MULLINS-SEKERKA STABILITY ANALYSIS FOR MELTING-FREEZING WAVES IN HELIUM-4

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MULLINS-SEKERKA STABILITY ANALYSIS FOR MELTING-FREEZING WAVES IN HELIUM-4

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Abstract. This paper considers the stability of the melt-solid interface to eigenfunction perturbations in a system of equations which describe the melting and freezing of helium. The analysis is carried out in both planar and spherical geometries. The principal results are that when the melt is freezing, under certain far-field conditions, the interface is stable in the sense of Mullins and Sekerka. On the other hand, when the solid is melting (at least when the melting is sufficiently fast), the interface is unstable. In some circumstances, these instabilities are oscillatory, with amplitude and growth rate increasing with surface tension and frequency. The last section considers the original problem of Mullins and Sekerka in the present notation.

Key words. melting-freezing waves, linear stability analysis, Lyapunov functions

AMS(MOS) subject classifications. 35R35

§1. Introduction.

The phenomenon known as *melting-freezing waves* where oscillations appear in the solidification of helium-4 from a melt has been observed in a number of experiments, e.g., Keshishev, Parshin & Babkin (1979) and Castaing, Balibar & Laroche (1980). A general review of surface effects for helium-4 was given by Maris & Andreev (1987). These waves occur in helium for two principal reasons: for helium the latent heat is essentially zero, and the thermal conductivity is very large. The motion is principally of the solid-liquid interface rather than of the material itself. In particular, material in the solid remains static during the melting-freezing process, and the melt moves only due to density changes. The density of the solid is roughly 20% greater than that of the liquid. It is also worth noting that the helium crystal is atomically rough (nonfaceted) at relatively high temperatures (above 1.28 K), while at lower temperatures it may be atomically rough or smooth.

Gurtin (1990) derived from first-principles a mechanical theory for this solidification process for an atomically rough interface. In this theory, solidification is described by the following system of equations:

$$\begin{aligned} &\Delta u = 0, && \text{melt} \subset \mathbb{R}^3, \\ u_t = -\sigma\kappa + \beta V, \quad &u_m = \alpha V, && \text{interface}. \end{aligned} \tag{1.1}$$

This system is consistent with a rigid helium crystal (solid) and an inviscid, irrotational, incompressible melt which satisfies the *weak-inertia approximation.* This approximation

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asserts that flow in the liquid phase is sufficiently slow so that higher order velocity terms may be ignored [Gurtin (1990), pp 299-300]. The functions of space and time are as follows:

$$\begin{aligned} \mathbf{v}(\mathbf{x},t) &: \text{ melt velocity,} \\ u(\mathbf{x},t) &: \text{ rescaled potential: } \nabla u(\mathbf{x},t) = -(\rho_c - \rho)\mathbf{v}, \\ \mathbf{X}(\mathbf{p},t) &: \text{ position of the interface (parameterized by } \mathbf{p}), \\ V(\mathbf{p},t) &: \text{ normal component of the interface velocity,} \\ \kappa(\mathbf{p},t) &: \text{ interface curvature (twice the mean curvature).} \end{aligned}$$

The system also includes several constants:

$$\begin{aligned} \sigma &: \text{ surface tension (interfacial energy),} \\ \beta &: \text{ kinetic coefficient,} \\ \alpha &\equiv (\rho - \rho_c)^2/\rho, \\ \rho &: \text{ density in the melt,} \\ \rho_c &: \text{ density in the crystal.} \end{aligned}$$

The subscript m denotes the normal derivative into the melt. The solid-melt region for helium is shown in Figure 1.

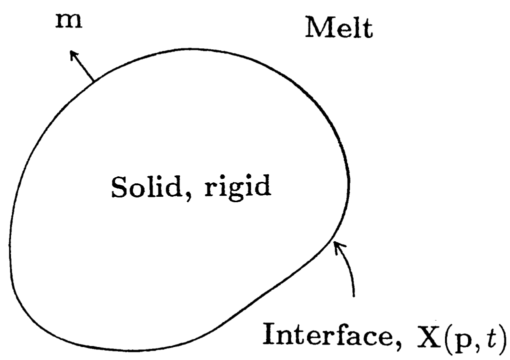

Fig. 1. Schematic of the solid-melt region for helium. **m** denotes the outer unit normal.

When no interface kinetics are considered, i.e., $\beta = 0$ in equation $(1.1)_2$, system (1.1) then has a form very similar to that of the classic Mullins–Sekerka problem for solidification from a melt:

$$\begin{aligned} \Delta u = 0, &\qquad \text{melt} \\ u = \sigma\kappa, \quad u_m = -\alpha V, &\qquad \text{interface} \end{aligned} \tag{1.2}$$

Mullins & Sekerka (1963) studied the question of interface stability for system (1.2). The goal of this paper is to consider this question for system (1.1) and hence for the solidification of helium. The issue of interface stability in a similar system describing flow in a Hele-Shaw cell has been studied by Saffman & Taylor (1958). The similarities between (1.1) and (1.2) inspire the following definition.

DEFINITION. An interface is **stable in the sense of Mullins and Sekerka** (or simply **stable**) on some initial time interval if, for the linearized system, all eigenfunction perturbations of the interface decay during that initial interval. On the other hand, an interface is **unstable in the sense of Mullins and Sekerka** (or **unstable**) at a time t if, for the linearized system, any eigenfunction perturbations of the interface grow starting from t.

Remarks.

