

Helium-4 Solid–Liquid Interface Mobility and Ultrasonic Energy Absorption Mechanisms

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The first measurements of acoustic reflection, together with acoustic transmission at the solid–liquid interface of ^4He , are used to determine the total relative acoustic energy absorption at the interface as a function of temperature. Based on these results, a new mechanism for the temperature dependence of the kinetics of growth and interface mobility is proposed.

Crystal growth from its liquid can be described phenomenologically by a relation $v = K \Delta\mu$, where v is the growth rate of the solid, $\Delta\mu$ is the difference in chemical potential between the liquid and the solid, and K is a kinetic coefficient. For a classical system in which the growth process is diffusion and/or nucleation dependent, K increases with temperature, usually according to an Arrhenius law. For quantum solids (such as helium) Andreev and Parshin¹ proposed that crystal growth proceeds in an entirely different way, that the solid–liquid interface has very high mobility, and that at $T = 0$ K the process is continuous and reversible, i.e., without dissipation. Thus at $T = 0$ the coefficient K is infinite; as the temperature increases, thermal excitations in the liquid and in the solid interact with the solid–liquid interface and cause dissipation. Consequently, unlike in classical systems, the coefficient K *decreases* with increasing temperature. This proposal led to a number of experimental and theoretical investigations on helium crystal growth.^{2–11} It was realized,^{3,4} in particular, that a sound wave propagating from liquid to solid could cause rapid freezing–melting when the interface is highly mobile. Thus, pressure changes would be taken up by the advancing or receding interface, where the pressure would be near or at the equilibrium melting value and sound transmission between the two media would be substantially reduced or even suppressed. This reduction in transmission provides a method of studying growth kinetics.

Here we report results of the first measurement of the reflection of sound, as well as an extension to higher temperatures and at a higher

frequency (10 MHz) of earlier results⁴ on transmission of sound across the ⁴He liquid-solid interface.

Helium crystals were grown under constant pressure conditions ($\pm 5 \times 10^{-3}$ bar) from a large pressure ballast, in a cylindrical brass cell, 1.67 cm high and 1.2 cm in diameter. Three pairs of 10-MHz LiNbO₃ longitudinal transducers were used. Each pair of parallel transducers consisted of a generator and receiver for propagating ultrasonic pulses (of $\sim 2 \mu\text{sec}$ duration) respectively along the cylinder (vertical) axis and in two horizontal, perpendicular directions. The crystals were grown slowly, on average 0.5 cm/h or less. During this slow growth, vertical sound transmission, liquid to solid, was monitored by measuring the received signal heights of the first transmitted signal and the two reflected signals from the interface. (See Fig. 1.) The transit times of these signals were determined to $\pm 0.5\%$. The majority of the crystals had growth axes (vertical direction) making angles in excess of 45° to the *c* axis. A crystal grown at 1.07 K (see below) had its growth axis within about 20° of the *c* axis. Ultrasonic amplitudes and tunings were established for the entire experimental run in the liquid, within 20 mK of the melting temperature T_m . A calibration measurement was made in the liquid. In addition, when possible, a similar calibration was carried out in the completed solid. Only "through" transmission, with one sending transducer and one receiving transducer, was used. The observed anisotropies in velocity indicate that single crystals were obtained.

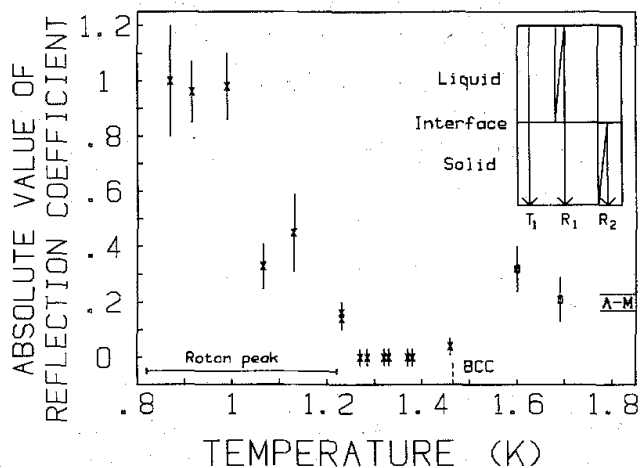


Fig. 1. Sound reflection coefficient for waves propagating from liquid to solid (R_1) as a function of temperature; (X) for hcp, (O) for bcc helium. Inset shows schematically the interior of the cell and labels the wave propagation directions. The horizontal lines marked A-M indicate the range of reflection coefficients expected from acoustic impedance mismatch between the liquid and all crystal orientations of hcp ⁴He.

