# The Growth Kinetics of <sup>3</sup>He Crystals

F. Graner,\* R. M. Bowley,† and P. Nozières

Institut Laue-Langevin, Grenoble Cedex, France

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Near the minimum of the <sup>3</sup>He melting curve, the dynamics of the liquid-solid interface is limited by surface dissipation instead of bulk diffusion of latent heat. Three dissipative coefficients characterize the intrinsic properties of the interface: the mobility k, the Kapitza resistance  $R_K$ , and the latent heat sharing coefficient  $\lambda$ . These parameters are studied at T = 0.32 kelvins, and at 29.3 bar, and the theoretical predictions are compared to recent measurements.

## **1. INTRODUCTION**

Usually crystal growth is limited by bulk diffusion of latent heat or impurities. In contrast, helium-3 or helium-4 can be free of impurities, and the solid-liquid latent heat L may vanish. Even when nonzero, the latent heat L is evacuated very quickly due to the good thermal conductivities present: in the solid for both <sup>3</sup>He and <sup>4</sup>He and in the superfluid for <sup>4</sup>He (or for <sup>3</sup>He at lower temperatures). Crystal growth is then limited only by surface dissipation, which can be experimentally studied.

The elementary excitations in each phase determine both the surface dissipation<sup>1</sup> and the transport properties. We discuss the kinetic coefficients around the minimum of the <sup>3</sup>He melting curve ( $T_m = 0.32$  K and P = 29.3 bar) before comparing them to experimental results. We focus on the Kapitza resistance  $R_K$ , for which we have developed a detailed calculation.

In the second section we present the notation used to describe the interface.<sup>2</sup> In Sect. 3, we interpret recent experiments<sup>3</sup> and deduce a measurement of the Kapitza resistance at  $T_m$ .  $R_K$  is measured by its effect on growth dynamics and not as the usual ratio of a temperature jump by a heat flux. We present our predictions at low temperature in Sect. 4 and compare them with the experiments in Sect. 5. The details of our calculations will be found in the appendices.

\*Permanent address: LPS-ENS, 24 rue Lhomond, 75231 Paris Cedex 05, France. †Permanent address: Department of Physics, University of Nottingham, NG7 2RD, U.K.

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#### 2. DESCRIPTION OF A MOVING INTERFACE

When a pure crystal is grown from its melt, the liquid-solid interface moving with a velocity v is crossed by two coupled currents: a flux mass  $J = \rho_S v$ , and an energy flux  $J_E$ . They are driven by the discontinuities of chemical potential  $\Delta \mu = \mu_L - \mu_S$  and temperature  $\Delta T = T_L - T_S$  between both phases. When rough, the interface responds linearly to small departures from the thermodynamical equilibrium; the fluxes are linked to the "forces" by linear Onsager relations<sup>2,4</sup>

$$\begin{pmatrix} J \\ J_Q \end{pmatrix} = \begin{pmatrix} A & B \\ B & C \end{pmatrix} \begin{pmatrix} \Delta \mu / T \\ \Delta T / T^2 \end{pmatrix}$$
(1)

Here  $J_Q$  is the heat flux  $J_E - \mu J$ ; in each phase it is the sum of a conduction current and a convection term TSJ

$$J_Q = J_E - \mu J = J_{QL} + TS_L J = J_{QS} + TS_S J$$
(2)

Expression (1) takes an equivalent but more useful form

$$\Delta T = R_K (J_O - \lambda J) \tag{3}$$

$$J = \rho_S k (\Delta \mu + \lambda \Delta T / T) \tag{4}$$

The coefficient k is the isothermal mobility of the interface: when  $\Delta T = 0$ , we have  $v = k\Delta\mu$ . The thermal Kapitza resistance  $R_K$  is defined in the static case (v = 0) by the usual relation  $\Delta T = R_K J_Q$ . The crossed term  $\lambda$  describes the sharing of the latent heat creation between both phases; for instance, in the isothermal case where  $\Delta T = 0$  and  $J_Q = \lambda J$ , the conduction currents on each side are  $J_{QS} = J(\lambda - TS_S)$  and  $J_{QL} = J(\lambda - TS_L)$ :  $\lambda$  determines separately  $J_{QS}$  and  $J_{QL}$ , while their difference  $J_{QS} - J_{QL} = JL$  is fixed by the production rate of the latent heat.

The motion of the interface is often started by a pressure variation. It is then convenient to distinguish in the "force"  $\Delta \mu$  a driving mechanical term  $\Delta \mu_M$  and a thermal brake  $\Delta \mu_T$ . The Gibbs-Duhem relation yields

$$\Delta \mu = \Delta \mu_M + \Delta \mu_T = \left(\frac{\delta P_L}{\rho_L} - \frac{\delta P_S}{\rho_S}\right) - \left(S_L \delta T_L - S_S \delta T_S\right)$$
(5)

where  $\delta P = P - P_{eq}(T)$  and  $\delta T$  are small fluctuations around the equilibrium values for a flat interface;  $\rho_L$  and  $\rho_S$  are the densities;  $\Delta \mu$ ,  $S_L$  and  $S_S$  are referred to the unit of mass. In the experiments discussed hereafter,  $\Delta \mu_T$  cannot be measured; in practice we eliminate the thermal terms and introduce an effective mobility of the interface by

$$v = k_{\rm eff} \Delta \mu_M \tag{6}$$

where  $k_{\rm eff}$  includes all information concerning heat creation and evacuation. In the linear regime, and as long as the phonons are in hydrodynamical regime,<sup>4</sup> heat conduction in each phase can be characterized by a bulk thermal impedance. If the fluxes are oriented positively from liquid to solid, the impedances  $\zeta$  are defined by