(1) There is an intuitive sense to the terms "grow" and "decay" which is not always easy to verify directly. For example, for an oscillatory system the amplitude of perturbations could be both increasing and decreasing on an interval where, say, one would wish to claim that the perturbations grow. To make the terms exact, one needs to establish conditions under which all (any) phase-plane trajectories of the perturbation amplitude monotonically converge to the origin (diverge to infinity), asymptotically for large time. If such convergence conditions then hold for some initial period of time, the interface will be stable in the sense of Mullins and Sekerka for that period. If the divergence conditions are satisfied starting at some time, the interface becomes unstable in the sense of Mullins and Sekerka at that time. Once the onset of such an instability has occurred, the planar or spherical symmetry is lost, and the linear analysis is of little value. As we shall see, requiring that trajectories converge to the origin implies that both the amplitude and the velocity of perturbations must go to zero for the interface to be stable. On the other hand, the interface will be unstable if either of the two become unbounded.

(2) The convergence or divergence conditions for this analysis will be established by finding Lyapunov (or energy) functions. Such functions prove that perturbations decay (grow) by proving that the energy associated with those perturbations is monotonically decreasing (increasing) for all (some) solution trajectories.

(3) The terms "linearly stable" and "linearly unstable" are often used for the type of stability described in the definition. The alternate terms used in the definition have the advantage of helping make distinct the concept defined here and the separate, though related, concept of linear stability as used for equilibrium points of dynamical system. Stable in the sense of Mullins and Sekerka, for example, is more akin to asymptotically stable in the sense of dynamical systems.

(4) These definitions do not exhaust all possible cases; one can imagine situations where the interface is neither stable nor unstable. For example, the perturbation could remain constant in time, or the energy might alternately increase and decrease without ever diverging or going to zero.

The principal results here are that under certain physically meaningful far-field conditions, system (1.1) is stable in the sense of Mullins and Sekerka for a freezing solid, but is unstable at least to perturbations of very high frequency when the solid melts sufficiently fast. When the solid melts slowly, stability also depends on the geometry. How fast the melt rate needs to be to assure instability depends on the size of β. When $\beta = 0$, even a slowly melting solid would be expected to be unstable. The instability of high frequencies (short wavelengths) is not surprising in view of experiments detecting oscillatory frequencies of 10^{11} Hz! Details of this analysis for both planar and spherical geometry are presented in §2 and §3. Three specific far-field conditions are considered in the spherical case: (1) constant growth rate, (2) constant radial flux, and (3) constant far-field pressure. In the planar case the far-field condition is the specification of a uniform flux. In §4, the present work is compared to the original analysis of Mullins & Sekerka (1963).

It is interesting to note that little is known about existence and uniqueness for either system (1.1) or (1.2) in $\mathbb{R}^3$. In $\mathbb{R}^2$ when the interface is of the form $y = f(x)$, short time existence and uniqueness were proven for (1.2) by Duchon & Robert (1984). If one considers system (1.2) with $(1.2)_2$ replaced by

$$u = \sigma\kappa - \beta V, \tag{1.3}$$

the situation is somewhat different. In general the presence of interface kinetics would be expected to make the problem somewhat easier since equation (1.3) is parabolic when $\beta > 0$ (then $V \sim \partial_t \mathbf{X}$ and $\kappa \sim \partial_{xx}\mathbf{X}$). Still existence and uniqueness for the problem $(1.2)_1$, (1.3), $(1.2)_3$ remain open. For the case of the Stefan problem where the Laplace equation is replaced by the heat equation, short time existence and uniqueness have been proven by Chen & Reitich (1990) when $\beta > 0$, and global existence of weak solutions has been proven by Luckhaus (1990) when $\beta = 0$.

Also note that since

$$\begin{aligned} u(\mathbf{X}(\mathbf{p},t),t) &= \int_0^t \frac{du}{ds}(\mathbf{X}(\mathbf{p},s),s)ds \\ &= \int_0^t V \cdot \nabla u + u_t \\ &= \int_0^t \alpha V^2 - \sigma\kappa + \beta V, \end{aligned} \tag{1.4}$$

one can replace the interface condition for u_t by (1.4). Indeed one can posed the problem equivalently by choosing any two of the three conditions for u, u_t and u_m.

§2. Mullins-Sekerka Analysis: Planar Geometry.

Consider first the case where the unperturbed interface is a plane separating two half spaces with the crystal (solid) below and the melt above. Under what conditions will small sinusoidal perturbations of this plane grow and under what conditions will they decay? To answer these questions, suppose that the unperturbed interface is at $z = Z(t)$. For uniform planar melting or freezing, the general solution of $\Delta u = u_{zz} = 0$ is $u(z,t) = u_0(t) + g(t)z$; then the far-field velocity in the melt is

$$\mathbf{v} = -g(t)\mathbf{k}/(\rho_c - \rho)$$

where $\mathbf{k}$ is the unit normal to the interface. System (1.1) will then have a single planar solution once a uniform far-field velocity (or flux) is specified by specifying $g(t)$.

