The effect of heat currents on the stability of the liquid solid interface

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The effect of heat currents on the stability of the liquid solid interface

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Abstract. — Rapid changing of the temperature of a liquid in equilibrium with its solid can lead to instabilities of the interface in two ways : the change in pressure, induced by a temperature change at the interface, leads to a uniaxial stress which can cause a Grinfeld instability at the capillary wavelength ; a temperature gradient is set up which modifies the effective gravity at the interface. When the effective gravity becomes negative, the interface is unstable at very long wavelengths. For a superfluid, such as ^{4}He, the situation is more complex. If we ignore surface dissipation, there is still a small critical temperature gradient across the solid above which the interface is unstable. However surface dissipation — in particular the growth resistance — pushes the instability to huge temperature gradients, ones which cannot be realised experimentally. The only instability that can be seen is caused by uniaxial stress.

Introduction.

Consider a solid in contact with its melt with the interface perturbed from equilibrium as illustrated in figure 1. If the solid is heavier than the liquid, both gravity and surface tension are stabilising : they act to push the interface back to the equilibrium position. A temperature gradient across the interface can be destabilising if the liquid is supercooled, the solid superheated, and the latent heat is positive. The solid would want to grow into colder liquid
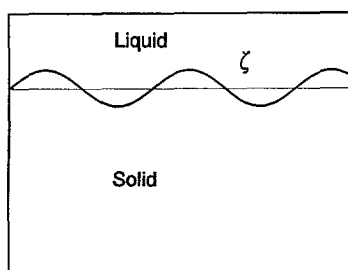

Fig. 1. — A disturbance of the interface between a solid and its melt.

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and so the perturbation would grow. Such an instability should be present for ordinary liquids.

Such a temperature gradient has been invoked in order to explain instabilities seen by Bodensohn *et al.* [1] on the interface between solid and liquid ^{4}He. They report that, when they cool the liquid, it spontaneously deforms with corrugations of the surface a distance of about 6 mm apart. This value is close to the capillary wavelength, $2 \pi (\gamma/g \, \Delta\rho)^{1/2}$, where γ is the surface stiffness and $\Delta\rho$ is the difference in density between solid and liquid, $\rho_S - \rho_L$. This interpretation cannot be correct because the instability appears at much longer wavelengths as we shall show. Rather they saw the effect of the Grinfeld [2, 3] instability caused by the creation of a uniaxial strain. Such a strain arises partly from the change in the liquid pressure at the interface : as the temperature drops so the pressure changes so as to keep on the melting curve. The strain also arises from the thermal expansion or contraction of the solid. This explanation of the observations of Bodensohn *et al.* was recently proposed by Balibar, Edwards and Saam [4].

In this paper we analyse the instability of a solid interface due to a temperature gradient. For an ordinary material, the temperature gradient is equivalent to a change in gravity, which can drive a standard Rayleigh instability. The situation is completely different in superfluid ^{4}He. In the absence of surface dissipation, the interface would be unstable if the temperature gradient exceeds a threshold ~ 1 mK/cm. However, the presence of surface dissipation increases the critical temperature gradient by a factor of 10^5 or more, so that it is impossible to observe the instability in ^{4}He crystals. A sudden change in temperature can lead to a uniaxial stain sufficient to trigger the Grinfeld instability ; we estimate the size of the temperature change which is needed.

1. Dynamics of a solid liquid interface in the absence of surface dissipation.

Assume that a d.c. heat current J_0 flows through the steady interface, implying zeroth order temperature gradients $G_S = - J_0/\kappa_S$, $G_L = - J_0/\kappa_L$ (κ_S and κ_L are the d.c. thermal conductivities in each phase). The interface is displaced by an amount

$$\zeta \simeq e^{ikx} \, e^{st} \tag{1}$$

The change in liquid pressure at the interface contains an hydrostatic part $- \rho_L \, g\zeta$ (due to exploration of the zeroth order profile), and an extra modulation ΔP_L which drives the flow through the liquid. If $s \ll ck$ (c = sound velocity), a quasistatic approximation is valid : ΔP_L is harmonic, such that $\Delta P_L \simeq e^{ikx - kz}$ The equation of motion at interface is then