$$\delta T_S = +\zeta_S J_{QS}; \qquad \delta T_L = -\zeta_L J_{QL} \tag{7}$$

The thermal circuit has then a simple electrical equivalent: the bulk impedances  $\zeta_L$ ,  $\zeta_S$  are shunted by the resistance  $R_K$  of the interface. The latent heat creation injects a flux  $J(TS_L - \lambda)$  on the liquid side and  $-J(TS_S - \lambda)$ in the solid (Fig. 1). The conduction current crossing the interface is  $(\delta T_L - \delta T_S)/R_K$ , and we have

$$J(TS_L - \lambda) = \frac{\delta T_L}{\zeta_L} + \frac{\delta T_L - \delta T_S}{R_K}$$
(8a)

$$J(\lambda - TS_S) = \frac{\delta T_S}{\zeta_S} + \frac{\delta T_S - \delta T_L}{R_K}$$
(8b)

or equivalently

$$\delta T_L = J\zeta_L \frac{\zeta_S L + R_K (TS_L - \lambda)}{R_K + \zeta_L + \zeta_S}$$
(9a)



Fig. 1. The electrical analogy of the thermal circuit. The latent heat creation is shared between the two phases and debits in the bulk thermal impedances  $\zeta_L$ ,  $\zeta_S$  shunted by  $R_K$ . The currents are oriented positively from the liquid towards the solid.

$$\delta T_{S} = J\zeta_{S} \frac{\zeta_{S}L + R_{K}(\lambda - TS_{S})}{R_{K} + \zeta_{L} + \zeta_{S}}$$
(9b)

Equation (4), after elimination of  $\delta T_L$  and  $\delta T_S$ , allows us to calculate the effective mobility  $k_{\text{eff}}$ . It corrects the intrinsic mobility k by a combination of the Onsager coefficients and bulk thermal impedances, describing the dissipative effects due to latent heat evacuation

$$\frac{1}{k_{\text{eff}}} = \frac{1}{k} + \frac{\rho_S}{T} \frac{R_K \zeta_L \zeta_S}{R_K + \zeta_L + \zeta_S} \left[ \frac{(TS_L - \lambda)^2}{\zeta_S} + \frac{(TS_S - \lambda)^2}{\zeta_L} + \frac{L^2}{R_K} \right]$$
(10)

We see that the thermal term slows down the growth  $(k_{\text{eff}} < k)$  whatever the sign of L or  $\Delta \mu_M$ .

In usual crystal-growth experiments, thermal conductivities are finite and  $R_{\kappa}$ , which decreases as  $T^{-3}$ , is negligible compared to  $\zeta_L$  and  $\zeta_S$ . Bulk dissipation is then dominant. Since the second term in Eq. (10) is greater than 1/k, the surface dissipation is completely hidden; discontinuities of temperature or chemical potential across the interface are negligible compared to the bulk gradients. In <sup>3</sup>He or <sup>4</sup>He the thermal impedances and L can be made sufficiently small, and the properties of the interface itself become open to experiment.

# 3. EXPERIMENTS ON THE <sup>3</sup>He LIQUID-<sup>3</sup>He SOLID INTERFACE

# 3.1. Excitations in Both Phases

The thermal excitations in <sup>3</sup>He solid are longitudinal and transverse phonons as well as spin degrees of freedom, but we will see that the latter are frozen on the relevant time scales. There is thus no essential difference with the case of <sup>4</sup>He. In contrast, <sup>3</sup>He liquid presents a "thermal bath" of quasi-particles that have a much greater thermal capacity than phonons and rotons of the superfluid <sup>4</sup>He. The kinetics of the interface, i.e., the mobility as well as heat transmission, is hence deeply modified (Table I).

Timeta		
Liquid	$u = 35 \text{ m} \cos^{-1}$	Dof 5
quasiparticles. $m/m = 5.55$ at 0 K	$v_F = 35 \text{ m sec}$	Rel. J
first sound	$c_1 = 402 \text{ m sec}^{-1}$	Ref. 6
longitudinal zero sound	$c_{ol} = 404.5 \text{ m sec}^{-1}$	<b>Re</b> f. 6
transverse zero sound	$c_{ot} \ge v_F$	Ref. 7
Solid		
longitudinal sound	380 to 480 m sec <sup><math>-1</math></sup>	<b>Ref.</b> 8
transverse sound	$120 \text{ to } 240 \text{ m sec}^{-1}$	Ref. 8

 TABLE I

 Velocities of Excitations under 29.3 bar

In the temperature range between 2.5 mK and a small fraction of one kelvin, liquid <sup>3</sup>He is a Fermi liquid described by the Landau theory. <sup>9-11</sup> It supports individual excitations, namely the quasiparticles with momentum close to the Fermi momentum  $p_F$  and also collective sound excitations. At a frequency  $\omega$  smaller than the collision frequency in the liquid ( $\omega \tau_{coll} \ll 1$ ) we enter the hydrodynamic regime, and the liquid has the usual viscous behavior. A longitudinal wave propagates, while a transverse shear wave is overdamped. But if  $\omega \tau_{coll} \gg 1$  (high frequency or low temperature), the collective modes are "zero sound"-like. Beside the longitudinal mode,<sup>6</sup> there appears a transverse mode,<sup>7</sup> which means that the liquid has an "elastic" behavior. The longitudinal zero sound propagates at a velocity slightly greater than first (normal) sound; transverse zero sound travels at a velocity of order the Fermi velocity  $v_F$ .<sup>12,13</sup>