Let the position of the perturbed interface be $z(x, y, t) = Z(t) + \delta(t)\sin(\omega\xi_1 x)\sin(\omega\xi_2 y)$ where $\delta(t)$ is the amplitude of the perturbation and $\xi_1^2 + \xi_2^2 = 1$. The various sinusoidal perturbations can be considered separately because these functions are eigenfunctions of the Laplacian. The solution of $\Delta u = 0$ corresponding to this perturbation and a uniform far-field velocity is

$$u(x, y, z, t) = u_0(t) + g(t)z + c(t)e^{-\omega(z-Z(t))}\sin(\omega\xi_1 x)\sin(\omega\xi_2 y).$$

In (1.1) and (1.2), κ is defined as twice the mean curvature, i.e.,

$$\kappa \equiv -\frac{1}{R_M} - \frac{1}{R_m}$$

where R_M and R_m are the two principle radii of curvature [cf. eg. O'Neill (1966), §§5]. Then to first order in δ,

$$\kappa(x, y, t) = -\omega^2\delta(t)\sin(\omega\xi_1 x)\sin(\omega\xi_2 y) + O(\delta^2).$$

Since $V(t) \equiv Z'(t) + \delta'(t)\sin(\omega\xi_1 x)\sin(\omega\xi_2 y)$, on substituting the expressions for u and κ into $(1.1)_{2,3}$ and linearizing about the planar solution, one obtains four equations. These yield that

$$Z(t) = \frac{1}{\alpha}\int_0^t g(s)ds,$$

and on eliminating $c(t)$ and $Z(t)$, the following equation for δ:

$$\delta'' + \omega\left(\frac{g}{\alpha} + \beta\right)\delta' + \omega(\omega^2\sigma - g')\delta = 0. \tag{2.1}$$

Equation (2.1) is the equation of principal interest in this section since it is the behavior of δ which determines whether and how the interface is becoming nonplanar. The first result addresses the stability question for a planar interface.

THEOREM 1. *Consider a planar interface dividing a helium melt above from a solid helium region below. Suppose that the motion of this interface is governed by (1.1) with far-field melt conditions given by specifying $g \in C^2$. Then the growth or decay of sinusoidal perturbations of the interface is governed by (2.1). If the solid is melting sufficiently fast so that $g + \alpha\beta$ is bounded less than zero, and if there is an M such that $g' \leq M$ and $g'' \leq 0$, then the planar interface is unstable in the sense of Mullins and Sekerka for all sinusoidal frequencies satisfying $\omega^2\sigma > M$. On the other hand, if the solid is either slowly melting or* *freezing such that $g + \alpha\beta$ is bounded greater than zero and if $g' \leq 0$ and $g'' \geq 0$, then the planar interface is stable in the sense of Mullins and Sekerka for all sinusoidal frequencies.*

Proof. Multiplying (2.1) by δ' and integrating by parts, one finds an energy for (2.1):

$$(2.2) \qquad (\delta')^2 + \omega(\omega^2\sigma - g')\delta^2 = C_p(\omega) - \omega \int_0^t g''\delta^2 - 2\omega \int_0^t \left(\frac{g}{\alpha} + \beta\right)(\delta')^2$$

where

$$C_p(\omega) \equiv (\delta'(0))^2 + \omega(\omega^2\sigma - g'(0))\delta^2(0).$$

is constant in time. From the phase plane, one sees that no trajectories of (2.1) stay in a neighborhood of the rays $\delta' = 0$, $\delta > \epsilon$ for any fixed ϵ. Therefore along any trajectory, the right-hand side of (2.2) is strictly increasing and becoming unbounded when $g'' \leq 0$ and $g + \alpha\beta$ is bounded less than zero. Under these conditions, provided $|g'|$ is bounded, equation (2.2) guarantees a growing elliptical region in the phase plane which solution trajectories must avoid. If g' is becoming unboundedly negative, then the coefficient of δ^2 is also increasing. However, from the phase plane, one sees immediately that trajectories for (2.1) diverge or are oscillatory. In the later case, δ' must be becoming arbitrarily large when $\delta = 0$. So the system is still unstable.

With $g + \alpha\beta$ bounded greater than zero, $g' \leq 0$ and $g'' \geq 0$, however, (2.2) implies that

$$\Psi_p(\delta, \gamma, t) \equiv \gamma^2 + \omega(\omega^2\sigma - g')\delta^2$$

is a strict Lyapunov function for (2.1). So $(\delta, \gamma) = 0$ is asymptotically stable in this later case. •

Remarks.

(1) One of the significant cases occurs when g is simply constant. In this steady-state case, (2.1) is just a constant coefficient, second order ODE, and its general solution can be given explicitly. The quantum mechanical theory for melting-freezing waves of Andreev & Parshin (1978) also predicts a stable steady-state regime [cf. Maris & Andreev (1987)].

(2) Not all freezing conditions are stable. For example, take $g = g_0 + t$. This accelerating freezing is sufficiently fast so that perturbations with $\omega < 1/\sqrt{2\sigma}$ can be shown to be unstable by considering the phase plane for (2.1) with $g' = 1$. In this case, however, there are no time oscillations.

(3) Clearly when $g + \alpha\beta \equiv 0$, perturbations neither grow or decay. Hence this case would be *marginally* stable.

Something more can be said about the rapid decay case of Theorem 1 (the unstable case). Let $g(t) = -\alpha v$ (constant) with $v > 0$. As mentioned in the remark above, the explicit general solution of (2.1) for $\omega > (v - \beta)^2/2\sigma$ then is simply

$$\delta(t) = e^{\frac{\omega(v-\beta)}{2}t} \left\{ c_1 \sin\left(\frac{\omega}{2}\sqrt{4\omega\sigma - (v-\beta)^2}\, t\right) + c_2 \cos\left(\frac{\omega}{2}\sqrt{4\omega\sigma - (v-\beta)^2}\, t\right) \right\}.$$

From this solution, one immediately sees that not only is the interface unstable (i.e., not only is δ growing in time), but that δ oscillates in time with a frequency which increases with ω, and that the growth rate of these oscillations also increases with ω. When g is not constant but changes slowly in time, one would expect that the behavior of δ would be similar. The next theorem addresses this situation.