$$k \, \Delta P_L = k (\delta p_L + \rho_L \, g\zeta) = \rho_L \, \dot{v}_L = - (\rho_S - \rho_L) \, \ddot{\zeta} \tag{2}$$

v_L is the normal liquid velocity at interface, ρ_S and ρ_L are specific masses. (In writing (2), we use conservation of matter). Similarly, temperatures at the interface change by amounts δT_L and δT_S on each side. Each of them contains a term $- G\zeta$ due to exploration of the zeroth order gradient, and an extra modulation that drives the heat current through liquid and solid. These heat currents, measured from solid to liquid, may thus be written as

$$Z_L J_{QL} = \delta T_L - G_L \, \zeta \qquad - Z_S J_{QS} = \delta T_S - G_S \, \zeta \tag{3}$$

where Z_L and Z_S are appropriate a.c. thermal impedances for each phase. J_{QL} and J_{QS} in turn must ensure energy conservation

$$J_{QL} - J_{QS} = \mathcal{L} \rho_S \, \dot{\zeta} \tag{4}$$

where $\mathcal{L}$ is the latent heat per unit mass.

We now assume that there is no surface dissipation. Then $\delta T_S = \delta T_L = \delta T$ (no Kapitza resistance). In the absence of capillarity, δp_L and δT must remain on the phase equilibrium curve,

$$\delta p_L = \frac{dp_L}{dT} \delta T = \frac{\mathcal{L} \rho_S \rho_L}{T(\rho_S - \rho_L)} \delta T . \tag{5}$$

From (3) and (4) we find δT. Using (2) and (5) we obtain the required dispersion relation

$$0 = \rho_L g + (\rho_S - \rho_L) \frac{s^2}{k} + \frac{dp_L/dT}{Z_L^{-1} + Z_S^{-1}} \left[sT \frac{\rho_S - \rho_L}{\rho_L} \frac{dp_L}{dT} - \left(\frac{1}{\kappa_L Z_L} + \frac{1}{\kappa_S Z_S} \right) J_0 \right] . \tag{6}$$

The generalization of (6) to more complex situations is straightforward. If we want to include *capillarity*, we must add to (5) the standard Gibbs correction. $\rho_L g$ is thus replaced by

$$\rho_L g + \gamma k^2 \frac{\rho_L}{\rho_S - \rho_L} = \rho_L g(k) \tag{7}$$

where γ is the surface energy (as usual, gravity is overtaken by capillarity as k increases). In order to include *surface dissipation*, we must allow for different temperatures at the interface, $\delta T_L \neq \delta T_S$, the growth being driven by the difference $(\mu_L - \mu_S)$. This will be done in the next section.

We first consider a normal liquid, with finite thermal conductivities in both phases. We assume $\rho_S > \rho_L$ — the liquid is on top such as to ensure mechanical stability. J_0 is oriented from solid to liquid : $J_0 > 0$ implies that the liquid is supercooled. The thermal impedances are diffusive (in a quasistatic approximation which holds near threshold) :

$$Z_L^{-1} = \kappa_L k \qquad Z_S^{-1} = \kappa_S k .$$

The dispersion relation (6) reduces to

$$0 = \rho_L g + (\rho_S - \rho_L) \frac{s^2}{k} \left[1 + \left(\frac{dp_L}{dT} \right)^2 \frac{T}{s\rho_L(\kappa_S + \kappa_L)} \right] - \frac{2 J_0}{\kappa_L + \kappa_S} \frac{dp_L}{dT} \tag{8}$$

When $J_0 = 0$, the Rayleigh waves are damped at low frequency by thermal dissipation (this limit is seldom achieved in practice). The effect of the d.c. heat current is just to replace g by an *effective gravity*

$$g_{\text{eff}} = g - \frac{2 J_0}{\rho_L(\kappa_S + \kappa_L)} \frac{dp_L}{dT} . \tag{9}$$

From now on, g is meant to be the combination $g(k)$ defined in (7). The interface undergoes a standard Rayleigh instability if g_{eff} becomes negative, which occurs for J larger than a critical heat current

$$J_c = \rho_L g \frac{dT}{dp_L} \frac{\kappa_S + \kappa_L}{2} \tag{10}$$

(J_c depends on the wave vector k through $g(k)$). Note that J_c is positive : heat must flow *into the high temperature phase* in order to destabilise the interface. The situation would be

reversed for water : the solid is then on top of the liquid, and J_c is negative — once again, heat flows into a supercooled liquid.