#### 3.2. The Cross Coefficient $\lambda$

The Onsager cross coefficient  $\lambda$  determines the partition of the latent heat between both phases: a heat flux  $TS_L - \lambda$  is created at the surface on the liquid side, and  $\lambda - TS_S$  on the solid side.\* At a microscopic level, this heat creation is due to inelastic processes occuring when an atom sticks to the solid or, inversely, is emitted from the solid into the liquid. If we ignore at first the acoustic modes (phonons) and the magnetic exchange in the solid, an atom in the solid has a well-defined enthalpy  $\varepsilon_s$ . When it gets into the liquid where it has an enthalpy  $\varepsilon$ , the difference  $(\varepsilon - \varepsilon_S) \sim T$  has to be provided by the surrounding thermal excitations. The only remaining degrees of freedom are those of quasiparticles in the liquid. The elementary process appears then as an interaction mechanism where an atom from the liquid sticks to the solid while another one removes the excess of enthalpy. As a result, the melting enthalpy is released on the liquid side, which implies  $\lambda = TS_s$ , as has been suggested by Puech, Bonfait, and Castaing (PBC).<sup>14</sup>

Of course, the solid does have internal degrees of freedom that could contribute to inelastic processes and energy conservation upon sticking. The spin degrees of freedom control the magnetic entropy  $k_B \log 2$ , but the corresponding exchange energy is far too small to provide an efficient evacuation mechanism, typically 1.25 mK for a pair of spins, compared to thermal energies ~0.3 K. The solid phonons are not efficient either in view of their long wavelengths: they are very weakly coupled to the *localized* sticking process, in contrast to liquid quasiparticles that can be localized into wave packets of size  $\sim k_F^{-1}$ . Besides, phonons have a small specific heat compared to that of quasiparticles. Such a qualitative argument can be

<sup>\*</sup>This distinction between both sides of the interface would not make sense if the Kapitza resistance were negligible compared to the bulk thermal impedances.

made more precise by calculating the acoustic energy radiated in the solid when an atom sticks to (or bounces on) the surface. This calculation is carried out in Appendix A, yielding a radiated energy

$$E_{\rm rad} = \frac{1}{3} \frac{p_{\rm F}^2}{2m} \left(\frac{T}{\Theta_{\rm D}}\right)^3 \tag{11}$$

where  $\Theta_D$  is the Debye temperature of the solid. The factor  $T^3$  is a statement of the mismatch mentioned above. Such a mechanism cannot dispose of an inelastic energy  $\sim T$ .

# 3.3. Experimental Measurement of k and $R_K$

During recent experiments reported elsewhere,<sup>3</sup> very pure <sup>3</sup>He crystals have been grown from their melt in an optical cryostat. A monocrystal is observed during its relaxation towards thermodynamic equilibrium. Its shape evolves by local melting and recrystallization under the effect of gravity (since the solid is slightly more dense than the liquid) and the surface tension  $\gamma$ . The size of each crystal is between 1 and 3 mm, and the relaxation takes 10 to 30 sec. The windows of the cryostat allow one to measure visually the speed of the interface. Moreover, the applied departure from mechanical equilibrium  $\Delta \mu_M$  is controlled only by gravity and capillary forces; it is also possible to measure it visually, by determining the heights and curvatures of various points of the interface. The effective mobility  $k_{\text{eff}}$  (Eq. 6) becomes directly measurable.

Experiments are done around the minimum of the melting curve P(T); the latent heat L, which is proportional to dP/dT, vanishes and becomes negative at  $T_m = 0.32$  K. By choosing the temperature, one can control L as a variable parameter. It is then possible to determine  $k_{\text{eff}}$  as a function of L and to compare its behaviour with theoretical predictions (Eq. 10).

For experiments performed close to the minimum of the <sup>3</sup>He melting curve, some approximations simplify the expression for  $k_{\text{eff}}$ .

The liquid has a very poor thermal conductivity<sup>15</sup>:  $4.5 \times 10^{-3}$  W m<sup>-1</sup> K<sup>-1</sup>, compared to more than 16 W m<sup>-1</sup> K<sup>-1</sup> for the solid.<sup>14,16</sup> The heat diffuses at a rate of  $4.6 \times 10^{-8}$  m<sup>2</sup> sec<sup>-1</sup>: on a time scale of 10 sec, the boundary layer is ~1 mm wide, i.e., about the size of the cell. The resistive (real) and capacitive (imaginary) parts of the thermal impedance  $\zeta_L$  are then both of order of 0.22 m<sup>2</sup> K W<sup>-1</sup>. The bulk thermal impedance of the liquid is large compared to the Kapitza resistance.

In the solid, the phonon mean free path is limited by reflections on the surface and on the copper walls.<sup>16</sup> One can only define, for the distribution of phonons present in the crystal, a *global* temperature  $T_s$ . The phonons come from the liquid, which is thermally inhomogenous; but after multiple reflections their distribution becomes isotropic:  $T_S$  represents the physical temperature that controls the growth kinetics. It is then possible to define a bulk thermal impedance  $\zeta_S$ , with a zero real (resistive) part. The capacitive part is due to the variation of the crystal mass, but the quantity of crystal remains almost constant during the experiments, so that here  $\text{Im}(\zeta_S)$  is also negligible.

We obtain then  $|\zeta_L| \gg R_K \gg |\zeta_S|$ . Another simplification is that the difference  $(TS_S - \lambda)$  can always be neglected\* in Eq. (10), which eventually becomes

$$\frac{1}{k_{\rm eff}} = \frac{1}{k} + \frac{\rho_S}{T_m} R_K L^2 = \frac{1}{k} + 3 \times 10^8 R_K (T - T_m)^2$$
(12)

(units: k in sec m<sup>-1</sup>;  $R_K$  in m<sup>2</sup> K W<sup>-1</sup>; T in K). The physical situation is clear (Fig. 2). The latent heat released by the growth is created on the liquid side, mostly in the quasiparticles bath. It eventually transfers to collective modes, which provide a channel to transmit it back to the solid. The limiting factor is the Kapitza resistance which must be crossed in order to ensure evacuation. In the melting area, the situation is reversed: heat crosses again the interface and is absorbed in the liquid.