THEOREM 2. *Suppose that δ satisfies (2.1) and suppose that δ_0 satisfies*

$$\delta_0'' + \omega\left(\frac{g(0)}{\alpha} + \beta\right)\delta_0' + \omega(\omega^2\sigma - g'(0))\delta_0 = 0. \tag{2.3}$$

Let

$$\omega > \frac{1}{4\sigma}\left(\frac{g(0)}{\alpha} + \beta\right)^2 + \frac{g'(0)}{\omega\sigma}$$

be fixed so that δ_0 oscillates. Let $T \equiv 2\pi N/\sqrt{\sigma\omega^3}$ so that δ_0 has approximately N oscillations in the interval $[0,T]$ when ω is large. Then $||\delta(t) - \delta_0(t)||_{C^1[0,T]}$ will be arbitrarily small through approximately N oscillations provided that $||g(t) - g(0)||_{C^2[0,T]}$ is sufficiently small.

Proof. This theorem is essentially just continuous dependence of the equation on its coefficients. Write (2.1) and (2.3) as first order systems:

$$\dot{u}(t) = A(t)u(t) \qquad \text{and} \qquad \dot{u}_0(t) = A(0)u_0(t). \tag{2.4}$$

Let $E(t) \equiv u(t) - u_0(t)$. Then taking the difference between $(2.4)_1$ and $(2.4)_2$, integrating, and applying the mean value theorem, one finds that for some $\xi \in [0,t]$,

$$E(t) = \int_0^t A(s)\left(u(s) - u_0(s)\right) + \left(A(\xi) - A_0\right)\int_0^t u_0(s)$$

where $A_0 \equiv A(0)$. Hence

$$|E(t)| \le (||A_0|| + \epsilon)\int_0^t |E(s)| + \epsilon \sup_{\tau\in[0,T]}\left|\int_0^\tau u_0(s)\right|$$

where $||g(t) - g(0)||_{C^2[0,T]}$ is sufficiently small so that $||A_0 - A(\cdot)|| < \epsilon$. Applying Gronwall's inequality, one obtains the estimate

$$|E(t)| \le \epsilon\left[\sup_{\tau\in[0,T]}\left|\int_0^\tau u_0(s)\right|\right]e^{(||A_0||+\epsilon)t}$$

which assures that provided $||g(t) - g(0)||_{C^2[0,T]}$ is sufficiently small, the difference between δ and δ_0 is arbitrarily small through N oscillations. •

Remarks.

(1) Theorem 2 implies that when the planar solid is rapidly melting, δ can become arbitrarily large and oscillates arbitrarily fast as $\omega \rightarrow \infty$. This result is particularly interesting in view of the observation of waves with frequencies as high as 10^{11} Hz [cf. eg. Huber & Maris (1981)]. The result also suggests a complete loss of smoothness for the interface. This is a very different from what is known and what is expected for the Mullins–Sekerka problem. Both Theorems 1 and 2 do not depend on the problem being posed in $\mathbb{R}^3$. In particular, the results are the same in $\mathbb{R}^2$. For the Mullins–Sekerka problem, Duchon & Robert (1984) have shown that in two dimensions the interface is at least C^1.

(2) The role of surface tension here is different from that in the classical Mullins-Sekerka problem. Here surface tension can act to *destabilize* the interface in the sense that it leads to sinusoidal oscillations in $\delta(t)$. The role of interface kinetics, on the other hand, *is* stablizing.

For general $g(t)$, it may be very difficult to decide whether solutions of (2.1) oscillate. Certainly behavior similar to the constant coefficient equation would not be expected if the coefficients are changing rapidly. But the oscillations of (2.1) may still increase with frequency. For example, let $g(t) = -\alpha\mu t$ for some positive constant μ. This case is of particular interest in regard to Theorem 1 since assuming that g' is bounded and that $g'' \leq 0$ implies that g' approaches a constant. Define $\tau \equiv \sqrt{\omega\mu/2}(t - \beta/\mu)$ and $\nu \equiv (\omega^2\sigma + \alpha\mu)/\mu$. Then (2.1) becomes the Hermite equation

$$\delta_{\tau\tau} - 2\tau\delta_\tau + 2\nu\delta = 0,$$

one of whose solutions is the Hermite function $H_\nu(\tau)$. From Lebedev (1972), the asymptotic behavior of $H_\nu(\tau)$ for large ν is

$$H_\nu(\tau) \sim 2^{(\nu+1)/2}\nu^{\nu/2}e^{\tau^2/2}\cos\left(\sqrt{2\nu+1}\,\tau - \frac{\nu\pi}{2}\right), \qquad \nu \rightarrow \infty.$$

Thus the solutions in this case also oscillate with frequency and growth rate which increases with ω. And as in the constant coefficient case, one can use Gronwall's inequality to show that if g is "near" being proportional to t, then solutions exhibit similar behavior.

§3. Mullins-Sekerka Analysis: Spherical Geometry.

