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Transmission of sound at the liquid-solid interface of helium: a new probe of melting kinetics

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Résumé. — Une onde acoustique produit une discontinuité alternative de potentiel chimique, $\Delta \mu$, lorsqu'elle traverse un interface liquide-solide. Si la fusion est assez rapide, elle peut répondre à une telle excitation. Nous montrons qu’un tel effet existe dans les systèmes quantiques, où la chaleur peut être évacuée par convection. Cet effet peut conduire à une réduction spectaculaire de la transmission acoustique; il permet en outre une conversion du premier son en second son. Nous analysons le phénomène en détail, et nous proposons une expérience qui devrait donner accès à la cinétique de la fusion, dans des conditions où celle-ci est trop rapide pour être observée en continu.

Abstract. — An incoming acoustic wave produces an a.c. discontinuity of chemical potential, $\Delta \mu$, at a liquid-solid interface. If melting is fast enough, it responds to such an excitation. We show that in quantum systems, in which heat can be evacuated via convection, such an a.c. melting does occur. It may give rise to a spectacular decrease in sound transmission, and to a conversion mechanism from first to second sound. We analyse that effect, and we propose an experiment that should give access to the kinetics of melting when it is too fast to be observed by d.c. methods.

In ordinary materials, melting and crystal growth are slow processes on an atomic scale. First of all, one must dispose of the latent heat of transformation, which usually is the controlling factor of the kinetics. Second, the growth itself depends on the nature of the interface [1]. If the latter is sharp and smooth, the nucleation of a new layer is the controlling step; it occurs on surface defects (cf. spiral growth on dislocations) and the process is slow. Fast growth can only occur if the interface is rough, with spontaneous formation of many steps and kinks: then each layer grows continuously. Such a rough surface is signalled by the absence of facets in the equilibrium shape of the crystals.

Quantum crystals are in that respect quite unusual. First of all, heat can be evacuated via heat convections instead of ordinary conduction (in the liquid for $^4$He, in the solid for both $^4$He and $^3$He). In an a.c. regime, a second sound wave replaces the ordinary diffusion process. Such a non dissipative phenomenon is very efficient: it takes care of the heat problem. Second, the interface of solid He is very likely rough, due to the large zero point motion of the atoms [2]. Indeed, the shape of $^4$He crystals seems to be isotrope, with no facets. Thus, fast crystal growth should be possible.

Experimentally, it is known that $^4$He crystallizes very easily. As stated by Simmons [3], it is difficult not to obtain a single crystal. Growth velocities of order mm/s may be achieved without any measurable difference of chemical potentials in the two phases (the relative distance, $\Delta p/p$, to the equilibrium curve in the $p-T$ plane being $< 10^{-4}$). With such velocities, a d.c. study of the kinetics would be difficult. Instead, it appears possible to study the response to an a.c. excitation, one then triggers melting at a finite frequency $\omega$ by modulating the distance to the phase equilibrium curve.

One such experiment was recently proposed by Andreev [2], who discussed the existence of thermocapillary waves at the interface. If a second sound wave is propagated parallel to the interface, the crystals melts in the hot points, freezes in the cold points. Because of the different densities, a coupled flow pattern of matter and energy is generated, in close analogy with capillary waves at the free surface of a liquid.

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We discuss here another possibility: a.c. melting of the interface under the influence of an incoming acoustic pressure wave. If the response of the interface is slow, it cannot follow the modulation of \( p \) : as usual the transmission of sound is controlled by the ratio of acoustic impedances in the liquid and solid phases. If instead melting responds instantaneously to \( p \), the pressure and temperature at the interface should remain on the phase equilibrium curve. At the minimum of that curve, the interface becomes a node of pressure: the transmitted sound wave should then disappear. Away from the minimum, a modulation of temperature ensues: first sound is then reflected partly as second sound. Thus, we expect rather spectacular effects, which should provide a sensitive probe to the kinetics of the phase transformation.

1. Transmission of sound at a liquid-solid interface. — We consider the simplest geometry: a flat interface separates two phases 1 and 2; the normal to the surface is assumed to be a symmetry axis of the crystal, so that longitudinal and transverse waves are decoupled in that direction. A plane acoustic wave at normal incidence is emitted in phase 1: we wish to calculate the reflected and transmitted amplitudes.