Experimentally,<sup>3</sup> the growth rate  $k_{\rm eff}$  reaches a maximum when the latent heat vanishes, as expected; namely  $k = 0.18 \pm 0.04 \sec m^{-1}$ . The position of the maximum at  $T = T_m$ , with an uncertainty of 10 mK, confirms that  $|\lambda/TS_s - 1|$  is certainly less than  $1.5 \times 10^{-2}$ . The curvature of  $1/k_{\rm eff}(T)$  yields  $R_{\kappa}$ ; the major uncertainty comes from the measured value of the surface tension<sup>18</sup>  $\gamma = 0.060 \pm 0.011 \, {\rm erg} \, {\rm cm}^{-2}$ . One finally obtains the following experimental value at 0.32 K:

$$R_K = 1.3 \pm 0.3 \text{ cm}^2 \text{ K W}^{-1}$$
 [i.e.,  $R_K T^3 = 0.043 \pm 0.01 \text{ cm}^2 \text{ K}^4 \text{ W}^{-1}$ ] (13)

whereas PBC<sup>14</sup> measured  $R_K T^3 = 0.8 \text{ cm}^2 \text{ K}^4 \text{ W}^{-1}$ . Their measurement is more direct, but more open to experimental complications: this large discrepancy is currently under examination (Castaing and Bossy, private communication).

Here  $R_{\kappa}$  is indirectly measured by its effect on the growth kinetics and not by any heat flux or temperature measurement. This is possible here because  $R_{\kappa}$  is not hidden by the bulk thermal impedances. If part of the latent heat were released in the solid, it would be evacuated without crossing  $R_{\kappa}$ ; the same effect would appear if  $R_{\kappa}$  were much less, or even much

<sup>\*</sup>When L is nonzero,  $(TS_S - \lambda) \ll L$ : indeed, with  $L = 900 \ (T - T_m) \ J \ kg^{-1}$  (Ref. 17) and  $TS_S \sim k_B T \ln 2/m = 615 \ J \ kg^{-1}$ , we see that  $|1 - \lambda/TS_S| < 6 \times 10^{-5}$  is smaller than  $L/TS_S = 1.5 \ (T - T_m)$  as soon as  $|T - T_m|$  exceeds 0.04 mK. And when L is zero, at  $T = T_m$ , the correction  $\rho_S R_K (TS_S - \lambda)^2/T$  is less than  $10^{-4} \ m \ sec^{-1}$ , i.e. negligible compared to the intrinsic mobility term 1/k.



Fig. 2. This egg-shaped <sup>3</sup>He crystal is 2.6 mm wide. Its shape relaxes, driven by gravity and surface tension. Within ~30 sec, the top melts while the bottom grows, until the equilibrium meniscus is reached. The thermal circuit of Fig. 1 is much simplified in our experiments: here  $\lambda = TS_S$  and  $|\zeta_L| \gg R_K \gg |\zeta_S|$ . The latent heat is created within the liquid in the growing area, crosses the interface, and is evacuated through the solid. The melting area acts to absorb the latent heat. The temperatures are indicated on the upper part of the figure, the heat flux in the lower part; here  $\delta T_L = R_K JL$ .

greater, than both  $\zeta_L$  and  $\zeta_S$ . In these cases  $R_K$  would play a negligible role in the dissipation.

# 4. THEORETICAL STUDY OF $R_{K}$

We will now present a calculation of the Kapitza resistance based on an acoustic mismatch method, stressing the importance of transverse zero sound in heat transmission.

The helium-3 solid-liquid interface is free of impurities and in a well-defined surface state; the acoustic impedances of both phases are comparable. It is impossible here to use semiclassical or fully quantummechanical treatments<sup>19</sup> based on perturbation developments in  $v_F/c_{sound}$ , as this number is not small for the transverse sound in the liquid. We use the acoustic mismatch model, modified to take into account the strong attenuation of these transverse modes. The arguments leading to this choice are reviewed in detail by Swartz and Pohl.<sup>20</sup>

The heat flux  $\Phi_{S \to L}$  from solid to liquid is determined by the flux of phonons incident on the interface on the solid side and by their transmission rate; at thermal equilibrium it is exactly compensated by an opposite flux  $\Phi_{L \to S}$  coming from the liquid. The acoustic mismatch model<sup>21-23</sup> relies on the assumption that  $\Phi_{S \to L}$  is a function of  $T_S$  only and not of  $T_L$ . Then, near equilibrium, with  $T_S = T_L + \Delta T$ , the resulting heat flux is

$$\Phi_{S \to L}(T_S) - \Phi_{L \to S}(T_L) = \Delta T \frac{\partial \Phi_{S \to L}}{\partial T}$$
(14)

The Kapitza resistance is determined if we know the function  $\Phi_{s \rightarrow L}(T)$ . As the incident heat flux depends only on the phonon distribution in the solid, the problem consists in calculating the energy transmission rate for one phonon.

When a phonon of the solid (longitudinal or transverse) hits the interface, the surface of the liquid is submitted to a pressure and velocity wave characterized by two parameters: its frequency  $\omega$  and its wavelength q in the plane of the surface (noted yz). Boundary conditions on the liquid side relate the stress tensor  $\sigma_{ij}$  to the liquid velocity v: we can define one acoustic impedance matrix  $[Z^L]$  for each mode of incident phonon according to

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{xy} \\ \sigma_{xz} \end{pmatrix} = -[Z^{L}(\omega, q)] \begin{pmatrix} v_{x} \\ v_{y} \\ v_{z} \end{pmatrix}$$
(15)

In the solid, acoustic impedances  $[Z^s]$  are defined in the same way. The knowledge of the acoustic impedances of both phases is enough to determine the transmission coefficient for phonons and to compute the Kapitza resistance (see Appendix B).

In the solid, such a "surface acoustic impedance"  $[Z^s]$  is just the projection onto the surface of the usual bulk impedance. When diagonalized, the latter reduces to one quasilongitudinal and two quasitransverse scalars respectively equal to  $\rho_s c_i$ , where  $c_i$  are the sound velocities. The solid has the phonons of an usual bcc crystal.<sup>8</sup> We do take into account the distribution of phonon velocities, but we average over the anisotropy of the crystal when calculating the transmission.