The result (8) for « normal » liquids is fairly obvious. It is not quite so for superfluid ^{4}He. In that case, the d.c. conductivity κ_L is infinite (i.e. $G_L = 0$) ; the thermal gradient exists only on the solid side. The a.c. thermal impedance Z_L is monitored by thermal inertia of second sound,

$$Z_L^{-1} = C_L\, u_{II}^2\, \frac{k}{s} \tag{11}$$

where C_L is the specific heat per unit volume of the liquid and u_{II} the second sound velocity. Carrying (11) into (6), we find

$$0 = \rho_L\, g + (\rho_S - \rho_L)\frac{s^2}{k}\left[1 + \left(\frac{dp_L}{dT}\right)^2 \frac{T/\rho_L}{u_{II}^2\, C_L + s\kappa_S}\right] - \frac{J_0\, s}{u_{II}^2\, C_L + s\kappa_S}\frac{dp_L}{dT} \tag{12}$$

J_0 is no longer equivalent to a mere change in effective gravity. For small s, we can expand equation (12) in a power series in s. The terms in s and s^3 are small. Neglecting them we get the dominant terms

$$\rho_L\, g + (\rho_S - \rho_L)\frac{s^2}{k}(1 + B) = 0 \tag{13}$$

where

$$B = \frac{T}{\rho_L}\left(\frac{dp_L}{dT}\right)^2 \frac{1}{C_L\, u_{II}^2} \tag{14}$$

B describes the « thermal inertia » due to second sound in the liquid. Its behaviour as a function of T is shown in figure 2 ; it is dominant at high temperatures.
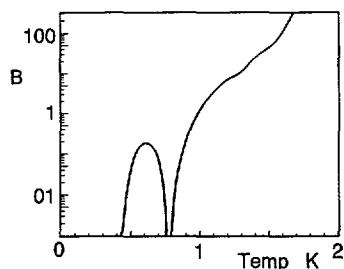

Fig. 2. — The parameter $B(T)$ for superfluid ^{4}He.

Equation (13) describes a purely oscillatory mode with s imaginary $(= i s_0)$. The terms which have been neglected describe the damping (or growth) of the mode. Putting $s = i s_0 + \delta s$ we get, after simple algebra,

$$\delta s = \left(\frac{dp_L}{dT}\right)\frac{(J_0 - J_c)}{u_{II}^2\, C_L} \tag{15}$$

where

$$J_c = \rho_L\, g(k)\left(\frac{dT}{dp_L}\right)\kappa_S \frac{B}{1 + B} \tag{16}$$

is the critical heat current. For J_0 larger than J_c the mode is unstable and it grows exponentially since the real part of s is positive. Equation (15) is very similar to the expression (10) obtained for a normal liquid with no second sound ; the main difference comes from the factor $B/(1 + B)$. But the nature of the instability is different : it occurs at finite frequency s_0 instead of zero frequency.

The instability appears first when $g(k)$ is minimal, i.e. at $k = 0$. We can readily calculate the critical temperature gradient using the data of Grilly [5] taken on the melting curve, and using estimates of second sound velocities and other quantities from the tables of Brooks and Donnelly [6]. The result is shown in figure 3. Below 0.78 K the latent heat is negative so that the critical temperature gradient is negative also. The largest magnitude for the temperature gradient is 0.93 mK/cm. Changing the temperature of the liquid by a few mK could have a destabilising effect if this were a complete description of the system. But we have ignored surface dissipation : we have ignored both the resistance to growth at the interface which leads to a difference in chemical potentials across the interface, and we have neglected the Kapitza resistance which gives rise to a discontinuity in the temperature across the interface.

