. (15) for θ_L, into Eq. (11) yields $B=2/\mathrm{St}'\sqrt{\pi\alpha}$, where $1/\mathrm{St}'=[(1/c)+(1/\mathrm{St})-1]$. This indicates that the asymptotic solution for $r(\tau)$ given above is just the first term in expansion of $r/\tau-1$ in powers of $\sqrt{\tau}/\mathrm{St}'$, adequate as long as $\tau\ll \mathrm{St}'^2$.

IV. LONG-TIME SOLUTIONS

We develop now the long-time asymptotic solutions. The previous studies [6,7,16] suggest that for long times the decay length of θ_L (in terms of the variable ξ) is typically of the order α/rr'. This fact, along with the overall heat balance, Eq. (11), indicates that θ_L is a function of τ and $\mu=(\xi-1)rr'/\alpha$. In terms of τ and μ, Eq. (7) reads

$$\theta_{L,\tau}=(r'^2/\alpha)[\theta_{L,\mu\mu}+(1+\mu\alpha g)\theta_{L,\mu}] ,$$
$$0\le\mu<\infty, \quad g=-r''/r'^3 . \qquad (18)$$

For the exact traveling-wave solutions [7] (St > 1) θ_L depends only on μ. For all other scenarios $r'(\tau)$ decreases monotonically as $\tau\to\infty$. These facts suggest that $\theta_{L,\tau}$ can be neglected in the leading order in Eq. (18) [19]. The resulting equation can be integrated explicitly [20]:

$$\theta_L=-1+(1-r')\mathrm{erfc}[\Omega(1+\mu\alpha g)]/\mathrm{erfc}\Omega ,$$
$$\Omega=1\sqrt{2g\alpha} . \qquad (19)$$

Such θ_L satisfies the boundary conditions (8) and (10). As shown below for $\tau\to\infty$ it yields the similarity solutions for St < 1, the traveling waves advancing with a constant speed for St > 1, and a rather peculiar form of θ_L in the critical case St = 1. The latter profile shares some features of kinetics-dominated and diffusion-dominated solutions. Inserting Eq. (19) into Eq. (11) and adopting the parabolic profile (17) for θ_S yields

$$-r'-2A/3+c(1-r')[W(\Omega)-1]/grr'=c(-1+1/\mathrm{St}) . \qquad (20)$$

Here $W(\Omega)=[\sqrt{\pi}\Omega\exp(\Omega^2)\mathrm{erfc}\Omega]^{-1}$. We now look for solutions of Eq. (20) in the form $r\approx\gamma\tau^\beta$ with $\frac{1}{2}\le\beta\le1$, and $A(t)$ defined by Eq. (17). For $\beta=\frac{1}{2}$, both r' and A are of the order $t^{-1/2}$. Thus in the leading order Eq. (20) reduces to St = $1/W(\Omega)$. It is identical to the equation governing the similarity solution of the classical Stefan problem [1]. It has real roots only for St < 1. Consequently, in the long-time regime $\theta_S=0(\tau^{-1/2})$ and

$$\theta_L\approx-1+\mathrm{erfc}(\Omega\xi)/\mathrm{erfc}\Omega+0(t^{-1/2}) . \qquad (21)$$

Thus for St < 1 the similarity solution of the corresponding Stefan problem is an asymptotic attractor of the solution to the continuum model, accounting for the interfacial kinetics.

Let us consider now Eq. (19) for $\frac{1}{2}<\beta\le1$. Using the asymptotic expansion of the error function for large arguments [1], one obtains in the leading order

$$\theta_L\approx-1+(1-r')[\exp(-\mu-\mu^2 g\alpha/2)]/(1+\mu\alpha g) . \qquad (22)$$

Consequently, the heat balance (20) reduces to

$$-r'-2A/3+c(1-r')\alpha/rr'=c(-1+1/\mathrm{St}) . \qquad (23)$$

For $\beta=1$, $A(t)$ is exponentially small as $\tau\to\infty$, and Eqs. (17), (22), and (23) yield for long times the traveling-wave-type solution [7] with $R(t)=c(\mathrm{St}-1)(L^*/ac_L)t$. For the sake of convenience we return to the x',τ frame in which

$$\theta_L\approx-1+(1-c+c/\mathrm{St})\exp\{-[x'-r(\tau)]r'/\alpha\} ,$$
$$\theta_S\approx-r'\approx-c(1-1/\mathrm{St}) . \qquad (24)$$