In the liquid, the surface impedance may in principle differ from the projected bulk impedance, due to distortion of the quasiparticle distribution near the walls, extending over a mean free path. We neglect this effect: we assume that the liquid response is controlled by the bulk impedance. The latter can be reduced to a longitudinal component  $Z_i^L$  and two transverse components which, due to the isotropy of the liquid, are equal: we call them  $Z_i^L$ . Let us now focus on the impedances of the liquid.

The longitudinal impedance  $Z_I^L$  is essentially due to the sound modes.<sup>24</sup> Indeed, the quasiparticles are strongly coupled and the longitudinal sound modes are well separated from the individual Fermi excitations continuum (c is much greater than  $v_F$ ).<sup>6,12,13</sup> Because the density and the compressibility of both phases are almost equal at the minimum of the melting curve, the acoustic mismatch is small. The phonon-phonon coupling then ensures a good heat transmission<sup>19</sup> from solid to liquid. Within the bulk of the liquid, the coupling between collective and individual excitations is also good (see discussion section), so that the heat eventually reaches the quasiparticle bath where it is stored. Conversely, the direct coupling at the interface between photons of the solid and quasiparticles is much weaker (see above and Ref. 23).

However, the longitudinal modes cannot account for all the heat transmission. The transverse zero-sound modes are 10 times slower. Their transmission rate is smaller, due to a poorer acoustic mismatch; but they contribute more to the phonon specific heat (which varies as  $1/c^3$ ) and hence to the heat flux (which varies as  $1/c^2$ ). A simple estimate which neglects this new transmission channel<sup>14</sup> leads to  $R_K T^3 \sim 0.2 \text{ cm}^2 \text{ K}^4 \text{ W}^{-1}$ : this value is five times larger than the experimental result. The transverse modes thus play a crucial role (Fig. 3). We need to know  $Z_t^L$  along the melting curve for thermal frequencies.

At low temperatures (<50 mK), the thermal phonons are in the zerosound regime and correctly described by Landau theory. But in contrast with longitudinal zero-sound,  $c_{ot}$  is very close to  $v_F$  and  $Z_t^L$  doesn't reduce



Fig. 3. The transmission rate  $\tau$  of phonons coming from the solid. The incident phonons are either: l = longitudinal, t1 =transverse vibrating in the incidence plane, t2 = transverse vibrating perpendicularly to the incidence plane. Here  $\tau \cos \theta \sin \theta$  is plotted vs. the incidence angle  $\theta$ , so that the relative importance of a mode is represented by the integral of the curve. (a) At high temperatures, without transverse modes in the liquid, the t2 mode is not transmitted at all. (b) At 0.32 K, with transverse modes in the liquid, the transmission is significantly enhanced.

to  $\rho_L c_{ol}$ . The whole spectrum of excitations contribute to  $Z_t^L$ , including the continuum of individual modes. A complete calculation should be based on a linear response function formalism.<sup>10</sup> Experimental measurements<sup>7</sup> yield a global value\* of  $Z_t^L$ . One can parametrize  $Z_t^L$  by defining an "effective sound velocity"  $c_{\text{eff}}$  according to

$$Z_t^L \approx \rho_L c_{\rm eff} \tag{16}$$

The extrapolated value at melting pressure is  $c_{\text{eff}} \approx 23.4 \text{ m/sec}$ . Note that  $c_{\text{eff}}$  is below  $v_F$ , expressing the contribution of the quasiparticle continuum. If we use this value, the phonon transmission rate is independent of T, and

<sup>\*</sup>Predictions from the Landau theory (Ref. 25) disagree with its frequency dependence. For a review of this problem, see for instance Ref. 12.

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 $R_K T^3$  remains constant. Our numerical calculations yield

$$R_K T^3 = 0.03 \text{ cm}^2 \text{ K}^4 \text{W}^{-1}$$
 (T < 50 mK) (17)

At higher temperatures (especially at 0.32 K) the transmission should be worse, since the transverse modes disappear in the first-sound regime:  $R_K T^3$  increases slightly with T. But a difficulty arises in the determination of  $Z_t^L$  as a function of T. This frequency and temperature range (10<sup>10</sup> Hz, 0.3 K) is both outside the validity domain of Landau's theory and yet inaccessible to direct measurements.

At still higher temperatures, we enter the hydrodynamic regime in which  $Z_i^L$  is monitored by the liquid viscosity  $\eta \sim T^{-2}$ 

$$\left(\frac{\rho_L}{Z_\iota}\right)^2 = i\frac{\rho_L}{\eta\omega} \tag{18}$$



Fig. 4.  $R_K T^3$  is computed at 0.32 kelvins as a function of  $c_{\text{eff}}$  in a range 5 to 50 m/sec (see text). Units:  $c_{\text{eff}}$  in m sec<sup>-1</sup>,  $R_K T^3$  in cm<sup>2</sup> K<sup>4</sup> W<sup>-1</sup>.

The best we can do is to interpolate between the two limits, using a "viscoelastic" model<sup>26</sup> in which

$$\left(\frac{\rho_L}{Z_t^L}\right)^2 = \frac{1}{c_{\text{eff}}^2} + i\frac{\rho_L}{\eta\omega}$$
(19)

The real (imaginary) part characterizes the propagation (attenuation) of an excitation.  $c_{\text{eff}}$  is then a parameter which follows from a fit to the experimental  $R_K$  (see Fig. 4 and Appendix B).