At first sight this neglect appears to be correct for surface dissipation leads to terms proportional to s in the dispersion relation. For normal fluids these dissipative processes do not influence the threshold of instability, only the rate at which the instability develops. But for superfluid systems, s is mainly imaginary. The instability due to heat currents comes from a competition between terms in s and in s^3 in the dispersion relation. Consequently any term which is of order s can strongly affect the stability of the interface.
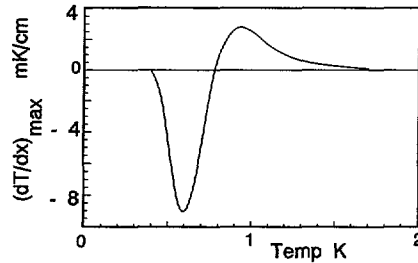

Fig. 3. — The critical temperature gradient for solid ^{4}He if there were no surface dissipation.

2. The effect of surface dissipation.

Let us first neglect Kapitza resistance. The thermal balance at interface is the same as before. But we allow for a resistance to growth : a finite velocity $\dot{\zeta}$ requires a chemical potential bias (per unit mass)

$$\mu_L - \mu_S = \Delta\mu = \frac{\dot{\zeta}}{\alpha} \tag{17}$$

α is a growth coefficient. (For ^{4}He, it is usually written as $m_L K_L$, where m_L is the atom mass : K_L then has the dimension of a velocity). (17) implies that the liquid pressure departs from the phase equilibrium curve $p_L(T)$, by an amount

$$\Delta p_L = \frac{\rho_S \rho_L}{\rho_S - \rho_L} \frac{\dot{\zeta}}{\alpha} \tag{18}$$

The dispersion relation (13) acquires an extra term

$$\frac{\rho_L \rho_S}{\rho_S - \rho_L} \frac{s}{\alpha} \tag{19}$$

which is dissipative and stabilizing. Such a contribution should be compared to the other dissipative terms in (6).

For *normal* materials, the dispersion relation reduces to (8). (19) competes with the bulk dissipation due to κ_S and κ_L. The ratio of surface to volume dissipation is equal to k/k^*, where

$$k^* = \frac{(\rho_S - \rho_L)^2}{\rho_S \rho_L^2} T \left(\frac{dp_L}{dT} \right)^2 \frac{\alpha}{\kappa_S + \kappa_L} \tag{20}$$

Under typical conditions, k^* is of atomic scale ($1/k^*$ if of order the phonon mean free path in either phase). On a macroscopic scale, surface dissipation is completely negligible as compared to bulk dissipation.

The situation is opposite for superfluid ^{4}He. Then (19) should be compared with the term

$$\frac{J_c s}{u_{II}^2 C_L} \frac{dp_L}{dT}$$

in (12). The ratio of surface to volume dissipation is

$$\left(\frac{dT}{dp_L} \right)^2 \frac{(\rho_S - \rho_L)}{\rho_S} \rho_L g \frac{B^2}{1 + B} \frac{\kappa_S}{T} \alpha \tag{21}$$

where B is defined in (14). Using values of α given by Bodensohn *et al.* [1], we find this ratio much larger than 1 at « high » temperatures (above 0.8 K) : it is about 10^5 at 0.8 K, 6×10^7 at 1.2 K and 4×10^9 at 1.6 K. The critical heat current and temperature gradient are accordingly pushed up by a factor $> 10^5$, up to 10 K/cm, a number beyond reach of any experiment. We conclude that surface dissipation is here largely dominant : it precludes any surface instability driven by J_0.