The amplitudes of the first transmitted signals (T_1 in Fig. 1) with solid filling 10–50% of the cell were normalized to the liquid calibration signal by division. Corrections for signal attenuation in the solid and liquid ^4He and for any focusing effects in the solid were made by determining the effective attenuation from two different heights of solid present in the cell.

The reflection and transmission coefficients (obtained from the relative amplitudes of reflected and transmitted signals) are shown in Figs. 1 and 2 for hcp and bcc crystals grown from the superfluid. First we note the striking result that in the temperature interval $1.25 \leq T \leq 1.38$ K the amplitude of the signal reflected from the interface on the liquid side, labeled R_1 in Fig. 1, is *zero* within the resolution of our observations. At the same time the signal R_2 (reflection on the solid side) is large and generally in excess of the value expected from acoustic impedance mismatch.* Since R_2 is not zero (and is, in fact, larger than the value expected from acoustic impedance mismatch) in the temperature interval where R_1 is zero, the vanishing of the latter cannot be attributed to such artifacts as a solid–liquid interface inclined to the direction of sound propagation. The observed zero value of R_1 is evidently not related to the roton attenuation peak¹² either, since for the ultrasonic frequency of 10 MHz used here this peak occurs at a lower temperature (see Fig. 1). Moreover, these observations are independent of which (top or bottom) transducer is used as a transmitter.

The transmission coefficient for hcp crystals grown from the superfluid (Fig. 2) is found to vary monotonically with temperature and to reach very small (~ 0.1) values below 0.9 K. At any given temperature the relation[†] $R = T_{LS} - 1$ is generally not satisfied. The following exceptions are noted: (1) A crystal grown at 1.07 K displayed substantial agreement, both in reflection and transmission, with acoustic impedance mismatch expectation. We do not have an explanation for this result; we can only conjecture that in this case the solid–liquid interface was “faceted.”[‡] (2) An hcp crystal grown at 1.8 K (from normal fluid) also displayed reflection and transmission properties in accordance with acoustic impedance mismatch. This result is consistent with what is expected when increased latent heat and thermal resistance begin to play a dominant role in the solidification process. (3) The only two crystals of bcc ^4He , grown from the superfluid, had transmission coefficients substantially smaller than expected from acoustic

*In acoustic mismatch theory the transmission and reflection coefficients for a pressure wave originating in medium a and entering medium b at normal incidence are, respectively, $T_{ab} = 2Z_b/(Z_a + Z_b)$ and $R_{ab} = (Z_b - Z_a)/(Z_b + Z_a)$, where $Z_i = \rho_i C_i$ and ρ and C are the mass density and sound velocity, respectively.

†See preceding footnote.

‡A faceted interface is expected to have low mobility and thus transmit sound according to acoustic impedance mismatch. See, for example, Ref. 13.

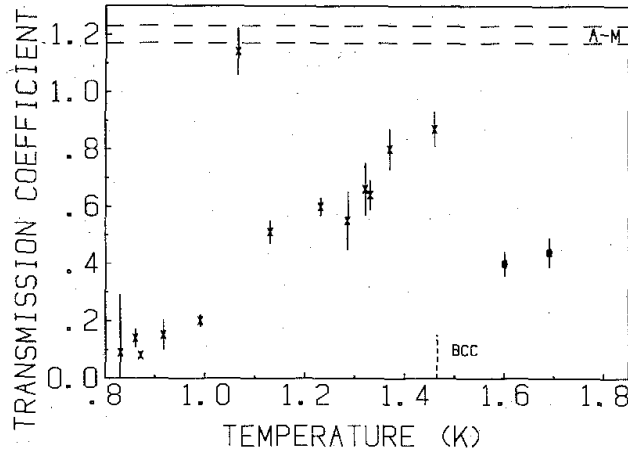


Fig. 2. Sound transmission coefficient for waves propagating from liquid to solid (T_1) as a function of temperature; (\times) for hcp, (\circ) for bcc helium. The horizontal lines marked A-M indicate the range of transmission coefficients expected from acoustic impedance mismatch between liquid and all crystal orientations of hcp ^4He .

impedance mismatch and from extrapolation of values for hcp crystals (see Fig. 2). This case will be treated separately.