In each phase, we neglect the small coupling between mechanical and thermal variables due to dilatation. A pressure wave \( \delta p \) propagating in the positive \( z \)-direction involves a velocity field:

\[
\begin{align*}
u &= \frac{1}{\rho_1 c_1} \delta p = \frac{1}{z_1} \delta p \\
\end{align*}
\]

where \( z_i \) is the acoustic impedance of phase \( i \) (\( \rho \) and \( c \) are the mass density and first sound velocity). Similarly, if it exists, a second sound wave involves a heat current \( J_Q \) and temperature modulation \( \delta T \) such that:

\[
J_Q = \lambda_i \delta T, \quad \lambda = \rho C_v c_{\text{II}}
\]

(\( C_v \) is the specific heat per unit mass, \( c_{\text{II}} \) the second sound velocity). In the absence of second sound, (2) would be replaced by the heat conduction equation:

\[
J_Q = -K \text{grad} T.
\]

What we need is boundary conditions at the interface.

In the absence of melting, \( u \) and \( \delta p \) are continuous at the interface. The amplitude \( R \) and \( \tau \) of the reflected and transmitted pressure waves are then given by the usual formulas

\[
R = \frac{z_2 - z_1}{z_1 + z_2}, \quad \tau = \frac{2z_2}{z_1 + z_2}.
\]

Since \( z_1 \) and \( z_2 \) are comparable, the transmission of sound is large.

We next assume that heat convection occurs only in phase 1. The other phase is a thermal insulator in which \( J_Q = 0 \). Such a situation may apply to liquid \( ^4\)He above 1 K, in a region where second sound does not exist in the solid. Then heat will be evacuated in region 1 only. Besides the reflected and transmitted pressure wave, a reflected second sound wave will appear, with an amplitude:

\[
\delta T = \xi \delta p
\]

\( \xi \) is a conversion coefficient from first to second sound. Let \( J \) be the mass that freezes per unit time and unit area (from phase 1 to phase 2). If \( v \) is the velocity of the interface, mass conservation implies:

\[
J = \rho_2 (u_2 - v) = \rho_1 (u_1 - v)
\]

\[
= \rho_1 \rho_2 \frac{u_1 - u_2}{\rho_2 - \rho_1}
\]

(3)

(all velocities are positive where going from 1 to 2).

In a linear approximation \( J \) is proportional to the difference in chemical potential (per unit mass) between the two phases in contact:

\[
J = k (\mu_1 - \mu_2)
\]

\( k \) plays the role of a transport coefficient between the two phases. We use the standard thermodynamic identity

\[
\delta \mu = \frac{\delta p}{\rho} - s \delta T
\]

and we further note that mechanical equilibrium requires that \( p \) be continuous at the interface. If the Kapitza resistance is small as compared to the thermal impedance fo phase 2, \( T \) is also continuous, and we may write

\[
J = k \left[ \delta p \frac{\rho_2 - \rho_1}{\rho_2} - L \frac{\delta T}{T} \right]
\]

(4)

where \( L = T(s_1 - s_2) \) is the latent heat. Together (3) and (4) replace the condition \( u_1 = u_2 \). We complement them with a thermal balance equation

\[
J_{Q_1} = -L J.
\]

(5)

According to (2), the reflected second sound wave is such that \( J_{Q_2} = -\lambda_1 \delta T \): the problem is closed.

The solution of these equations is straightforward. From (3), (4) and (5) we infer an effective boundary condition for the acoustic wave:

\[
\xi = k \frac{[\rho_1 - \rho_2]}{\rho_1 \rho_2} \left[ 1 + \frac{kL^2}{\lambda_1 T} \right]^{1/2}
\]

(6)

\( \xi \) incorporates all the information regarding melting triggered by the sound wave. The transmission and reflection coefficients are given by

\[
\tau = \frac{2z_2}{z_1 + z_2 + z_1 z_2 \xi}
\]

(7)

\[
R = \tau - 1.
\]
The transfer coefficient from first to second sound is similarly
\[
\eta = \frac{\delta T}{\delta p} = \frac{kL}{\lambda_1} \frac{\rho_2 - \rho_1}{\rho_1 \rho_2} \frac{1}{1 + \frac{kL^2}{\lambda_1 T}}.
\] (8)

Note that similar results would be obtained if the emitters were in the solid phase instead of the liquid or equivalently if heat were evacuated as second sound in phase 2 instead of phase 1: we need only to replace \(\lambda_1\) by \(\lambda_2\) in (6) and (8) (it does not matter which side heat chooses to go).