Now consider a spherical crystal in an infinite melt with radius $R(t)$ at time t. The results in this geometry are similar to those for a planar interface, but the analysis is now somewhat more complicated. Suppose that the position of the solid–liquid interface is given by

$$r(\theta, \phi, t) = R(t) + \delta(t)Y_{\ell,m}(\theta, \phi) \tag{3.1}$$

where $Y_{\ell,m}$ is the (ℓ,m)-th spherical harmonic and again δ is the amplitude of the perturbation. For this unbounded domain, the appropriate solution of $\Delta u = 0$ can be written as

$$u(r,\theta,\phi,t) = u_\infty(t) + \frac{B(t)}{r} + \frac{D(t)}{r^\ell} Y_{\ell,m}(\theta,\phi) \tag{3.2}$$

where u_∞ is the far-field value of u and $B(t) = (\rho_c - \rho)\Phi(t)/4\pi\rho$ with Φ the far-field flux. Since κ is defined as twice the mean curvature, for this geometry to lowest order in δ,

$$\kappa(\theta,\phi,t) = -\frac{2}{R(t)} + \frac{2-\ell(\ell+1)}{R^2(t)}\delta(t)Y_{\ell,m}(\theta,\phi) + O(\delta^2).$$

Define volume (or mass):

$$\mathcal{V} \equiv \frac{\alpha}{3}R^3.$$

Then proceeding as before, now with $V(t) \equiv R'(t) + \delta'(t)Y_{\ell,m}(\theta,\phi)$, linearization about the spherical solution yields that $B = -\mathcal{V}'$. Eliminating B and D, one finds that δ satisfies

$$3\mathcal{V}\delta'' + \alpha R^2(\ell+4)\left[R' + \frac{\ell+2}{\ell+4}\frac{\beta}{\alpha}\right]\delta' + (\ell-1)\left[(\ell+1)(\ell+2)\sigma - \alpha R^2 R''\right]\delta = 0. \tag{3.3}$$

Again this δ-equation is the one of principal interest in determining the stability of the interface.

In addition, one finds that u_∞ satisfies

$$Ru'_\infty = 2\sigma + \mathcal{V}'' + \beta R R'. \tag{3.4}$$

Equation (3.4) gives the connection in spherical geometry between the far-field value of the potential and the uniform flux. As in the planar case, one must specify some far-field condition in order to have a single symmetric ($\delta = 0$) solution. In general by specifying various values for u_∞, one would expect a wide range of behaviors for δ in (3.3). There are, however, three far-field conditions which seem of particular physical interest: (1) constant spherical growth rate $R' \equiv g_R$, (2) constant uniform flux $\mathcal{V}' \equiv g_V$, and (3) constant far-field pressure. Using (3.4) to find u_∞ for the first two conditions is straightforward. As for the third, we make use of the *Bernoulli Equation* [Eqn. (5.8), Gurtin (1990)]:

$$p = -\rho(\phi_t + \mathbf{v}^2/2) \tag{3.5}$$

where $\nabla\phi \equiv \mathbf{v}$. The relationship between the potentials u and ϕ is

$$u = -(\rho_c - \rho)\phi - Ft \tag{3.6}$$

where F is a constant determined by the solid and melt energies. This relationship (3.6) is used in the derivation of (1.1). Combining (3.5) and (3.6), and using the weak-inertia approximation to drop the $\mathbf{v}^2$-term, one derives that

$$(\rho_c - \rho)p = \rho(u_t + F) \tag{3.7}$$

[cf. Eqn. (5.15), Gurtin (1990)]. Therefore constant far-field pressure implies that u_t is constant at infinity.

THEOREM 3. *In $\mathbb{R}^3$ suppose that a solid helium sphere of radius R exists in an infinite helium melt. Suppose also that the motion of the melt-solid interface is governed by (1.1) with far-field boundary conditions such that either (1) the growth rate of the sphere is constant, (2) the uniform radial flux rate is constant, or (3) the far-field pressure is constant. Then the growth or decay of spherical harmonic perturbations is governed by equation (3.3). Suppose the melt is freezing, i.e, $R' \geq 0$, and for the constant pressure case with $u'_\infty > 0$, suppose that the initial conditions lie in the region of the phase plane where $f' \geq 0$ (f defined below; cf. Figure 2). Then the sphere is stable in the sense of Mullins and Sekerka for all spherical harmonic frequencies. On the other hand, suppose that the sphere is melting sufficiently rapidly so that*

$$R' < -\frac{\beta(\ell_0+2)}{\alpha(\ell_0+5/2)},$$

and for the constant pressure case with $u'_\infty > 0$, that the initial conditions lie in the region of the phase plane where $f' \leq 0$. Then the sphere is unstable in the sense of Mullins and Sekerka for all spherical harmonic perturbations with index $\ell \leq \ell_0$ (and for all ℓ if $R' \leq -\beta/\alpha$).

Proof. The proof is of course similar to that of Theorem 1: multiply (3.3) by δ' and integrate by parts. The resulting energy is

$$\begin{aligned}&\alpha R^3(\delta')^2 + (\ell-1)\left[(\ell+1)(\ell+2)\sigma - \alpha R^2 R''\right]\delta^2 = \\ &\qquad C_s(\ell) - \alpha(2\ell+5)\int_0^t R^2\left[R' + \frac{\beta(\ell+2)}{\alpha(\ell+\frac{5}{2})}\right](\delta')^2 - \alpha(\ell-1)\int_0^t (R^2R'')'\,\delta^2\end{aligned} \tag{3.8}$$

where the initial constant is

$$C_s(\ell) = \alpha R^3(0)(\delta'(0))^2 + (\ell-1)\left[(\ell+1)(\ell+2)\sigma - \alpha R^2(0)R''(0)\right]\delta^2(0).$$

For constant spherical growth rate, $R' = g_R$ (constant), and (3.8) reduces to

$$\alpha R^3(\delta')^2 + (\ell-1)(\ell+1)(\ell+2)\sigma\delta^2 = C_s(\ell) - \alpha(2\ell+5)\left[g_R + \frac{\beta(\ell+2)}{\alpha(\ell+\frac{5}{2})}\right]\int_0^t R^2(\delta')^2. \tag{3.9}$$

