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Sound propagation in a superleak covered with solid ^4He

M. Uwaha

Institut Laue-Langevin, BP 156X, 38042 Grenoble Cedex, France

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Résumé. — On étudie les effets dus à la croissance cristalline sur la propagation du quatrième son, dans le cadre de l'hydrodynamique. Le système particulier que nous envisageons est une superfuite en graphite exfolié remplie d'hélium 4. L'hélium 4 solide se forme sur la surface du graphite et s'accroît d'une façon continue au fur et à mesure que la pression du liquide s'accroît lors de la propagation du son. La vitesse du son et les déplacements Doppler pour un écoulement superfluide permanent sont calculés en fonction de la force du potentiel du substrat (ce qui revient à l'épaisseur de la couche solide) et de la vitesse de croissance. Dans la limite d'un fort potentiel de substrat (couche solide mince) ou d'une croissance lente, on retrouve les propriétés usuelles du quatrième son. Par contre, pour un potentiel faible et une cristallisation rapide, on observe des phénomènes nouveaux : la vitesse du son tend à s'annuler pour $T = 0$ car la vitesse rapide de croissance empêche les changements de pression du liquide, mais à température finie la variation de densité d'entropie associée à la cristallisation (ou la fusion) permet la propagation du son. Le déplacement Doppler est beaucoup plus grand que la vitesse de l'écoulement et de sens opposé. Les conditions expérimentales pour une superfuite utilisant du grafoil sont considérées.

Abstract. — Effects of crystal growth on the fourth sound propagation are studied in the framework of hydrodynamics. The particular system we have in mind is superfluid ^4He in a grafoil superleak. Solid ^4He is formed on the surface of graphite and it grows continuously when the pressure of liquid is increased during passage of a sound wave. Sound velocity and Doppler shifts for steady superfluid flow are calculated as a function of the strength of the substrate potential (or equivalently the thickness of the solid layer) and the rate of crystal growth. In the limit of strong substrate potential (thin solid layer) or slow crystal growth they are the same as the usual fourth sound. On the other hand, if the potential is weak and the crystal growth is fast, we find several new features : the sound velocity approaches zero at $T = 0$ because the fast crystal growth does not allow pressure changes of the liquid, but at finite temperatures the change of entropy density on crystallization (or melting) assures propagation of the sound. The Doppler shift is much larger than the velocity of the flow and is in the opposite sense. Experimental conditions for grafoil superleaks are considered.

1. Introduction.

At low temperatures solid ^4He grows from superfluid exceedingly fast [1-3] and this causes various interesting phenomena at the interface between solid and liquid such as crystallization waves [3-8], a striking decrease in sound transmission [9, 10] and anomalous temperature dependence of the Kapitza resistance [11-14, 7]. The fast crystal growth can affect some bulk properties of superfluid ^4He if the ratio of the interface area to the volume is sufficiently large. Recently Maynard, Jelatis and Roth [15] found an anomalous oscillation in fourth sound velocity in grafoil as a function of external pressure. Ramesh and Maynard [16] attributed this phenomenon to layer-by-layer crystal growth on the graphite substrate. Bearing this system in mind, we study propagation of hydrodynamic sound in a super-

leak which is covered with solid ^4He ⁽¹⁾, where the crystal growth can change the essential features of fourth sound.

We consider a superfluid in a very narrow channel between two parallel plates which are covered with layers of solid ^4He formed by their van der Waals potentials. When a sound wave propagates in this system, the increase of pressure causes crystal growth of the solid. If the crystal growth is fast enough, the

⁽¹⁾ In the cases usually studied solid will not be formed in superleaks [17-20]. Even if solid is formed on the substrates, it does not grow continuously [21]. In this respect solid ^4He on graphite is an exception ; it grows continuously to a bulk solid [22-24]. In this paper the words « a superleak covered with solid » always imply such a case in which the solid can grow continuously from the thin layer to a bulk phase.

equilibrium between the solid and the liquid is maintained and the sound velocity will be modified. The extent of crystal growth is controlled by the strength of the substrate potential and therefore the change of the sound velocity depends on the thickness of the solid layer.

Liquid ^4He in a superleak is not Galilean invariant at finite temperatures since the normal flow of the liquid is locked to the wall. In the present system, in addition, Galilean invariance is broken by the existence of crystal growth even at zero temperature. For this reason we study sound propagation in a steadily flowing superfluid and look at how the Doppler shift is changed. We formulate a theory within the framework of hydrodynamics [25-27] and are not concerned with crystal growth at atomic scales, a subject which remains a fertile field to be explored.

We give a simple analysis of the problem, in 2, based on conservation laws at zero temperature. When the crystal growth is fast, we find a remarkable change of the sound velocity as the strength of the substrate potential changes. The Doppler shift also shows a drastic change, and even its sign is changed when the potential is weak. The treatment of 2 is justified in the appendix and extended to finite temperatures in 3. At finite temperatures entropy plays an important role as in the case of second sound. We find appreciable temperature dependences of both the sound velocity and Doppler shift. In the limit of weak potential, where sound does not propagate at zero temperature, there is a new sound mode due to the change of entropy density on crystallization. In 4 the results are summarized and realistic situations are discussed.

2. Zero temperature.

We use the following model to study sound propagation in a solid-covered superleak: The superleak consists of two parallel plates separated by a distance $2d$ ($2d$ is about 400 \AA for grafoil [24]). They are covered with ^4He solid formed by their van der Waals potentials (Fig. 1). Let z be the coordinate normal to the surface, x -axis at the centre of the channel, and ζ and $-\zeta$ the symmetrical positions of the interface (we assume $d - \zeta \ll d$). The superfluid ^4He is flowing steadily with a velocity v_0 .