(7) displays all the qualitative features we expect. If \(\xi\) is small either because melting is difficult or because \(L\) is large — the transmission of sound is not much affected. If on the other hand, \(\xi\) is large, transmission may be reduced dramatically. The maximum value of \(\xi\) one can achieve (if \(k \to \infty\)) is
\[
\xi_m = \frac{\lambda_1 T}{L^2} \frac{(\rho_1 - \rho_2)^2}{\rho_1 \rho_2} = \frac{\lambda_1}{T} \left(\frac{dT}{dp}\right)^2.
\] (9)

\(\xi_m\) is infinite at the minimum of the melting curve: there, as predicted, instantaneous melting yields a zero transmission (the interface being a node of pressure). At other temperatures, \(\xi_m\) should be compared to \(1/\xi_1\); \(\xi_m\) remains large well above \(1\) K: we can therefore expect a sizeable decrease of \(\xi\) in a broad range of temperatures — giving access to the fundamental coefficient \(k\).

Note that the transfer coefficient \(\xi\) may be written simply as:
\[
\xi = \frac{dT}{dp} \xi_m.
\]

The conversion into second sound is quite efficient if \(k\) is large.

2. Energy transport across the interface. — In the preceding discussion, we skipped one difficulty when writing (4): does it make sense to disregard the flow of energy across the interface during melting? The issue is crucial if second sound can propagate on both sides 1 and 2: the way the released latent heat splits between the two phases will depend precisely on the energy balance across the interface.

The pressure is continuous at the interface. Let \(\mu_1, T_1\) and \(\mu_2, T_2\) be the chemical potential and temperature on either side.

The corresponding discontinuities drive the flow of matter and energy. The problem is a standard one in irreversible thermodynamics. We work in a frame in which the interface is at rest (\(v = 0\)). We first write the energy balance when a mass \(dm = J dt\) goes from phase 1 to phase 2. At constant pressure, the heat received from outside by a cylinder of matter sitting halfway across the interface is:
\[
dt | J_{Q_1} + J_{Q_2} = (h_2 - h_1) J dt.
\]

\(h_1, h_2\) are the enthalpies per unit mass of each phase, \(J_{Q_1}\) and \(J_{Q_2}\), the heat conduction current on each side (note the analogy with a Joule Kelvin process). We see that:
\[
J_{Q_1} + J_{Q_2} = J_{E} = J_{E_1} + J_{E_2} = \int (h_2 - h_1) J dt.
\]

(10) expresses conservation of the energy current \(J_E\) across the interface (\(J_E\) contains two parts: a convection term \(Jh\) and a genuine conduction term \(JQ\)).

We next evaluate the irreversible entropy production, \(S\), per unit time. It results from a balance between the entropy received from outside and that stored in the two phases. Thus
\[
\dot{S} = J_{Q_1} \frac{T_1}{T_1} - J_{Q_2} \frac{T_2}{T_2} - J (\mu_1 - \mu_2) T_1 T_2.
\]

Within a linear approximation, \(\dot{S}\) may also be written as
\[
\dot{S} = J_E \frac{\Delta T}{T^2} + J \frac{\Delta \mu}{T},
\] (11)
in which
\[
J_E = J_{E_1} - \mu J = J_{Q_2} + T \delta T
\]
is the energy current measured from the (equilibrium) chemical potential. \(\Delta T = \delta T_2 - \delta T_1\) and \(\Delta \mu\) are the discontinuities across the interface. From (11), we infer the linear Onsager equations describing irreversible transport across the interface
\[
\frac{\Delta \mu}{T} = a J + b J_E,
\]
\[
\frac{\Delta T}{T^2} = b J + c J_E.
\] (12)

Our previous analysis amounted to setting \(b = c = 0\) (and also \(J_{Q_2} = 0\)): our coefficient \(k\) is then \((-1/\alpha T)\). We note that \(CT^2\) is nothing but the usual Kapitza thermal resistance of the interface. The cross coefficient \(b\) corresponds to a Peltier effect for helium atoms.