In principle, we could also replace  $\eta$  by an effective  $\eta_{\text{eff}}(\omega)$ , which would go to  $\eta$  as  $\omega$  decreases: such a refinement is useless at 0.32 K as in practice the result does not depend much on  $\eta_{\text{eff}}$ . It seems that  $\eta$  has never measured exactly at the minimum of the melting curve, but extrapolation of measurements at different temperatures<sup>27</sup> and pressures<sup>28</sup> indicates the same value as predictions deduced from  $Z_t^{L7}$  or from Landau theory,<sup>12</sup> namely

$$\eta \sim 4 \times 10^{-5}$$
 Poise (at 0.32 K and 29.3 bar). (20)

### 5. DISCUSSION AT 0.32 K

The experimental value of  $R_K$  agrees with our prediction.  $R_K T^3$  at 0.32 K is higher than our predictions at lower temperatures, which corresponds to the transition range between the first- and zero-sound regimes. Then Fig. 4 yields the "effective transverse sound velocity"  $c_{\text{eff}}$  at the minimum of the melting curve and at thermal frequencies (10<sup>10</sup> Hz)

$$10 \,\mathrm{m \, sec^{-1}} < c_{\rm eff} < 20 \,\mathrm{m \, sec^{-1}} \tag{21}$$

in good agreement with the value deduced from lower frequencies data<sup>7</sup>:  $c_{\text{eff}} = 15 \text{ m sec}^{-1}$  at 10<sup>8</sup> Hz.

The major heat transmission channel across the interface is the coupling between sound modes in both phases. Harrison<sup>23</sup> reviewed the question of thermalization in each phases, applying Khalatnikov's method.<sup>21</sup> In the liquid, the heat is essentially stored in the quasiparticle bath. The bulk thermal resistance between the phonons of the liquid and the quasiparticles for a volume V is

$$R_{\Phi L-QP}V = \frac{15}{4\pi^2} \frac{\hbar^3}{k_B^4} \frac{c^2}{\alpha T^3}$$
(22)

The longitudinal sound attenuation is  $\alpha \sim 2 \times 10^6 \text{ m}^{-1}$  at the minimum of the melting curve<sup>6,12</sup>; i.e., for a typical thickness of 1 mm the resistance per surface A is

$$R_{\phi L-OP}A \approx 2 \times 10^{-4} \text{ cm}^2 \text{ K W}^{-1}$$
 (23)

at  $T_m = 0.32$  K. This resistance is then negligible compared to  $R_K$ . Note that  $\alpha$  varies as  $T^2$ , which means that  $R_{\Phi L-QP}T^3$  increases quickly at low temperatures. As for the transverse sound in the liquid, it is slower and much more attenuated, hence even better thermalized with the quasiparticles.

Conversely, in the solid, a similar evaluation yields the bulk resistance between the phonons and the spin-exchange bath

$$\frac{1}{R_{\phi S-sp}} = \frac{C_{\rm spin}}{t} \tag{24}$$

Here t is the relaxation time of the spins in the lattice,  $C_{\rm spin}$  is the specific heat of the spins. Numerically, at the minimum of the melting curve,  $t \sim 1 \sec^{29}$ ;  $C_{\rm spin}/R = 4.6 \times 10^{-5}$ , i.e.,  $C_{\rm spin} = 1.5 \times 10^{-5}$  J K<sup>-1</sup> cm<sup>-3</sup> per unit volume. For a typical thickness of 1 mm and an area A, the resistance is

$$R_{\Phi S-sp}A \sim 10^6 \,\mathrm{cm}^2 \,\mathrm{K} \,\mathrm{W}^{-1}$$
 (25)

This resistance is much bigger than  $R_K$ : the spins are decoupled from the heat-transmission processes across the interface.

## 6. CONCLUSION

We presented here a coherent description of the dissipative coefficients that control the growth kinetics of the <sup>3</sup>He solid-liquid interface. Predictions based on the study of the elementary excitations in both phases are compared to experimental results at 0.32 K.

- (1) The coupling between heat and matter fluxes is described by the cross coefficient  $\lambda$ . The latent heat is released in the quasiparticles of the liquid, except for a negligible part in the phonons of both phases.
- (2) The heat has then to cross the Kapitza resistance of the interface, before being evacuated through the solid. The value of  $R_K$  is predicted from the phonon transmission; it is weaker than previously expected, as the transverse modes which exist in the liquid offer an essential transmission channel. It is in agreement with our unconventional measurement of  $R_K$  at  $T_m$ .
- (3) We predict that the variation of  $R_{\kappa}$  with T is near from the classical law  $R_{\kappa}T^3$  = constant; the transmission should be slightly improved at lower temperature, due to an increase of the importance of transverse zero sound. However, more direct measurements of  $R_{\kappa}$ should be useful to check our indirect value, as well as our predictions at lower temperatures.

# APPENDIX A: STUDY OF $TS_s - \lambda$

We will determine the amount of energy radiated in the phonons bath by an atom of the liquid sticking (or bouncing) on the crystal. This elasticity calculation requires us first to calculate the response of the solid to an external constraint, i.e. its acoustic impedance.

#### A.1. Acoustic Impedance of the Solid

Let  $\sigma_{xx}$ ,  $\sigma_{xy}$  be stresses applied to the surface yz of the solid, with a frequency  $\omega$  and a wavevector **q** in the y direction (Fig. 5). The surface responds by moving at a velocity  $(\dot{u}_x, \dot{u}_y)$ : this velocity is linearly related to the stresses. As a percussion creates stresses only in the incidence plane, we don't consider here any  $\sigma_{xz}$ , but this case is easy to carry out.