At zero temperature, the propagation of long wavelength sound ($k\zeta \ll 1$) is determined by a continuity equation, an equation for superfluid velocity, and a growth rate equation at the interface:

$$\frac{\partial}{\partial t}(n_2 \zeta) - n_1 \frac{\partial \zeta}{\partial t} + \frac{\partial}{\partial x}(n_2 \zeta v) = 0, \quad (1)$$

$$\frac{\partial v}{\partial t} + \frac{1}{m} \frac{\partial}{\partial x} \left(\mu_2 + \frac{1}{2} m v^2 \right) = 0, \quad (2)$$

$$\frac{1}{K} \frac{\partial \zeta}{\partial t} + \left(\mu_2 + \frac{1}{2} m v^2 - \mu_1 \right) \Big|_{\zeta} = 0, \quad (3)$$

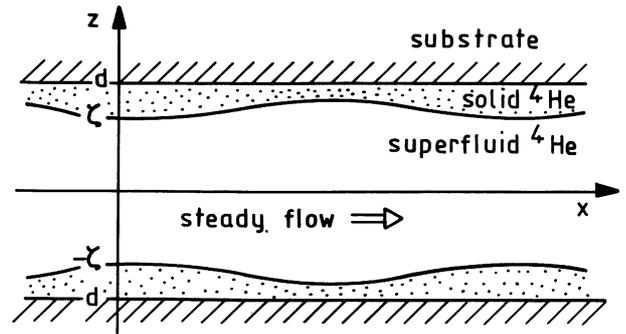


Fig. 1. — The superleak consists of two parallel plates, which are covered with solid ^4He formed by their van der Waals potentials. The upper half and the lower half are symmetrical. The solid thickness $d - \zeta$ is much smaller than the width $2d$. The superfluid ^4He is flowing steadily with velocity v .

where 1 and 2 represent solid and liquid respectively, n is the number density, μ the chemical potential per atom, and K the crystal growth coefficient defined as in reference [3].

In (1) and (2) we have neglected the z -dependence due to the substrate potential, which will be examined later. For simplicity the solid has been assumed to be rigid; n_1 is constant and there is no equation of motion for the solid. We will make a similar assumption for the entropy of the solid in 3. The inclusion of these effects would bring corrections of the order of $(d - \zeta)/d$, which we have assumed small compared to unity. Equation (3) is a natural generalization of the growth rate equation [3] in the presence of the steady superfluid flow. The chemical potentials in (3) are evaluated at the interface. Melting or crystallization will produce a tangential force on the solid, which will be transmitted directly to the substrate as we have assumed that the solid is rigid. The interfacial tension and the interfacial inertia [7] are not included for the following reasons: the ratio of the pressure change due to the surface tension to the compression is very small:

$$\frac{k^2 \alpha \zeta'}{m c_0^2 n_2} \sim \frac{k^2 \alpha \zeta}{m c_0^2 n_2} \lesssim k a \cdot k \zeta \ll 1, \quad (4)$$

where the quantities with a prime indicate the deviations from the equilibrium, and α is the interfacial tension, c_0 the first sound velocity of liquid, and a the atomic distance. The inertia of the interface [7] σ would appear in (3) as a term

$$\frac{\sigma}{n_1 n_2} n_1 \frac{\partial^2 \zeta}{\partial t^2},$$

and the ratio of this term to the first term of (3) is

$$\frac{\omega^2 \sigma n_2^{-1} \zeta'}{\omega K^{-1} \zeta'} \sim \frac{c_0}{(mK)^{-1}} k a; \quad (5)$$

we suppose that this value is much smaller than unity (see 4).

The two-phase equilibrium condition for chemical potentials with the steady flow v_0 is given by

$$\mu_2(\zeta_0) + \frac{1}{2}mv_0^2 - \mu_1(\zeta_0) = 0. \tag{6}$$

When there is a small perturbation, (6) becomes

$$\begin{aligned} \mu_2(\zeta_0 + \zeta') + \frac{1}{2}m(v_0 + v)^2 - \mu_1(\zeta_0 + \zeta') &= \frac{1}{n_{20}} \left(P_2' + \frac{\partial P_2}{\partial z} \zeta' \right) + mv_0 v' - \frac{1}{n_{10}} \left(P_2' + \frac{\partial P_2}{\partial z} \zeta' \right) = \\ &= \left(\frac{1}{n_{20}} - \frac{1}{n_{10}} \right) \left(mc_0^2 n_2' - n_{20} \frac{\partial U}{\partial z} \zeta' \right) + mv_0 v', \end{aligned} \tag{7}$$

where we have used the equilibrium condition

$$\frac{\partial P_2}{\partial z} = -n_{20} \frac{\partial U}{\partial z} \tag{8}$$

to relate the pressure to the substrate potential. Substituting (7) into (3), we obtain the system of linearized equations :

$$-(n_1 - n_2) \frac{\partial \zeta'}{\partial t} + n_2 v \frac{\partial \zeta'}{\partial x} + \zeta \frac{\partial n_2'}{\partial t} + \zeta v \frac{\partial n_2'}{\partial x} + n_2 \zeta \frac{\partial v'}{\partial x} = 0, \tag{9}$$

$$\frac{c_0^2}{n_2} \frac{\partial n_2'}{\partial x} + \frac{\partial v'}{\partial t} + v \frac{\partial v'}{\partial x} = 0, \tag{10}$$

$$\frac{1}{mK} \frac{\partial \zeta'}{\partial t} - \frac{n_1 - n_2}{n_1} \frac{1}{m} \frac{\partial U}{\partial z} \zeta' + \frac{n_1 - n_2}{n_1 n_2} c_0^2 n_2' + vv' = 0. \tag{11}$$

where we omit the suffix 0 for equilibrium values.