One can see from the phase plane that the the system is oscillatory and that no trajectories remain near either ray $\delta' = 0$, $|\delta| > \epsilon$ for any fixed ϵ. Therefore if

$$g_R < -\frac{\beta(\ell_0+2)}{\alpha(\ell_0+\frac{5}{2})},$$

the right-hand side of (3.9) is monotonically increasing for $\ell \leq \ell_0$ (and for all ℓ if $g_R < -\beta/\alpha$). And again there is an elliptical region whose size increases monotonically where trajectories are forbidden. For $g_R \geq 0$, on the other hand, equation (3.9) implies that

$$\Psi_s(\delta,\gamma,t) \equiv 3\mathcal{V}(t)\gamma^2 + (\ell-1)(\ell+1)(\ell+2)\sigma\delta^2$$

is a Lyapunov function for (3.3) in the constant growth rate case.

When the uniform radial flux is constant, $\mathcal{V}''(t) = 0$. So $R^2R'' = -2R(R')^2$, and $(R^2R'')' = 6(R')^3$, and hence (3.8) becomes

$$\begin{aligned} &\alpha R^3(\delta')^2 + (\ell-1)\left[(\ell+1)(\ell+2)\sigma + \frac{2g_V^2}{V}\right]\delta^2 = \\ &\qquad C_s(\ell) - \alpha(2\ell+5)\int_0^t R^2\left[R' + \frac{\beta(\ell+2)}{\alpha(\ell+\frac{5}{2})}\right](\delta')^2 - 3\alpha(\ell-1)\int_0^t (R')^3\delta^2. \end{aligned} \tag{3.10}$$

As in the constant growth rate case, the right-hand side of (3.10) is monotonically decreasing when $R' > 0$ so that

$$\Psi_s(\delta,\gamma,t) \equiv 3\mathcal{V}(t)\gamma^2 + (\ell-1)\left[(\ell+1)(\ell+2)\sigma + \frac{2g_V^2}{V}\right]\delta^2$$

is a Lyapunov function for this case. When the sphere is rapidly melting, the right-hand side of (3.10) is monotonically increasing. On the left-hand side, however, the coëfficient of δ^2 is also growing as $R \to 0$, and this makes the conclusion that all trajectories are diverging less than obvious. But since equation (3.3) is oscillatory, from (3.10) there are trajectories for which δ' must become arbitrarily large for $\delta = 0$ and R near zero. Therefore the system is unstable in this case.

Finally when the far-field pressure is constant, equation (3.7) allows one to define a constant $C \equiv -u'_\infty$. This constant may be positive, negative or zero; first consider $C \geq 0$. Then using (3.4), one finds that

$$-\alpha R^2R'' = 2\sigma + CR + 2\alpha R(R')^2 + \beta RR' \tag{3.11}$$

and

$$(R^2R'')' = \frac{8\sigma}{R}\left(R' + \frac{\beta}{4\alpha}\right) + \left[3C + 6\alpha R'\left(R' + \frac{\beta}{2\alpha}\right)\right]\left(R' + \frac{\beta}{3\alpha}\right). \tag{3.12}$$

Combining (3.8), (3.11) and (3.12), one again finds that the energy of each eigenmode is monotonically decreasing when $R' > 0$. Specifically,

$$\Psi_s(\delta,\gamma,t) \equiv 3\mathcal{V}(t)\gamma^2 + (\ell-1)\left[(\ell^2+3\ell+4)\sigma + CR + 2\alpha R(R')^2 + \beta RR'\right]\delta^2$$

is a Lyapunov function.

For $C < 0$, the situation is more complicated. For convenience, let $Q \equiv -C > 0$. Then write the equation for the radius of the sphere (3.4) as a first-order system:

$$\begin{aligned} \alpha R^2 y' &= -2\sigma + R(Q - 2\alpha y^2 - \beta y), \\ R' &= y. \end{aligned} \tag{3.13}$$