As the temperature goes down, the Kapitza resistance becomes significant : one must rely on coupled equations for the mass flow through the interface $\mathfrak{J} = \rho_S \dot{\zeta}$, and the energy flow $\mathfrak{J}_E$. These are conveniently written as [3]

$$\mathfrak{J} = \rho_S \alpha \left[\Delta\mu + \lambda \frac{\Delta T}{T} \right]$$
$$\Delta T = R \left[\bar{\mathfrak{J}}_E - \lambda \mathfrak{J} \right] \tag{22}$$

in which $\bar{\mathfrak{J}}_E = \mathfrak{J}_E - \mu \mathfrak{J}$ is the heat current, $\Delta\mu$, ΔT are discontinuities across the interface, R is the Kapitza resistance and λ is a cross coefficient that specifies how entropy is shared between solid and liquid. For a given pressure δp_L the interface temperature δT_L and δT_S should be calculated according to these new boundary conditions [3]. The result is equivalent to a shift of α, which becomes

$$\frac{1}{\bar{\alpha}} = \frac{1}{\alpha} + \frac{\rho_S}{T} \frac{RZ_L(TS_L - \lambda)^2 + RZ_S(TS_S - \lambda)^2 + Z_L Z_S \mathcal{L}^2}{R + Z_L + Z_S} \tag{23}$$

In the absence of surface dissipation ($R = 0$, $\alpha = \infty$),

$$\frac{1}{\bar{\alpha}_0} = \frac{\rho_S}{T} \frac{Z_L Z_S \mathcal{L}^2}{(Z_L + Z_S)}$$

which is just the bulk dissipation embodied in the first term in the bracket of (6). The genuine surface dissipation involves the excess to $\bar{\alpha}_0$, i.e. an effective α_{eff}.

$$\frac{1}{\alpha_{eff}} = \frac{1}{\alpha} + \frac{\rho_S R}{T(R + Z_L + Z_S)} \frac{[Z_L(TS_L - \lambda) + Z_S(TS_S - \lambda)]^2}{Z_L + Z_S}. \tag{24}$$

The thermal corrections to (24) become important below 0.6 K. But they are somewhat uncertain as the values of R and λ are not obvious. One possible choice is that of Andreev and Knizhnik [7], who assume an interface completely impenetrable to phonons — i.e. $\alpha = R = \infty$, $\lambda = 0$. In the long wavelength capillary limit, $s \simeq k^{3/2}$, $k \Rightarrow 0$, it seems more reasonable to assume

$$Z_L \ll R \ll Z_S$$

in which case

$$\frac{1}{\alpha_{eff}} = \frac{1}{\alpha} + \frac{\rho_S R}{T} (TS_S - \lambda)^2 \tag{25}$$

In any event, thermal corrections can only *enhance* surface dissipation : any instability is pushed up to very large heat currents, well out of reach of experiment.

3. The effect of a Grinfeld instability.

It was shown by Grinfeld [2, 3] that a non hydrostatic stress near the interface could drive a melting instability — a melting freezing wave relinquishing some of the bulk elastic energy. Assuming a one dimensional geometry in which the strain $u_{yy} = 0$, the instability is monitored by the difference $\sigma_{xx} - \sigma_{zz}$. It can be described as an additional term in $g(k)$, which becomes

$$g(k) = g + \frac{\gamma k^2}{\rho_S - \rho_L} - \frac{2\beta k}{\rho_S - \rho_L}$$
$$\beta = (\sigma_{xx} - \sigma_{zz})^2 \frac{1 - \sigma^2}{E} \tag{26}$$

(E is Young's modulus, σ is Poisson's ratio). While gravity and capillarity are stabilising, the elastic term leads to an instability if $g_{eff} \leq 0$.

In the absence of a heat current, the instability appears at the capillary wave vector

$$k_c = \left[\frac{(\rho_S - \rho_L) g}{\gamma} \right]^{1/2} \tag{27}$$

when β exceed a threshold

$$\beta_c^0 = [g \gamma (\rho_S - \rho_L)]^{1/2} \tag{28}$$

When β exceeds β_c^0, one faces the usual wavelength selection problem.