Solutions of (9), (10) and (11) proportional to $e^{ikx - i\omega t}$ satisfy the dispersion relation,

$$\left(\omega + kv \frac{n_2}{n_1 - n_2} \right)^2 + \left(-i\omega \frac{\zeta}{mKc_0^2} + \frac{n_1 - n_2}{n_1} \frac{\zeta}{mc_0^2} \left| \frac{\partial U}{\partial z} \right| \right) \frac{n_1 n_2}{(n_1 - n_2)^2} [(\omega - kv)^2 - k^2 c_0^2] = 0. \tag{12}$$

The first term is essentially a result of continuity at the interface; the interface moves $n_2/(n_1 - n_2)$ times faster than the flow of the liquid and in the opposite direction. The second term gives the usual fourth sound modes with the Doppler shift v . The coupling factor consists of a term proportional to the substrate potential and a term related to the crystal growth rate. The latter causes a delay of the phase and thereby attenuation of the wave.

If the solid grows very slowly ($K \rightarrow 0$) or the substrate potential is very strong, we have the usual fourth sound propagation with the Doppler shift v :

$$\omega = kv \pm kc_0. \tag{13}$$

On the other hand, when the solid grows fast ($K \rightarrow \infty$), the frequency is given by (including only linear terms in v)

$$\omega = kv \frac{p - 1}{p + r} \pm kc_0 \sqrt{\frac{p}{p + r}}, \tag{14}$$

where p and r are the dimensionless quantities :

$$p = \frac{\zeta}{mc_0^2} \left| \frac{\partial U}{\partial z} \right|, \quad r = \frac{n_1 - n_2}{n_2}. \tag{15}$$

r is about 0.1 and, for a graphite substrate, p is unity when the solid thickness $d - \zeta$ is about 12 Å. The sound velocity is smaller than c_0 because the pressure change is absorbed partially by the immediate crystal growth and the liquid therefore appears to be more compressible [16]. When the potential is weak (p is small), the displacement of the interface is large and this effect is enhanced. In the limit of weak potential the pressure change is completely absorbed and sound does not propagate. The Doppler shift also becomes smaller and it changes its sign. This anomalous Doppler shift is a result of the continuity as mentioned before and a characteristic phenomenon of this system.

In the limits of slow and fast crystal growth the attenuation of sound is negligible. It becomes important when $\omega\zeta(mKc_0^2)^{-1} \sim 1$. When the attenuation is small, the imaginary part of the frequency, ω'' , is given by

$$\omega'' = -\frac{1}{2} \frac{mKc_0^2}{\zeta} \frac{(n_1 - n_2)^2}{n_1 n_2} \tag{16}$$

for slow growth and

$$\omega'' = -\frac{1}{2} \frac{\zeta}{mKc_0^2} \frac{n_1 n_2}{(n_1 - n_2)^2} \left(\frac{r}{p+r}\right)^2 c_0^2 k^2 \tag{17}$$

for fast growth. The change of the complex frequency $\omega = \omega' + i\omega''$ for fixed k , v , and p , with K changed, is depicted schematically in figure 2.

In the above argument, when we use (1) and (2), we have neglected the z -dependence of the substrate potential $U(z)$, which is essential to have a solid only near the walls. We examine whether this simplification is allowed in the Appendix. For small k ($k\zeta \ll 1$) the result is very simple; in (12) we should use the values at melting point for n_2 and c_0^2 except the last c_0^2 , which should be replaced by $\langle n_2 \rangle \langle n_2/c_0^2 \rangle^{-1}$, where $\langle \dots \rangle$ indicates the average value over the liquid volume. Therefore using (1) and (2), with the equilibrium values at the melting pressure, is a good approximation unless pressure at the center is too low.

3. Finite temperature.

At finite temperatures the normal component of the ^4He liquid appears, and it is completely locked to the solid wall since ζ is much shorter than the mean free paths of the thermal excitations. In addition to the corresponding equations to (1), (2), and (3), we need an equation for entropy. With the condition $v_n = 0$ taken into account, the necessary set is

$$\frac{\partial}{\partial t}(n_2 \zeta) - n_1 \frac{\partial \zeta}{\partial t} + \frac{\partial}{\partial x}(n_s \zeta v) = 0, \tag{18}$$

$$\frac{\partial}{\partial t}(n_2 s_2 \zeta) - n_1 s_1 \frac{\partial \zeta}{\partial t} = 0, \tag{19}$$

$$\frac{\partial v}{\partial t} + \frac{1}{m} \frac{\partial}{\partial x} \left(\mu_2 + \frac{1}{2} m v^2 \right) = 0, \tag{20}$$

$$\frac{1}{K} \frac{\partial \zeta}{\partial t} + \left(\mu_2 + \frac{1}{2} m v^2 - \mu_1 \right) \Big|_{\zeta} = 0, \tag{21}$$

where v represents the superfluid velocity v_s , n_s the number density of the superfluid flow, and s the entropy per atom. We have assumed $\partial n_1/\partial P = 0$ and $\partial s_1/\partial T = 0$ to avoid uninteresting complications. The entropy production in crystal growth is not included in (19) since it is proportional to $(\partial \zeta/\partial t)^2$ and therefore a higher order effect. We neglect all dissipative terms in these equations. The growth rate equation (21) is written in the frame of reference of the solid, and in an arbitrary frame it is written as ⁽²⁾

$$\frac{1}{K} \left(\frac{\partial \zeta}{\partial t} + v_1 \frac{\partial \zeta}{\partial x} \right) + \left(\mu_2 + \frac{1}{2} m v_s^2 - m v_s v_n - \mu_1 + \frac{1}{2} m v_1^2 \right) \Big|_{\zeta} = 0, \tag{22}$$

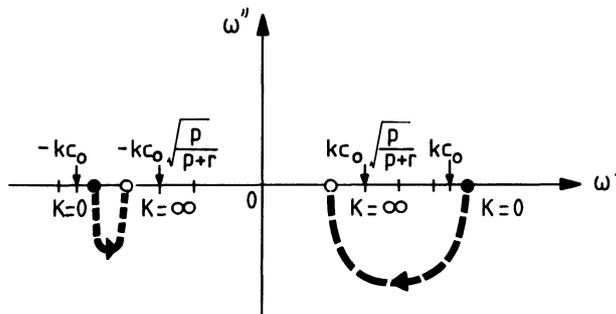


Fig. 2. — The complex frequencies of the sound for a given potential, flow velocity v , and wave number k is shown schematically as a function of the growth coefficient. The points $K \rightarrow 0$ are the fourth sound modes without crystal growth and $K \rightarrow \infty$ is the fast growth limit. The Doppler shift can be in the opposite sense in the latter.