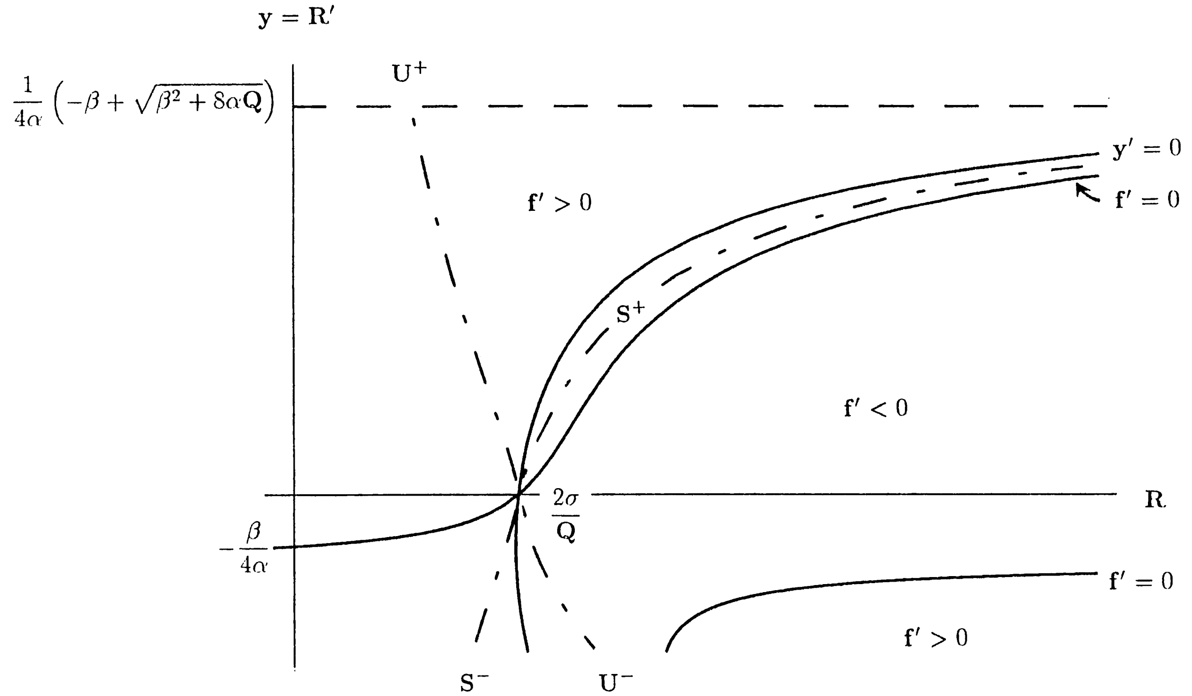

Fig. 2. Phase plane for spherical geometry when $u'_\infty > 0$ and $\beta > 0$. On the solid curves, $y' = 0$ or $f' = 0$. The dot-dash curves denote the stable and unstable manifolds of the phase plane. The dashed line is the asymptote for the solid lines and therefore for S^+.

The phase plane for system (3.13) when $\beta > 0$ is shown in Figure 2. When $QR \leq 2\sigma$, the argument is the same as for $C \leq 0$. When $QR > 2\sigma$, the key result is given by the following lemma:

LEMMA 1. *Let $f(t) \equiv R(Q - 2\alpha y^2 - \beta y)$. Consider trajectories which lie to the right of the unstable manifolds U^+ and U^- (cf. Fig. 2). On these trajectories,*

$$\lim_{t\to\infty} f(t) = \frac{8\sigma}{3 + \beta/\mathcal{B}} \tag{3.14}$$

where $\mathcal{B} \equiv \sqrt{\beta^2 + 8\alpha Q}$. Moreover, $f(t)$ is strictly increasing on trajectories above the stable manifold S^+, while for $\beta > 0$ on trajectories below S^+, a minimum value is achieved as the trajectories cross the curve

$$R = \frac{2\sigma(4\alpha y + \beta)}{(3\alpha y + \beta)(Q - 2\alpha y^2 - \beta y)}. \tag{3.15}$$

Once trajectories cross above (3.15), $f(t)$ is again strictly increasing.

Proof of Lemma. Using (3.13), one can compute that

$$f'(t) = \frac{2\sigma(4\alpha y + \beta) - f(t)(3\alpha y + \beta)}{\alpha R}, \tag{3.16}$$

and hence that $f'(t) = 0$ on the curve (3.15). One can also compute that the slope of (3.15) satisfies

$$R^2(3\alpha y + \beta)\left[1 + \frac{2\sigma\alpha\beta}{R(3\alpha y + \beta)(4\alpha y + \beta)}\right]\frac{dy}{dR} = 2\sigma,$$

while trajectories crossing (3.15) satisfy

$$R^2(3\alpha y + \beta)\frac{dy}{dR} = 2\sigma.$$

Therefore when $\beta > 0$, $f(t)$ is strictly increasing on any trajectory for sufficiently large t. Since by (3.16) the function $f(t)$ is also bounded above, the limit as $t \to \infty$ must exist. Using l'Hôpital's rule, (3.13) and the phase plane, one can find that this limit is given by (3.14). •

Lemma 1 and $(3.13)_1$ immediately imply that

$$\lim_{t\to\infty} \alpha R^2 R'' = 2\sigma\frac{\mathcal{B} - \beta}{3\mathcal{B} + \beta}$$

and that the limit is approached monotonically on trajectories above (3.15). Since $f'(t) = (\alpha R^2 R'')'$, the proof is complete provided that $f'(t) \geq 0$ and

$$\alpha R^2 R''(0) \leq 2\sigma\frac{\mathcal{B} - \beta}{3\mathcal{B} + \beta}.$$

These conditions are satisfied provided the initial conditions of the trajectory lie to the right of U^+ and above the curve (3.15) where $f' = 0$.

When the sphere is rapidly decaying, regardless of the value of C, the right-hand side of (3.8) is monotonically increasing, but from the phase plane for (3.11), the coefficient of δ^2 on the left-hand side is also increasing. Still as in the case of constant radial flux, equation (3.8) assures that δ' becomes arbitrarily large for at least some of the trajectories as $R \to 0$. So again the system is unstable. •

Remarks.

(1) Theorem 3 begs the question as to what happens when the sphere is slowly melting, i.e., $-\beta/\alpha \leq R' < 0$, or when initial conditions lie in other parts of the phase plane for the constant pressure case. For such cases, the right-hand-side terms compete with each other and/or with the coefficients on the left making simple energy arguments insufficient

to say that both the velocity and the displacement are tending to zero, or that either is necessarily becoming large. There are two cases of this type particularly worth noting: The first occurs when the growth rate is constant and

$$g_R \equiv -\frac{\beta(\ell_0 + 2)}{\alpha(\ell_0 + \frac{5}{2})}.$$

Then the right-hand side of (3.9) is constant, and since $R \to 0$, δ' must be unbounded while the maximum value of δ remains constant during each oscillation. But the relative size of δ to R of course is increasing. The second occurs when the pressure is constant and the initial conditions are $(R, y)(0) = (R_0, 0)$ with $R_0 u'_\infty \geq 2\sigma$. From Figure 2, one sees that $R' > 0$ for $t > 0$, but that what one would hope would be the Lyapunov function for this case will be *increasing* on some trajectories during some initial period of time before becoming monotonically decreasing.