In order to exploit (12) we note that:
\[
\Delta \mu = \rho_2 - \rho_1 + s_1 \delta T_1 - s_2 \delta T_2.
\] (13)

Moreover, \(J_{Q_1}\) and \(\Delta P_1\) are related by the thermal impedance of the liquid and solid phases:
\[
\begin{align*}
J_{Q_1} &= J_E - J T s_2 = \lambda_2 \delta T_2, \\
\Delta P_1 &= J T s_1 = -\lambda_1 \delta T_1.
\end{align*}
\] (14)

From (12), (13), (14) we may eliminate \(J_E, \delta T_1\) and \(\delta T_2\). We are left with a relationship between \(J\) and
\[ \xi^{-1} = \left( \frac{\rho_1 \rho_2}{\rho_1 - \rho_2} \right)^2 \left[ aT + \frac{-T(s_1 - s_2)^2 + \lambda_1 \lambda_2 b^2 T^3 + 2(\lambda_1 s_2 + \lambda_2 s_1) b T^2 + (\lambda_1 s_2^2 + \lambda_2 s_1^2) c T^3}{\lambda_1 + \lambda_2 - \lambda_1 \lambda_2 c T^2} \right] . \]

\[ (15) \]

Let us consider for instance the simple case described by (6). At low frequency \( \omega \), \( \lambda \) is small : the resulting \( \xi \) is also small, and we recover standard acoustic transmission. Note \( \lambda \approx \xi_m \) has an imaginary part : a.c. melting, albeit small, gives rise to phase lag at reflection. When the frequency is increased, \( \lambda \) becomes larger and larger, (a.c.) melting becomes more efficient, and in the end \( \xi \) will be entirely controlled by the melting coefficient \( k \). A detailed discussion would require more information on \( \lambda \) and \( k \). Here, we only make the qualitative point that in a conduction regime, a.c. melting becomes important at high frequencies \( \omega \) (at which evacuation of the latent heat is no longer a problem). In contrast, in the second sound convection regime, \( \xi \) is frequency independent : the importance of a.c. melting depends only on the dissipative coefficient \( k \).

3. Reflection of a second sound wave. — We may also look at the reverse process : a second sound wave is emitted in the liquid and reflected on the interface. We again assume that phase (2) is a thermal insulator : no heat is transferred to the solid. On the other hand, temperature modulation at the interface may trigger melting [4] : the reflection coefficient of second sound, \( R \), is reduced, while a subsidiary acoustic wave is radiated on both sides, due to the resulting pressure modulation.

Let \( \delta T \) be the temperature modulation at the interface; as before, \( J \) is the mass that goes per unit time and area from phase 1 to phase 2. The net heat influx of the second sound waves to the interface is

\[ J_Q = \lambda_1 \delta T \frac{1 - R}{1 + R} = -LJ \]

\[ (17) \]

\( J \) gives rise to a velocity modulation according to (3), which in turn generates a pressure shift \( \delta p \). Since \( u_1 = \delta p/z_1, u_2 = -\delta p/z_2 \), it follows that

\[ J = -\frac{\rho_1 \rho_2}{\rho_2 - \rho_1} \delta p \frac{z_1 + z_2}{z_1 z_2} . \]

\[ (18) \]

Finally, \( J \) is given by the kinetic equation (4), which closes our set of equations. We easily find

\[ \frac{\delta p}{\delta T} = k \frac{\rho_1 \rho_2}{(\rho_1 - \rho_2)^2} \frac{z_1 + z_2}{z_1 z_2} \frac{1}{1 + \alpha} \]

\[ (19) \]
with
\[ \alpha = \frac{T}{\lambda_1} \left( \frac{dp_1}{dT} \right)^2 \frac{k}{z_1 z_2 + \left( \frac{\rho_1 \rho_2}{\rho_2 - \rho_1} \right)^2}. \]

\( \mathbf{R} \) will be equal to 1 either when melting is slow \((k \to 0)\) or at the minimum of the melting curve \((\frac{dp}{dT} = 0)\). Otherwise, \( \mathbf{R} \) may depart from 1 appreciably and a sizeable fraction of the incoming energy may be converted into first sound (the source of which is the modulation \( \delta p \)).

4. Discussion. — In the absence of a clear picture of melting, any estimate of \( k \) is rather hazardous. It is certainly smaller than an atomic limit, in which the atoms would approach the interface on either side at the sound velocity
\[ k \leq \frac{\rho c}{\mu}. \]

Unfortunately, such a requirement is not very stringent—very likely a gross overestimate. It is thus wiser to resort to experiment, and to measure \( k \) indirectly using (6) and (7).