Assume now that a d.c. heat current J_0 is applied to a normal material (with finite κ_L, κ_S). g is replaced by

$$g \left[1 - \frac{J_0}{J_c} \right]$$

where J_c is given by (10). The thermal and elastic instabilities compete : the corresponding phase diagram is shown in figure 4. Note that the wave vector at threshold changes as J_0 increases,

$$k_c = \left[\frac{(\rho_S - \rho_L) g}{\gamma} \left(1 - \frac{J_0}{J_c} \right) \right]^{1/2} = \frac{\beta_c}{\gamma} \tag{29}$$
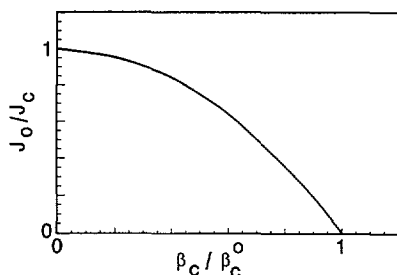

Fig. 4. — The stability diagram for a normal liquid as a function of heat current and uniaxial strain, denoted by the parameter β given by equation (26).

As J_0 increases, the instability moves to large wavelength. Conversely, one could apply a heat current in the reverse direction and cause a decrease in critical wavelength, enabling more waves to appear across a sample.

Such a simple picture no longer holds for ^{4}He, since J_0 only acts on damping of the wave. The Grinfeld instability is not affected by a d.c. heat flow ; it always appears at the capillary wavelength (27).

We now examine how these conclusions apply to the experiments of Bodensohn *et al.* [1]. They applied a sudden decrease in temperature to the liquid, thereby setting up a heat flow, but also a non hydrostatic stress in the solid. The difference $(\sigma_{xx} - \sigma_{zz})$ arises partly from thermal expansion in the x and y directions (in a vessel which we assume free of expansion), partly from the change in liquid pressure p_L needed in order to remain on the phase equilibrium curve. Mechanical equilibrium implies $\delta \sigma_{zz} = - \delta p_L$, leading to a non hydrostatic stress if we impose the constraint $\delta u_{xx} = \delta u_{yy} = 0$. More precisely, we may write

$$d\sigma_{xx} = \frac{- E \alpha_S \, dT + \sigma \, d\sigma_{zz}}{1 - \sigma}$$

where α_s is the thermal expansion coefficient. We thus find

$$d(\sigma_{zz} - \sigma_{xx}) = \frac{E \alpha_S \, dT - (1 - 2\sigma) \, dp_L}{1 - \sigma} = - \frac{(1 - 2\sigma)}{(1 - \sigma)} \, dp_L (1 - g_1) \,. \tag{30}$$

Here we have introduced the dimensionless parameter $g_1 = E \alpha_S (dT/dp_L)/(1 - 2\sigma)$ which expresses the relative importance of thermal expansion. For typical materials g_1 is small, but
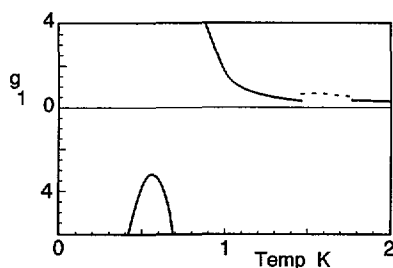
Fig. 5. — The variation of g_1 with temperature.

for helium it is of order unity for temperatures about 1 K. The variation of g_1 with temperature is shown in figure 5. Here we have used the estimates given by Balibar *et al.* [4] of $E = 3.05 \times 10^8$ ergs/cm^2 and $\sigma = 0.33$.

The threshold for the Grinfeld instability is therefore a pressure change, dp_L, given by Balibar *et al.* [4] :

$$dp_L^2 = \frac{E(1 - \sigma) \sqrt{(\rho_S - \rho_L) g \gamma}}{(1 + \sigma)(1 - 2\sigma)^2 (1 - g_1)^2} .\qquad(31)$$
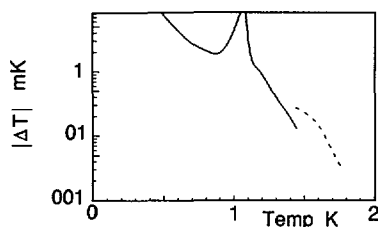

Fig. 6. — The temperature change needed to induce the Grinfeld instability.

We can generate the pressure change either mechanically or through a small change in temperature, ΔT. Provided we work away from the region where $g_1 = 1$ we can take ΔT to be small. In this way we find the variation in ΔT shown in figure 6. Above 1.1 K it is fairly easy to generate the Grinfeld instability by changing the temperature.

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