(2) When the far-field pressure is constant and $u'_\infty \leq 0$, Gurtin (1990) [p. 309] describes the melt as being *stable relative to the crystal* in the sense that when interface stability issues are ignored, consideration of the phase plane yields that the radius will go to zero in finite time. For $u'_\infty > 0$, and initial conditions of the form $(R, y)(0) = (R_0, 0)$, the solid will grow arbitrarily large if $R_0 u'_\infty > 2\sigma$, and will melt in finite time if $R_0 u'_\infty < 2\sigma$. Gurtin refers to the melt in this freezing case as being *unstable relative to the crystal.* Unfortunately when $\beta > 0$, the entire R-axis lies outside the region where $f' > 0$. When $\beta \equiv 0$, the phase plane for R is somewhat different from Figure 2. The significant change is that now the curve on which $f' = 0$ is

$$R = \frac{8\sigma}{3(Q - 2\alpha y^2)} \tag{3.17}$$

rather than (3.15). This implies that for $\beta \equiv 0$ and initial conditions of the form $(R, y)(0) = (R_0, 0)$ with $2\sigma < R_0 u'_\infty \leq 8\sigma/3$, the sphere is freezing and stable in the sense of Mullins and Sekerka. Also note that when $\beta \equiv 0$, the curve (3.17) is an explicit stable trajectory of the phase plane. Thus for $\beta \equiv 0$, trajectories outside the stable region never cross into that region.

(3) Note that when the solid is freezing at a constant radial growth rate ($R(t) = R_0 + g_R t$) and $\beta = 0$, the far-field pressure is given by

$$p_\infty(t) = 2(\rho_c - \rho) g_R^2 + \frac{\rho}{\rho_c - \rho}\left[\frac{2\sigma}{R(t)} + F\right]$$

which *decreases* and approaches a constant value as the radius grows. Hence the constant growth rate and constant far-field pressure cases converge asymptotically when $\beta = 0$. On the other hand, in the constant radial flux case, $u'_\infty \to 0$ when $\beta = 0$.

(4) As in the planar case, when the sphere is freezing, not all far-field conditions lead to a stable interface. Consider a simple example where $R'/R = \mathcal{V}'/3\mathcal{V}$ is constant

(exponential growth). If this constant is large, as in the planar case, one can examine the phase plane to see that perturbations with small ℓ will grow (though again there will be no time oscillations).

(5) When $u'_\infty \equiv 0$, the radially symmetric problem with $\beta = 0$ has an explicit solution:

$$R^3 = R_0^3 - B_0 t - \sigma t^2$$

where B_0 is essentially the initial flux. Consist with the second remark above, however, for these parameter values, the sphere will always eventually be melting, and thus unstable in the sense of Mullins and Sekerka.

(6) One can also find an explicit solution of the form $2R^2R'' = C_2$ which is stable provided $0 \leq C_2 < 24\sigma$. But the form of this solution is sufficiently complicated so that it seems of little practical importance.

(7) Results similar to Theorem 2 can be obtained for this spherical case when the coefficients vary slowly as they were for the planar case.

§4. Classical Mullins-Sekerka Problem.

It is instructive now to compare the problem of the previous section and the classical Mullins-Sekerka problem for thermally-driven solidification from an undercooled melt. The system of equations describing the Mullins-Sekerka problem is (1.2) where now u is a nondimension temperature. Specifically,

$$u \equiv \frac{T - T_M}{T_M}, \qquad \alpha \equiv \frac{L_v}{k_L T_M}$$

where T_M is the melting temperature of a flat interface, L_v is the latent heat, and k_L is the thermal diffusivity in the melt. All other quantities are defined as before. Note that there is no heat flow in the solid, i.e., $k_S \equiv 0$. Also recall that (1.2) describes thermally-driven solidification only to the extent of the validity of the quasisteady-state approximation, i.e., only when the heat equation can be replaced by the Laplace equation.

Considering spherical geometry, let the solid-liquid interface be given by (3.1); again the corresponding solution of $\Delta u = 0$ to first order is (3.2). Proceeding as before, one finds that the equation for the perturbation is

$$\delta' = \frac{\ell - 1}{3\mathcal{V}} \left[\mathcal{V}' - (\ell + 1) \left((\ell + 2)\sigma + \frac{\beta R^2}{\ell - 1} \right) \right] \delta \tag{4.1}$$

where again $\mathcal{V} \equiv \frac{\alpha}{3} R^3$.

Equation (4.1) has two principal implications: First, it indicates that for this problem, melting is stable and freezing is stable provided that the rate of change in volume is smaller than a quantity depending on surface tension and interface kinetics. Note that this stability condition is *independent* of the far-field condition. When the condition that the temperature approach a constant undercooled value ($u \to u_\infty < 0$) and $\beta \equiv 0$, equation (4.1) becomes

$$\delta' = \frac{\ell - 1}{3\mathcal{V}}\big[- Ru_\infty - (\ell^2 + 3\ell + 4)\sigma\big]\delta,$$

and this is precisely the classical result balancing surface tension and the product of the radius and undercooling [Mullins & Sekerka (1963)]. The second point is that since (4.1) is a first-order equation, there are no oscillatory solutions for $\delta(t)$.

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