⁽²⁾ This equation is a natural generalization of the growth rate equation [3] when the velocities are non-zero. The equilibrium condition for this case (i.e. the first term of (22) vanishes) is given in [27].

with $v_1 = v_n$ in the present case. The chemical potential μ_2 depends on the relative velocity $v (= v_s - v_n)$

$$d\mu_2 = \frac{1}{n_2} dP - s_2 dT - m x_n v dv, \tag{23}$$

where $x_n = n_n/n_2$. To second order in velocity the number density n_2 and the entropy are given by

$$n_2 = n_2(P, T) + \frac{1}{2} m v^2 n_2^2 \frac{\partial x_n}{\partial P}, \tag{24}$$

$$s_2 = s_2(P, T) + \frac{1}{2} m v^2 \frac{\partial x_n}{\partial T}. \tag{25}$$

The linearized equations of (18)-(21) are the following:

$$\left[- (n_1 - n_2) \frac{\partial}{\partial t} + n_s v \frac{\partial}{\partial x} \right] \zeta' + \zeta \left[\frac{\partial n_2}{\partial P} \frac{\partial}{\partial t} + \left(\frac{\partial n_2}{\partial P} \frac{n_s}{n_2} - n_2 \frac{\partial x_n}{\partial P} \right) v \frac{\partial}{\partial x} \right] P' - \zeta n_2 \frac{\partial x_n}{\partial T} v \frac{\partial}{\partial x} T' + \zeta \left[m v n_2^2 \frac{\partial x_n}{\partial P} \frac{\partial}{\partial t} + \left(n_s + m v^2 n_s n_2 \frac{\partial x_n}{\partial P} \right) \frac{\partial}{\partial x} \right] v' = 0, \tag{26}$$

$$(n_1 s_1 - n_2 s_2) \zeta' - \zeta \frac{\partial n_2}{\partial P} s_2 P' - \zeta n_2 \frac{\partial s_2}{\partial T} T' - \zeta m v n_2 \left(n_2 s_2 \frac{\partial x_n}{\partial P} + \frac{\partial x_n}{\partial T} \right) v' = 0, \tag{27}$$

$$\frac{1}{m n_2} \frac{\partial}{\partial x} P' - \frac{s_2}{m} \frac{\partial}{\partial x} T' + \left(\frac{\partial}{\partial t} + \frac{n_s}{n_2} v \frac{\partial}{\partial x} \right) v' = 0, \tag{28}$$

$$\left(\frac{1}{mK} \frac{\partial}{\partial t} - \frac{n_1 - n_2}{n_1} \frac{1}{m} \frac{\partial U}{\partial z} \right) \zeta' + \frac{1}{m} \frac{n_1 - n_2}{n_1 n_2} P' + \frac{s_1 - s_2}{m} T' + \frac{n_s}{n_2} v v' = 0, \tag{29}$$

where we have neglected the coupling between the mechanical variables and the thermal variables. We also put $T'_1 = T'_2$ because the Kapitza resistance is normal at the temperature range where the thermal effects discussed here are important. To first order in v , the dispersion relation obtained from (26)-(29) has the simple structure

$$(\omega^2 + 2 \omega k v_t - k^2 c_t^2) + g(\omega) (\omega^2 - 2 \omega k v_f - k^2 c_f^2) = 0, \tag{30}$$

where c_f and v_f are the usual fourth sound velocity [26, 27] and its Doppler shift [28],

$$c_f^2 = \frac{n_s}{n_2} \frac{1}{m} \left[\left(\frac{\partial n_2}{\partial P} \right)^{-1} + s_2^2 \left(\frac{\partial s_2}{\partial T} \right)^{-1} \right], \tag{31}$$

$$v_f = v \left[\frac{n_s}{n_2} - n_2 \frac{\partial x_n}{\partial P} \left(\frac{\partial n_2}{\partial P} \right)^{-1} + s_2 \frac{\partial x_n}{\partial T} \left(\frac{\partial s_2}{\partial T} \right)^{-1} \right], \tag{32}$$

the coupling factor $g(\omega)$ is defined by

$$g(\omega) = g_1 - i \omega g'_2 = \frac{n_1 - n_2}{n_2} \frac{\zeta}{m c_0^2} \left| \frac{\partial U}{\partial z} \right| \frac{1}{R} - i \omega \frac{\zeta}{m K c_0^2} \frac{n_1}{n_2} \frac{1}{R}, \tag{33}$$

$$R = \left(\frac{n_1 - n_2}{n_2} \right)^2 + \left(\frac{n_1}{n_2} \right)^2 (s_1 - s_2)^2 \frac{\partial n_2}{\partial P} \left(\frac{\partial s_2}{\partial T} \right)^{-1}, \tag{34}$$

and c_t and v_t are given by

$$c_t^2 = \frac{n_s}{n_2} \frac{1}{m} \left(\frac{n_1 s_1 - n_2 s_2}{n_2} \right)^2 \left(\frac{\partial s_2}{\partial T} \right)^{-1} \frac{1}{R}, \tag{35}$$

$$v_t = v \left[\frac{n_s}{n_2} \frac{n_1 - n_2}{n_2} - \frac{n_s}{n_2} \left(\frac{n_1}{n_2} \right)^2 s_1 (s_1 - s_2) \frac{\partial n_2}{\partial P} \left(\frac{\partial s_2}{\partial T} \right)^{-1} + \frac{n_1}{n_2} (s_1 - s_2) (n_1 s_1 - n_2 s_2) \frac{\partial x_n}{\partial P} \left(\frac{\partial s_2}{\partial T} \right)^{-1} - \frac{n_1 - n_2}{n_2} \frac{n_1 s_1 - n_2 s_2}{n_2} \frac{\partial x_n}{\partial T} \left(\frac{\partial s_2}{\partial T} \right)^{-1} \right] \frac{1}{R}. \tag{36}$$