Two phonons are emitted, a longitudinal and a transverse, with amplitudes L and T, at angles  $\phi$  and  $\theta$  defined by

$$\frac{\sin \phi}{c_l} = \frac{\sin \theta}{c_l} = \frac{q}{\omega}$$

where the sound velocities  $c_i$ ,  $c_i$  are supposed isotropic for simplicity. They



Fig. 5. Notation used in Appendix A: phonons radiated in the solid by a percussion on the surface.

correspond to a bulk displacement field in the solid

$$\begin{pmatrix} u_x \\ u_y \end{pmatrix} = (q/\omega) e^{i(qy-\omega t)} \begin{pmatrix} 1 & \cot \phi \\ -\cot \theta & 1 \end{pmatrix} \begin{pmatrix} T e^{i(qx \cot \theta)} \\ L e^{i(qx \cot \phi)} \end{pmatrix}$$

The stress tensor is<sup>30</sup>

$$\sigma_{jk} = 2\rho c_t^2 u_{jk} + \rho (c_l^2 - 2c_t^2) u_{ll} \delta_{jk}$$

where  $u_{jk} = \frac{1}{2}(\partial_k u_j + \partial_j u_k)$ . Then at the surface (x = 0), some algebra yields

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{xy} \end{pmatrix} = -i\omega\rho \begin{pmatrix} \sin 2\theta & \cos 2\theta \\ -\cos 2\phi & (c_t/c_l)^2 \sin 2\phi \end{pmatrix} \begin{pmatrix} T \\ L \end{pmatrix} e^{i(qy-\omega t)}$$

$$\begin{pmatrix} \dot{u}_x \\ \dot{u}_y \end{pmatrix} = iq \begin{pmatrix} 1 & \cot \phi \\ -\cot \theta & 1 \end{pmatrix} \begin{pmatrix} T \\ L \end{pmatrix} e^{i(qy-\omega t)}$$

We obtain then the acoustic impedance matrix [Z] by writing

$$\begin{pmatrix} \dot{u}_x \\ \dot{u}_y \end{pmatrix} = -[Z]^{-1} \begin{pmatrix} \sigma_{xx} \\ \sigma_{xy} \end{pmatrix}$$

and

$$\rho[Z]^{-1} = \frac{1}{\Delta} \begin{pmatrix} \frac{\cos\phi}{c_l} & \frac{\sin(2\theta - \phi)}{c_l} \\ -\frac{\sin(2\theta - \phi)}{c_l} & \frac{\cos\theta}{c_t} \end{pmatrix}$$

where  $\Delta = 1 + 2(c_t/c_l) \sin 2\theta \sin(\theta - \phi)$ .

# A.2. Energy Radiated by a Percussion

Let us consider an atom which sticks to the solid. When it hits the surface, its momentum **p** has a modulus  $p \sim p_F$  and an incidence angle  $\psi$ . We can describe the shock with a form function  $f(\mathbf{r}, t)$  such as:  $\iiint f(\mathbf{r}, t) d^2\mathbf{r} dt = 1$ . The force per unit surface is  $\mathbf{\sigma} = \mathbf{p} \times f(\mathbf{r}, t)$ , and we have

$$\sigma_{xx} = \sigma \cos \psi;$$
  $\sigma_{xy} = \sigma \sin \psi;$   $\sigma_{xz} = 0$ 

The atom radiates in the solid an acoustic energy  $\operatorname{Re}(\bar{\sigma}_{xj}\dot{u}_j)$ . After decomposition of  $\sigma_{xj}$  on all the modes  $(\omega, q)$  the energy radiated as phonons is

$$E = \operatorname{Re} \iiint \bar{\sigma}_{xj} Z_{jk}^{-1} \sigma_{xk} d^2 r dt$$
$$= \frac{1}{\rho} \operatorname{Re} \iiint \frac{1}{\Delta} |\tilde{\sigma}_{q\omega}|^2 \left( \cos^2 \psi \frac{\cos \phi}{c_l} + \sin^2 \psi \frac{\cos \theta}{c_l} \right) d^2 q d\omega$$

This integration can be solved numerically using the fact that the impact is short and localized on an atomic distance a = 3 Å. The upper limits of the integral are  $q_{\max} = 2\pi/a = 2 \times 10^{10} \text{ m}^{-1}$  and  $\omega_{\max} = 2\pi v_F/a =$  $7 \times 10^{11} \text{ rad sec}^{-1}$ : as these limits are greater than the thermal frequencies and wavevectors, the integration domain includes the whole phonon spectrum.

But a simple estimate of the order of magnitude is obtained by roughly replacing  $\Delta$  by 1. For a given incidence angle  $\psi$  this expression reduces to

$$E(\psi) \approx \frac{p_F^2}{36\pi^2 \rho} \left(\frac{k_B T}{\hbar}\right)^3 \left(\frac{\sin^2 \psi}{c_l^3} + \frac{\cos^2 \psi}{c_l^3}\right)$$

An average on  $\psi$  lets appear the Debye temperature of the solid  $\Theta_D$ 

$$E_{\rm rad} = \frac{1}{3} \frac{p_F^2}{2m} \left(\frac{T}{\Theta_D}\right)^3$$

This energy represents the fraction of the latent heat released on the solid side by an atom sticking to the crystal; the remaining heat is retained in the liquid. We then obtain the total latent heat creation per unit mass in the solid:  $(\lambda - TS_S) = E_{rad}/m$ . It can be compared to the entropy in the solid, i.e.,  $S_S = k_B \log 2/m$ . With  $\Theta_D \approx 17.3 \text{ K}^{8,31}$  around the melting pressure, we obtain

$$\left|\frac{\lambda}{TS_S} - 1\right| \approx \frac{E_{\text{rad}}}{k_B T \log 2} \approx 6 \times 10^{-4} \text{ T}^2$$

This gives an upper limit  $|\lambda/TS_s - 1| \le 6 \times 10^{-5}$  at  $T_m = 0.32$  K. The energy radiated as phonons in the liquid is probably not larger.

# APPENDIX B: DETERMINATION OF $R_{\kappa}$ BY THE ACOUSTIC MISMATCH METHOD

This appendix is devoted to the discussion of  $R_K$ . At first, we justify briefly the use of Little's calculation of  $R_K$  for a solid-solid interface.<sup>22</sup> We then determine the energy transmission coefficient in the <sup>3</sup>He solid-<sup>3</sup>He liquid interface; we follow Cheeke, Hebral, and Martinon (CHM),<sup>32</sup> but the transverse acoustic impedance of the liquid is complex.