For St = 1, Eqs. (17) and (23) yield in the leading order $r=\gamma\tau^{2/3}$, $A=\gamma\tau^{-1/3}$, $\gamma=(9c\alpha/8)^{1/3}$, $R(t)=(9\alpha_L cL^*/8ac_L)^{1/3}t^{2/3}$. (These results also follow from the heat balance at the interface, Eq. (9), combined with Eqs. (17) and (22).) Consequently, in the x',τ frame

$$\theta_S\approx-(\tfrac{2}{3})\gamma\tau^{-1/3}[1+3\{(x'^2)/(\gamma^2\tau^{4/3})-1\}/2] , \qquad (25)$$

$$\theta_L=1+\mathrm{erfc}\{\gamma\tau^{1/6}\sqrt{2/3\alpha}[1+(x'-\gamma\tau^{2/3})]/(2\gamma\tau^{2/3})\}/\mathrm{erfc}(\tau^{1/6}\gamma\sqrt{2/3\alpha})$$
$$\approx-1+\frac{\exp[-r'(x'-\gamma\tau^{2/3})/\alpha]\exp\{-[(x'-\gamma\tau^{2/3})/\sqrt{6\alpha\tau}]^2\}}{1+(x-\gamma\tau^{2/3})/2\gamma\tau^{2/3}} . \qquad (26)$$

The liquid temperature profile describes a pulse, propagating with a time-dependent velocity r', and involves three time-dependent scales, characterizing the decay of this pulse along the x' axis. The largest one is that of the denominator $l_1(\tau)=r(\tau)=\gamma\tau^{2/3}$. The next scale is the diffusion scale $l_2(\tau)=\sqrt{6\alpha\tau}$, associated with the "amplitude" $\exp[(x'-\gamma\tau^{2/3})/\sqrt{6\alpha\tau}]$. The shortest length scale $l_3(\tau)=3\alpha\tau^{1/3}/2\tau$ is that of the wave part $\exp[-(r'/\alpha)(x'-r)]$. It is this scale that dominates the spatial decay of the liquid temperature profile and hereby enters the enthalpy balance (23). Thus, effectively, the liquid temperature profile is dominated by the wave-type behavior

$$\theta_L(x',\tau)\approx-1+\exp\{-(r'/\alpha)[x'-r(\tau)]\} , \qquad (26a)$$

which differs from that of (24) by the time-dependent velocity and by a preexponential factor, which in the present case is just equal to the Stefan number St = 1. This manifest the relaxation of the interfacial temperature to its equilibrium value, contrary to the traveling-wave-type solutions for St > 1. It is worthwhile mentioning that the solution (26) stands in agreement with the ansatz suggested in [4] and [6] within the phase-field approach.

V. DISCUSSION

Let us compare the above results with those of [15] and [16]. In terms of dimensionless variables the overall heat balance, stated in [16], is identical to Eq. (23) with $A=0$, $c=1$, $\alpha=1$. It implies a uniform solid temperature $\theta_S=-r'$ and assumes that the decay length of θ_L (in terms of ξ) is $1/rr'$. At short times the nonuniformity of θ_S in the solid is indeed negligibly small, but the decay length of θ_L is of the order $1/\sqrt{rr'}$ [see Eq. (15)]. At long times our results for $c=\alpha=1$ are identical to Oswald's for St > 1. For St < 1, the long-time solution developed above tends to the similarity solution of the Stefan problem, corresponding to the parabolic law $r\approx\gamma\tau^{1/2}$. Oswald's solution also yields $r=\gamma\tau^{1/2}$, but with a different value of γ. For St$=1$ and $A=0$, $c=\alpha=1$, assumed in [16], Eq. (23) yields $r=(\frac{9}{4})^{1/3}\tau^{2/3}$, whereas the heat balance at the interface, Eq. (9), gives $r=(\frac{9}{8})^{1/3}\tau^{2/3}$. This inconsistency is due to the nonuniformity of θ_S, neglected in [16]. It does not occur in our analysis [see the term $-2A/3$ in Eq. (23)].