There are several important differences between (30) and the zero-temperature result (12). The first term of (30), which gives the dispersion relation in the limit of fast crystal growth, $K \rightarrow \infty$, and weak substrate potential, $p \rightarrow 0$, is quite different from that of (12). At finite temperatures there is a sound mode with the velocity c_i and the Doppler shift v_i . The mechanism which brings about the sound is the following. We consider the case of $v=0$. Suppose an inhomogeneous fluctuation of the superfluid velocity produces crystal growth at some point. The superfluid flow enters this region without carrying entropy. The conservation of entropy (27) relates the amount of the crystal growth to the resultant changes of pressure and temperature :

$$(n_1 s_1 - n_2 s_2) \zeta' = \zeta \left(s_2 \frac{\partial n_2}{\partial P} P' + n_2 \frac{\partial s_2}{\partial T} T' \right). \quad (37)$$

In the limit of fast crystallization (and melting), the pressure and the temperature deviate from their unperturbed values, with deviations tangential to the melting curve. The deviations P' and T' satisfy

$$\left(\frac{1}{n_1} - \frac{1}{n_2} \right) P' = (s_1 - s_2) T'. \quad (38)$$

The changes of pressure and temperature imply a change of chemical potential μ'_2 . The gradient of μ'_2 accelerates the superfluid ((28) with $v = 0$);

$$v' = \frac{k}{m\omega} \left(\frac{1}{n_2} P' - s_2 T' \right). \quad (39)$$

For ${}^4\text{He}$ both the latent heat and the difference of the entropy densities change their signs near $T = 0.8$ K and several different relative signs of the amplitudes, ζ' , P' and T' , are possible along the melting curve. However the chemical potential change μ'_2 obtained from (37) and (38),

$$\mu'_2 = \frac{(n_1 s_1 - n_2 s_2)^2}{n_1 n_2 (s_1 - s_2)} \left[\frac{\partial n_2}{\partial P} s_2 - \frac{\partial s_2}{\partial T} \frac{n_1 - n_2}{n_1 n_2 (s_1 - s_2)} \right]^{-1} \frac{\zeta'}{\zeta}, \quad (40)$$

is always positive at the point where the solid is thicker ($\zeta' < 0$) and decelerates v' to produce outgoing flow. Thermal effects are important at high temperature $T \gtrsim 1$ K, where $s_1 < s_2$ and $n_1 s_1 < n_2 s_2$. In this temperature region, when the solid grows, the decrease of the liquid volume causes increases of the pressure and the temperature. At the point of minimum melting pressure on the melting curve, where $s_1 = s_2$, the sound is a pure temperature wave ($P' = 0$). In this case, contrary to that of high temperature, the point where the crystal is growing is cooled by the superfluid and the temperature decreases. This is true as long as $n_1 s_1 > n_2 s_2$. The velocity of this pure thermal wave is

$$c_i^2 = \frac{n_n}{n_2} c_s^2, \quad (41)$$

where c_s is the second sound velocity

$$c_s^2 = \frac{n_s}{n_n} \frac{s_2^2}{m} \left(\frac{\partial s_2}{\partial T} \right)^{-1} \quad (42)$$

The velocity c_i is very small at low temperatures and only in the high temperature regime may these effects be observed in practice. The Doppler shift v_i does not depend on temperature very much.

If either the substrate potential is strong, $p \rightarrow \infty$, or the crystal growth is slow, $K \rightarrow 0$, the crystal does not grow very much. The second term of (30) is dominant and gives the usual fourth sound mode with Doppler shift. The finite temperature correction of the Doppler shift v_i (32), is important; the second term in the bracket becomes of the order of unity at 1.2 K and v_i is negative at higher temperatures.

These two sound modes are combined by the crystal growth and the substrate potential, i.e., the coupling factor $g(\omega)$. When the crystal growth is fast enough that the imaginary part of $g(\omega)$ is negligible ($\omega g'_2 \rightarrow 0$), the sound velocity c is given by

$$c^2 = c_0^2 \frac{n_s}{n_2} \frac{rp \left[1 + \frac{s_2^2}{mc_0^2} \left(\frac{\partial s_2}{\partial T} \right)^{-1} \right] + \left(\frac{n_1 s_1 - n_2 s_2}{n_2} \right)^2 \frac{1}{mc_0^2} \left(\frac{\partial s_2}{\partial T} \right)^{-1}}{r^2 + rp + \left(\frac{n_1}{n_2} \right)^2 \frac{(s_1 - s_2)^2}{mc_0^2} \left(\frac{\partial s_2}{\partial T} \right)^{-1}}, \quad (43)$$

which is compared to (14). At high temperatures ($T \gtrsim 1.2$ K) the three terms containing $(\partial s_2/\partial T)^{-1}$ are of about the same magnitude since $s_2 - s_1 \gg s_1$. Unless the potential is very weak the main temperature dependence comes from n_s/n_2 . In figure 3 we give crude pictures of the velocity c and the Doppler shift v_d , which is given by

$$v_d = \frac{-v_t + g_1 v_f}{1 + g_1}, \quad (44)$$

as a function of p . We plot c and v_d versus $p^{-1/4}$, which is approximately proportional to the solid thickness. They change drastically in the range $1 \lesssim p^{-1/4} \lesssim 4$.