# **B.1.** Little's Model

Little uses a kinetic theory to describe the phonon heat transmission between two solids. In the solid 1, there are three phonon modes. For a given mode i, the incident heat flux on the interface is

$$J_{Q1}^{i} = \frac{1}{4\pi} \iiint \hbar \omega N(\omega) c_{i} \cos \theta \, d\Omega \, d\omega$$

where  $c_i$  is the velocity of the mode i,  $\theta \in [0, \pi/2]$  is the incidence angle, and  $N(\omega)$  is the Bose-Einstein distribution. Each phonon has a transmission rate  $\tau_{1-2}$ ; by taking into account the anisotropy of the sound velocity, the heat flux across the interface from solid 1 to 2 writes (within the Debye approximation)

$$J_{Q1-2}(T) = \frac{\pi^2}{30} \frac{(k_B T)^4}{\hbar^3} \sum_{\text{modes } i} \int \frac{d\Omega}{4\pi} \cos \theta \frac{\tau_{1-2}(\theta, \phi)}{c_i^2(\theta, \phi)}$$

Or, after derivation:

$$\frac{dJ_{Q1-2}(T)}{dT} = \frac{\pi^2}{30} \frac{k_B^4}{\hbar^3} 4T^3 \sum_{\text{modes } i} \frac{1}{4} \left\langle \frac{\tau_{1-2}}{c_i^2} \right\rangle$$

where the term 1/4 comes from the average on the incidence angles  $\theta$ ,  $\phi$  on the half-space. Eventually, the Kapitza resistance is given by

$$\frac{1}{R_K T^3} = 1.0 \times 10^6 \sum \left\langle \frac{\tau_{1-2}}{c_i^2} \right\rangle \qquad (R_K \text{ in } \text{cm}^2 \text{ K W}^{-1}; c \text{ in } \text{m sec}^{-1})$$

Both media play a symmetrical role, which is not obvious in this approach where the heat flux is studied only from one side. But the transmission coefficient contains all the physical information on the second medium, and allows to calculate  $R_{\kappa}$ .

Little's model presents a paradox: when both solids are identical, i.e., when the interface doesn't exist and  $\tau = 1$ , it predicts a finite surface resistance. Katerberg, Reynolds, and Anderson<sup>33</sup> carefully studied the validity of this model. They compared it with an alternative derivation<sup>34</sup> in which  $\tau$  is replaced by  $\tau/(1-\tau)$ . Both derivations are equivalent but use a different definition of the temperature.<sup>20</sup> In our case, the growth kinetics is controlled by the distribution of the incident phonons, and we consider the temperature as defined by Little.

#### **B.2.** The Phonon Transmission Rate

CHM describe in detail the complete calculation of  $\tau$  for a solid-solid interface. The same algebra can be used here, within a "modified" model because the transverse acoustic impedance  $Z_t^L$  of the liquid is complex instead of being real. The Snell-Descartes law formally relates the acoustic impedance of each mode to the directions of the phonons, but here the complex angles may lose their geometrical signification. The wave-vectors get an imaginary part describing their damping. The four boundary conditions, given by the continuity of the stress tensor and by mass conservation at the interface, yield the amplitude transmission rate for an incident pressure wave. The calculation involves the inversion of a  $4 \times 4$  matrix, formally identical as CHM. One must be careful to come back to real quantities when determining the energy transmission rate; the energy flux associated to each wave is

$$J_O = \operatorname{Re}(\bar{\sigma}_{xx}\dot{u}_x + \bar{\sigma}_{xy}\dot{u}_y)$$

CHM can define a critical cone: inside this cone, an incident phonon is partially transmitted and emits in solid 2 a phonon with a real wavevector; outside, the incident phonon is totally reflected and emits only an evanescent wave (overdamped complex wavevector). Here, the emitted transverse phonon has always a complex wavevector, and the transition between the transmitted and the reflected domains is very smooth.

Another difference with a solid-solid interface comes from its mobility. The crystal can grow or melt. The interface tends to keep the pressure to its equilibrium value  $P_{eq}(T)$ , thus creating a pressure node at the surface.<sup>2</sup> The more mobile the interface, the more it hinders the phonon transmission.  $R_{K}$  increases with the mobility; the effect of k on the transmission is described by the dimensionless parameters:  $kc_i(\Delta\rho)^2/\rho_S\rho_L$ . If it is more than one,  $R_{K}$  is greatly modified; for instance, <sup>4</sup>He is an extreme case where at zero temperature k diverges, and  $R_{K}T^3$  also diverges.<sup>35</sup> Here, even for the quickest mode ( $c_i = 480 \text{ m/sec}$ ) this parameter reaches only 0.2 and  $R_{K}$  is not much modified.

To take this effect into account, the boundary conditions for a massless interface is expressed as

$$\dot{u}_{x}^{(2)} = \dot{u}_{x}^{(1)} + \left(\frac{\Delta\rho}{\rho_{L}}\right)^{2} \frac{k}{\rho_{S}} \sigma_{xx}^{(2)} \qquad \sigma_{xx}^{(2)} = \sigma_{xx}^{(1)}$$
$$\dot{u}_{y}^{(2)} = \dot{u}_{y}^{(1)} \qquad \sigma_{xy}^{(2)} = \sigma_{xy}^{(1)}$$

We included the value k = 0.18 sec m<sup>-1</sup> in the numerical calculations presented above. If we do them again by taking k = 0, we obtain a diminution of  $R_K$  of 7%.

CHM also studied the validity of the Debye approximation in Little's model. They tabulated the error as a function of  $T/\Theta_D$ . Here, with  $\Theta_D = 17.3$  K, the correction is of order 3%.

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