The long-time asymptotic solutions for $R(t)$ derived above generalize those of [15], developed within the one-phase model ($k_S=0$), and ignoring the temperature dependence of the latent heat. For a uniform initial undercooling with St>1, the value of $R(t)$ found in [15] is $1/c$ times smaller than that obtained in the present paper. For St$=1$, our analysis yields $R(t)$, which is greater than that of [15] by a factor $c^{1/3}$. For St<1 the long-time solution for $R(t)$ found above is identical to that of [15].

The one-phase formulation ($k_S=0$) implies that no heat is supplied to the solid. For the classical Stefan problem this means $T_S=T^*$, whereas for the model with interfacial kinetics it yields

$$c_S\rho\int_0^R (T^*-T_S)dx = ac_S\rho\int_0^t R_t'^2 dt = c_S T^{*2}S/L^*, \quad S(t)=\int_0^t \sigma dt\ . \tag{27}$$

The left-hand side of this equation is the difference between the amount of heat needed to raise the temperature of emerging solid to T^*, and the actual solid heat content. The right-hand side of Eq. (27) involves the entropy $S(t)$ produced at the interface. From Eqs. (5) and (27) follows the one-phase formulation of the liquid heat balance,

$$c_L\rho\int_{R(t)}^{\infty}(T_L-T^*)dx = c_S T^{*2}S/L^* + \rho R L^*(1-\text{St})\ . \tag{28}$$

Inserting Eq. (19) for T_L into Eq. (28) yields asymptotic solutions for $R(t)$, which generalize those of [15] for arbitrary values of c. These solutions for $R(t)$ obtained within the one-phase formulation are identical to those derived above using the two-phase model. This can be attributed to a negligibly small heat flux into solid at long times. Indeed, $\partial\theta_L/\partial\xi|_{\xi=1}$ is of the order rr', whereas $\partial\theta_S/\partial\xi|_{\xi=1}$ is of the order r' for St≤ 1 and is exponentially small for St>1. Equation (27) is satisfied by T_S, found in the two-phase formulation, when St≥ 1. Therefore, for St$=1$ the heat content of the melt is proportional to the entropy produced at the interface and is equal to the difference between the heat needed to raise the solid temperature to T^* and the actual heat content of the solid. For St<1, the solid temperature obtained within the two-phase formulation violates Eq. (27). Yet, when $t\to\infty$, the solid contributions in Eqs. (5) and (28) are small compared with the remaining terms. In this regime Eqs. (28) and (5) are identical in the leading order, so that the resulting long-time solutions for $R(t)$ also coincide.

To summarize: We derived the short-time and the long-time asymptotic solutions for the temperature fields and for advancing fronts. The short-time regime is determined by the effective Stefan number St$'$, which accounts for the temperature dependence of the latent heat. At this stage, the heat diffusion in the solid is negligible. At long times, the profiles of temperature for the diffusion-dominated (St<1), the kinetics-dominated (St>1), and for the critical (St$=1$) growths follow as particular limits of the general expressions for the liquid and for the solid temperatures. The heat flux into solid diminishes in the course of the process and the long-time asymptotic solutions can be treated within the one-phase formulation. For St<1 the present solution tends to the similarity solution of the corresponding Stefan problem. For St$=1$ the solid temperature is not uniform. This nonuniformity, and the temperature dependence of the latent heat, affect the interface propagation, which advances as $R(t)=[9\alpha_L cL^*/8ac_L]^{1/3}t^{2/3}$. For St$=1$ the liquid temperature profile exhibits a wave-type behavior. This wave decays in space on the shortest (kinetic) scale, whereas its amplitude decays on the diffusional (long) scale. One of the peculiarities of the case St$=1$ is the proportionality of the sensible heat content of the melt to the heat associated with the entropy production at the interface. For St>1 the long-time solution tends to the asymptotic attractor of the traveling-wave type [7], propagating with a constant velocity: $R(t)=c(\text{St}-1)(L^*/ac_L)t$.

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