In the limit of weak potential ($p \rightarrow 0$) the coupling $g(\omega)$ is purely imaginary, and the solutions of (30) for

a given k are complex numbers. The traces of the complex $\omega (= \omega' + i\omega'')$ for $v = 0$, as the growth coefficient K changes, are drawn schematically in figure 4. At zero temperature only the usual fourth sound modes $\omega = \pm c_f k$ exist when $K = 0$. Introduction of crystal growth causes damping of the sound and they become overdamped modes when K is sufficiently large. At finite temperatures there are two slightly different cases. If $c_t < c_f/3$, there are usual fourth sound and a relaxational mode with very small ω'' when K is small. With increasing K all the modes become overdamped. When K is large enough, new sound modes appear and the relaxational mode relaxes very fast, reflecting fast crystal growth. If $c_t > c_f/3$, the general features are similar. The fourth

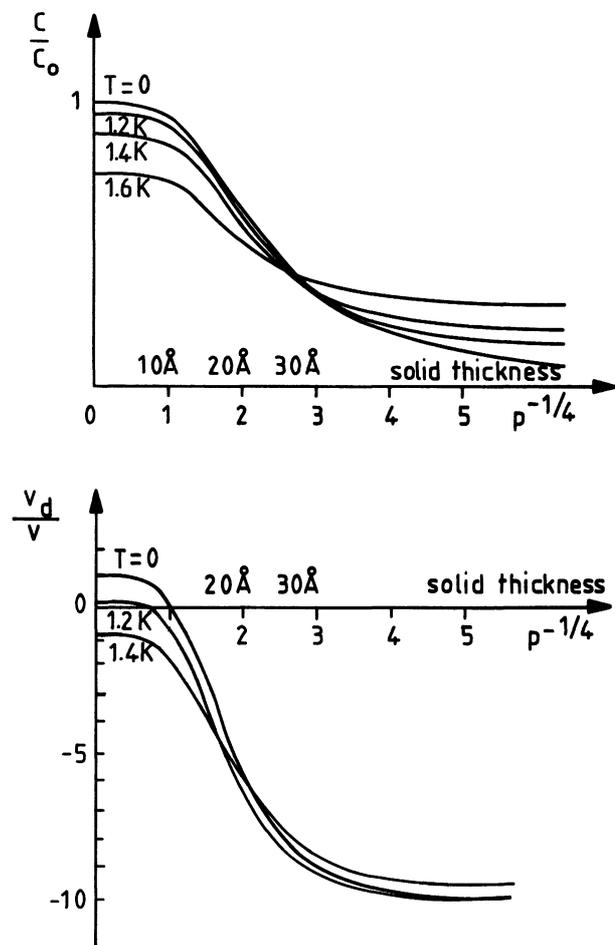


Fig. 3. — Sound velocity and Doppler shift for $K \rightarrow \infty$. [Thermodynamic quantities which are necessary for this estimate can be found in the following papers : MAYNARD, J., *Phys. Rev.* **B14** (1976) 3868, GRILLY, E. R., *J. Low Temp. Phys.* **11** (1973) 33].

a) The sound velocity c (c_0 is the first sound velocity) becomes smaller as the potentials, $p = (\zeta/mc_0^2) |\partial U/\partial z|$, becomes weaker (the solid layer is thicker). At finite temperatures c remains finite when $p \rightarrow 0$.

b) The Doppler shift v_d changes drastically from v to $-vn_2/(n_1 - n_2)$ at $T = 0$.

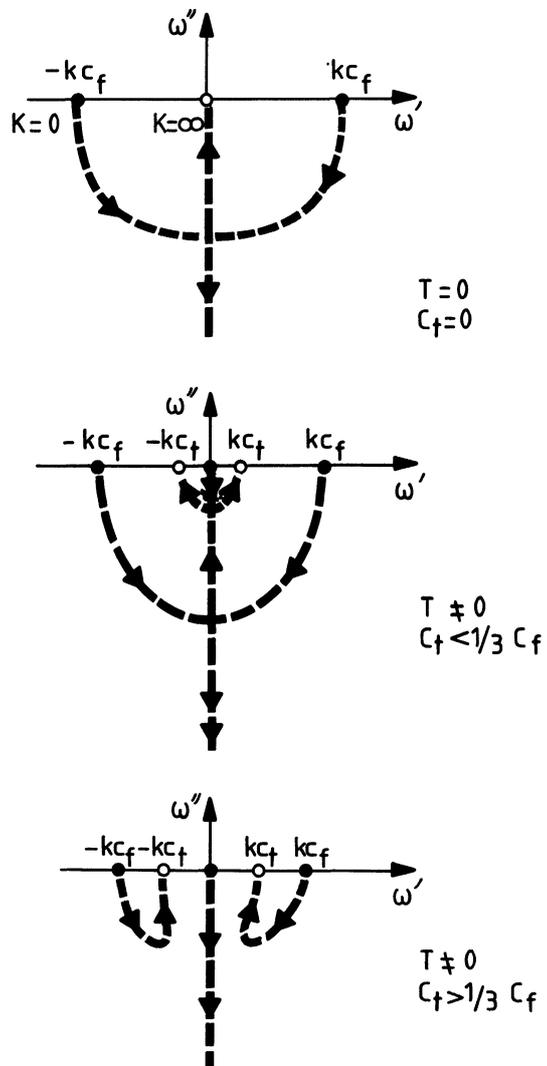


Fig. 4. — The traces of the complex frequencies, $\omega = \omega' + i\omega''$, for $v = 0$ and a given k in the limit of weak potential, $p = 0$, as the growth coefficient K changes. The frequencies move from the full circles for $K = 0$ to the open circles for $K = \infty$ along the broken lines. The fourth sound modes $\omega = \pm c_f k$ becomes overdamped modes ($T = 0$) or new sound $\omega = \pm c_t k$ ($T \neq 0$).

sound modes, however, are transformed directly into the other sound modes (ω' always remains finite), and the slowly relaxing mode becomes the fast relaxing mode when K is increased. The transition from the usual fourth sound $\omega = \pm kc_t$ to the new sound $\omega = \pm kc_l$ occurs when g_2 is of the order of $(kc)^{-1}$. Note that in reality the crystal growth coefficient K is not a controllable parameter : K is a function of temperature. We have treated K as an independent parameter for convenience only.