The simplest experiment is sketched on figure 1. Two transducers are placed above each other in liquid \(^4\text{He}\). An acoustic wave is emitted by the upper one, detected by the lower one. By monitoring the pressure, one can adjust the height of the horizontal liquid solid interface. A comparison of the signals when the receiver lies either in the liquid or in the solid gives access to the transmission coefficient of the interface (defined as the ratio of pressure amplitudes in the liquid and solid phases). The main difficulty is to ensure the alignment of the systems. The parallelism of the two transducers can be checked beforehand. But it is much harder to assess the parallelism with the interface. The latter need not be flat; in principle, its shape depends on the orientation of the crystalline phase. Experimentally, its seems that the interface remains horizontal within experimental accuracy, a sign that the surface is rough (the interfacial energy would then be isotropic, and the shape of the surface should be controlled by gravity, as for a liquid-liquid interface). It is not clear whether the flatness of the surface is sufficient for an acoustic transmission experiment. Before reaching any conclusion, it will be necessary to discuss in detail the dependence on frequency, on transducer size, on the crystal orientation, etc... Another possibility would be to measure the reflection coefficient instead of transmission. By propagating a heat wave along the surface, one can generate the Andreev thermocapillary mode (an a.c. heater on the edge of the vessel should be enough). The resulting oscillation of the interface can be observed by an acousto stroboscopy method, which gives access to the reflection coefficient.

A further difficulty stems from crystal anisotropy. Unless the normal to the interface is a symmetry axis, an incoming normal wave will split in the solid, into a pseudolongitudinal and a pseudotransverse mode. The former will collect most of the energy. Still, its group velocity is not parallel to the wavevector, and the refracted beam can depart from the normal direction. The thickness of solid between the interface and receiver must be kept small in order to guarantee that all the transmitted signal reaches the transducer. Another possibility would be to detect the second sound mode emitted under the impact of the incident first sound wave. The corresponding amplitude also gives access to the kinetic coefficient \( k \). The detection would then require a bolometer that responds to the frequency \( \omega \) of the emitting transducer. It should be noted that our proposed experiment can produce second sound waves with very high frequency, in the

Fig. 1. — Proposed experimental set-up.

Fig. 2. — \( z_1 \xi_m \) versus \( T \), assuming thermal equilibrium in the waves (no ballistic regime). At low temperatures, \( z_1 \xi_m \) is inversely proportional to the minimum transmitted signal.
acoustic range (> 100 MHz) in which ordinary heat sources are of no avail. Such a source of high frequency heat modulation may be of interest as such.

Assuming that the experimental problems are solved, a measurement of \( \tau \) will be significant only if \( \xi \) is large enough. We know that \( \xi < \xi_m \), the maximum corresponding to the limit of small dissipative coefficients \( a, b, c \). In order to achieve a qualitative estimate, we use the expression (9) of \( \xi_m \), valid when heat is evacuated in one phase only, the liquid for \( T > 0.8 \) K and the solid for \( T < 0.8 \) K. In figure 2, we plot \( \xi_m \) \( z_1 \) as a function of temperature for liquid \(^4\)He. We see that in any event the a.c. melting correction to \( \tau \) will be negligible above \( 1.4 \) K. Below \( 1 \) K, the reduction of \( \tau \) can be very spectacular if \( k \) is large enough.

5. Conclusion. — A measurement of acoustic transmission (or reflection) at a solid-liquid helium interface should give access to the melting kinetic coefficient \( k \). One should thus be able to clarify the nature of the interface.

(i) Either the surface is rough : as conjectured by Andreev, the energy of a step on the surface is zero (because of zero point motion). Then there is no problem of nucleation in crystal growth, and \( k \) is large. A large reduction of \( \tau \) as the temperature is lowered should be observed.

(ii) Or the step energy is positive (that might be the case for specific crystal orientations). Then crystal growth should be much the same for helium and for ordinary crystals, proceeding via the side motion of steps. The growth should be slow, yielding a small \( k \). The transmission \( \tau \) should be mostly unaffected by melting, irrespective of the temperature. Note that if the amplitude of the sound wave increases, the discontinuity \( \Delta \mu \) at the interface increases : the growth velocity could eventually become large again, leading to a non linear reduction of \( \tau \).

The experiment provides a new way to probe the kinetics of melting. It applies to quantum crystals where heat is evacuated via convection. It might be of use in ordinary materials at high enough frequencies when the modulation of \( \Delta \mu \) by the acoustic wave becomes large enough.

References

[1] See for example :
[4] In the case of classical solid-melt systems, the triggering of melting by an exponentially decaying thermal wave has been discussed and experimentally used by :
In the case of the \(^4\)He superfluid-gas system the reflection of a second sound wave has been discussed, for example by :
WIECHERT, H., J. Phys. C 9 (1976) 553 whose equations are similar to ours.