4. Discussion.

As we have seen, sound propagation in a superleak covered with solid ^4He will be quite different from the usual fourth sound propagation. With fast crystal growth the sound velocity changes drastically as the thickness of the solid changes (see Figs. 2 and 3), and the Doppler shift is of the opposite sense to the superfluid flow when the solid layer is thick. At zero temperature with a vanishing substrate potential (thick solid layer), sound does not propagate since the instantaneous crystal growth absorbs the pressure change of the liquid. However at finite temperatures a sound mode exists ; the change of entropy density in crystallization or melting produces a change of chemical potential along the melting curve, and the gradient of chemical potential accelerates the superfluid flow. Let us discuss briefly the range of parameters that pertain to realistic cases.

Solid ^4He grows epitaxially on a graphite substrate. The typical width $2d$ of a grafoil superleak is about 400 Å and the van der Waals potential is approximately [24]

$$U(z) = - \frac{\alpha}{(d-z)^3}, \quad (45)$$

with $\alpha = 3 \times 10^{-37}$ erg.cm³ (this value has been used to relate the potential strength p to the solid thickness in the figures). The crystal growth coefficient for rounded faces of ^4He crystals seems to be limited only by scattering of phonons and rotons aside from crystal defects. The agreement of the theoretical calculations [3, 29, 30] and the experimental results [5, 10, 31] is reasonably good. The value of the growth coefficient is strongly temperature dependent : $(\text{mK})^{-1} \sim 0.1$ cm/s at 0.4 K and $(\text{mK})^{-1} \sim 10^3$ cm/s at 1.3 K. Its value for the c -axis facet can be estimated from the acoustic transmission experiments [10] and the direct observation in melting [14] to be of the order of 10^2 cm/s or smaller. Recently Wolf, Balibar, and Gallet [32] reported that the value for growing facet was $(\text{mK})^{-1} \sim 10^4$ cm/s. However, for facets there seem to be a threshold chemical potential difference $\mu_2 - \mu_1$ below which the crystal does not grow [33] (i.e. $K = 0$). The growth rate of facets is therefore still somewhat uncertain. The imaginary

part of $g(\omega)$ is given by

$$\frac{\omega\zeta}{mKc_0^2} \frac{n_1}{n_2} \frac{1}{R} \sim k\zeta \frac{(mK)^{-1}}{c_0} \left(\frac{n_2}{n_1 - n_2} \right)^2. \quad (46)$$

If there is no threshold, this quantity is much smaller than unity for sound of moderate frequency. It is certainly small, at least, above the roughening transition temperature [34, 35, 5]. In other words crystallization and melting occur fast enough to maintain the equilibrium between the solid and the liquid. If the imaginary part of $g(\omega)$ is not negligible in an experiment, one can use the experiment to determine K .

Since it is most likely that the fast growth limit is realized, we expect that the drastic change of sound velocity and the anomalous Doppler shift occur as the solid thickness is changed. Experimental detection of the latter may not be easy because one cannot have a permanent superfluid flow of large velocity in this system. In the fourth sound experiment of Kojima *et al.* [28] the critical velocity, below which there are no vortices, is about 20 cm/s and the saturated superfluid velocity is about 60 cm/s at 1.3 K for Al_2O_3 powder, of which the grain size is 170-320 Å.

The critical velocity is smaller when the pore size is larger. Although we do not know exactly how high a velocity is attainable in our system, the effect on the Doppler shift is large and even the sign is changed so that its detection would be clear proof of the role of crystal growth. The Doppler shift of the opposite sign is a qualitatively new feature which our present system displays.

Recently Maynard, Jelatis and Roth [15] performed fourth sound resonance measurements with a grafoil ring and found an oscillation of the resonance frequency, which is attributed to layer-by-layer crystal growth. Our theory does not take into account the atomic layer structure of the solid and is not fully capable of giving a quantitative explanation of this phenomenon. Ramesh and Maynard [16] have given an explanation based on the solid growth. Their basic idea seems common to ours. In the case of strong substrate potential our result gives the same expression as theirs for the sound velocity and attenuation. There are two important parameters in their theory (and in our theory, too). One is the equilibrium position of the interface as a function of the pressure $\partial P/\partial\zeta$, which reflects the step-wise layer structure. As our theory is based on hydrodynamics, we have related $\partial P/\partial\zeta$ to the substrate potential by (8) although it is not really necessary. The other parameter is the relaxation time τ in their theory, which is related to the growth coefficient $K(g_2/g_1)$ corresponds to τ . If one introduces oscillatory changes of $\partial P/\partial\zeta$ or K (equivalently τ) due to the layered structure of the solid, one can get oscillatory changes in the sound velocity and attenuation [16].

The temperatures in the experiment are mostly higher than the roughening transition for the bulk system and we expect that the experiment corresponds to the case of fast growth, where decrease of K increases the sound velocity and the attenuation (see Fig. 2). This point is argued by Adler, Kuper, and Schulmann [36]. It seems that in the experiment decrease of the sound velocity and increase of the attenuation occur simultaneously. If that is the case, the effective growth coefficient of this thin layered solid is much smaller than we think. The strong substrate potential (10^{13} times stronger than the gravitational effect) could change the nature of the roughening transition. Sound was not detectable in the experiment when the solid became thick ($\sim 20 \text{ \AA}$). This is about the same thickness as we expect the drastic change. Capillary condensation is one possible explanation [15]. When

the solid is thick ($d - \zeta \gtrsim 30 \text{ \AA}$ in the present case), our « equilibrium » state is only a metastable state and in the ground state the channel is blocked by the solid. However, the metastable state is separated from the ground state by a very high potential barrier, and the observed isocores [22, 24] indicate that effects due to pore condensation are negligibly small. Further experimental studies are desirable in order to clarify these points and see the sound mode discussed here in the case of a thick solid layer.

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Appendix.

We examine the z -dependence of the hydrodynamic equations due to the substrate potential. The situation is similar to that in the third sound problem [37]. The number density n_2 , the superfluid velocity v and the chemical potential are z -dependent, and instead of (1) and (2) we should use

$$\frac{\partial n_2}{\partial t} + \frac{\partial}{\partial x}(n_2 v_x) + \frac{\partial}{\partial z}(n_2 v_z) = 0, \quad (\text{A.1})$$

$$\frac{\partial v_x}{\partial t} + \frac{1}{m} \frac{\partial}{\partial x} \left(\mu_2 + \frac{1}{2} m v^2 \right) = 0, \quad (\text{A.2})$$

$$\frac{\partial v_z}{\partial t} + \frac{1}{m} \frac{\partial}{\partial z} \left(\mu_2 + \frac{1}{2} m v^2 + U \right) = 0. \quad (\text{A.3})$$

In equilibrium the velocity of the steady flow is irrotational and independent of z . The equilibrium number density and pressure are determined by (A3), which leads to (8). The linearized equations of (A1), (A2) and (A3) are

$$\frac{\partial n'_2}{\partial t} + n_2 \frac{\partial v'_x}{\partial x} + \frac{\partial n'_2}{\partial x} v + \frac{\partial}{\partial z}(n_2 v'_z) = 0, \quad (\text{A.4})$$

$$\frac{\partial v'_x}{\partial t} + \frac{c_0^2}{n_2} \frac{\partial n'_2}{\partial x} + v \frac{\partial v'_x}{\partial x} = 0, \quad (\text{A.5})$$

$$\frac{\partial v'_z}{\partial t} + \frac{\partial}{\partial z} \left(\frac{c_0^2}{n_2} n'_2 \right) = 0, \quad (\text{A.6})$$

where all quantities except v are z -dependent and c_0 is determined by the local number density and the local compressibility. Boundary conditions at the interface are (11)

$$\frac{1}{mK} \frac{\partial \zeta'}{\partial t} - \frac{n_1 - n_2}{n_1} \frac{1}{m} \frac{\partial U}{\partial z} \zeta' + \frac{n_1 - n_2}{n_1 n_2} c_0^2 n'_2 + v v' = 0, \quad (\text{A.7})$$

and the continuity equation

$$(n_1 - n_2) \frac{\partial \zeta'}{\partial t} + n_2 v'_z - n_2 \frac{\partial \zeta'}{\partial x} v = 0. \quad (\text{A.8})$$

Assuming the solutions to be proportional to $e^{ikx - i\omega t}$, we eliminate v'_x and v'_z from (A4) to obtain

$$(\omega - kv)^2 n'_2 - k^2 c_0^2 n'_2 + \frac{\omega - kv}{\omega} \frac{\partial}{\partial z} \left[n_2 \frac{\partial}{\partial z} \left(\frac{c_0^2}{n_2} n'_2 \right) \right] = 0. \quad (\text{A.9})$$

Similarly the boundary conditions (A7) and (A8) can be written as

$$\left(i\omega \frac{\zeta}{mKc_0^2} + \frac{n_1 - n_2}{n_1} \frac{\zeta}{mc_0^2} \frac{\partial U}{\partial z} \right) \frac{\omega - kv}{\omega} \frac{\partial}{\partial z} \left(\frac{c_0^2}{n_2} n_2' \right) \Big|_{\zeta} = \left(\omega \frac{n_1 - n_2}{n_2} + kv \right)^2 \Big|_{\zeta}. \quad (\text{A.10})$$

With the obvious boundary condition at $z = 0$,

$$n_2 \frac{\partial}{\partial z} \left(\frac{c_0^2}{n_2} n_2' \right) \Big|_0 = 0. \quad (\text{A.11})$$

the integration of (A9) yields

$$(\omega - kv)^2 \int_0^z dz' n_2' - k^2 \int_0^z dz' c_0^2 n_2' + \frac{\omega - kv}{\omega} n_2 \frac{\partial}{\partial z} \left(\frac{c_0^2}{n_2} n_2' \right) \Big|_z = 0. \quad (\text{A.12})$$

This equation, along with (A10), determines the dispersion relation. For small k and ω ($k\zeta \ll 1$), $c_0^2 n_2'/n_2$ is almost independent of z , and the dispersion relation takes the simple form,

$$\left(\omega + kv \frac{n_2}{n_1 - n_2} \right)^2 + \left(-i\omega \frac{\zeta}{mKc_0^2} + \frac{n_1 - n_2}{n_1} \frac{\zeta}{mc_0^2} \left| \frac{\partial U}{\partial z} \right| \right) \frac{n_1 n_2}{(n_1 - n_2)^2} \left[(\omega - kv)^2 - k^2 \langle n_2 \rangle \left\langle \frac{n_2}{c_0^2} \right\rangle^{-1} \right] = 0, \quad (\text{A.13})$$

where the average values are defined by

$$\langle n_2 \rangle = \frac{1}{\zeta} \int_0^{\zeta} dz n_2, \quad \left\langle \frac{n_2}{c_0^2} \right\rangle = \frac{1}{\zeta} \int_0^{\zeta} dz \frac{n_2}{c_0^2}. \quad (\text{A.14})$$

The other quantities are evaluated at the interface. The result (A13) replaces (12).

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