In what follows, we confine the discussion to hcp crystals grown from superfluid, excluding the crystal grown at 1.07 K (see above). In examining the data we note that our transmission coefficients (Fig. 2) have values similar to those of Castaing *et al.*⁴ (CBL) obtained at 1 MHz in the temperature range $0.8 \lesssim T \lesssim 1$ K in which the two experiments overlap. We analyzed our transmission results through the expressions developed by Castaing and Nozières³ and used by CBL. In particular, we extracted from our transmission data values of the quantity ξ^{-1} (ξ is the interface mobility) according to Eq. (7) of Ref. 3, i.e.,

$$T_{LS} = 2Z_2 / (Z_1 + Z_2 + Z_1 Z_2 \xi) \quad (1)$$

We note, however, that in deriving³ Eq. (1), $R = T - 1$ was implied and that our data do not satisfy this condition. The behavior of ξ^{-1} obtained here should be considered, therefore, as an indication of a trend, rather than a precise result. With this reservation, ξ^{-1} was analyzed by fitting temperature dependences of the form $\xi^{-1} = AT^4 + \xi_0^{-1} \exp(-T_0/T)$ in an attempt to identify separately phonon and roton contributions,^{2,8} as well as $\xi^{-1} = \xi_0^{-1} \exp(-T_0/T)$, in keeping with the analysis of Ref. 4. The first form yielded a negative constant A , while the second one provided a good fit with $T_0 = 7.5 \pm 0.3$ K and $\xi_0^{-1} = (1.6 \pm 0.5) \times 10^{-6}$ in cgs units. These values are in fair agreement with those of previous studies at lower temperatures and frequencies.^{2,4}

As mentioned previously, our results generally do not satisfy the relation $R = T_{LS} - 1$. Accordingly, we calculated from our data the relative acoustic energy dissipation ε at the interface as a function of temperature, using the energy conservation expression¹⁴

$$\varepsilon = 1 - R^2 - T_{LS}^2(Z_1/Z_2) \quad (2)$$

ε represents all of the acoustic energy dissipation, including that^{3,14} consistent with the condition $R = T_{LS} - 1$, as well as the additional dissipation found when $R = T_{LS} - 1$ is *not* satisfied (i.e. the case of our results). Figure 3 shows a plot of ε (relative acoustic energy loss) as a function of temperature. The shape of this plot is strongly suggestive of a relaxation mechanism. We fitted, therefore, the data to a relation of the form

$$\varepsilon(T) = C \frac{\omega\tau(T)}{1 + \omega^2\tau^2(T)} \quad (3)$$

where C is an "energy relaxation strength," $\tau = \tau_0 \exp(U/T)$, and τ_0 is taken as the reciprocal of the Debye frequency, i.e., $\tau_0 = 1.6 \times 10^{-12}$ sec. The resulting activation energy is $U = 11.52 \pm 0.04$ K and the prefactor $C = 1.520 \pm 0.036$. The curve in Fig. 3 represents this fit.

With regard to the interpretation of our results, we note the following. Previous deduction of interface mobility ξ from the damping of melting-freezing waves² and from acoustic transmission⁴ measurements show that up to ~ 1 K, ξ^{-1} is thermally activated. We obtain similar behavior of ξ^{-1} from our acoustic transmission measurements alone. When we examine our acoustic transmission together with the acoustic reflection (which has not

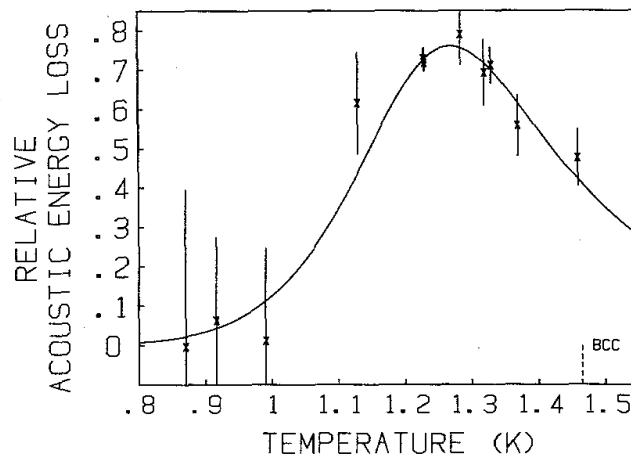


Fig. 3. Relative acoustic energy loss ε [see Eq. (2)] at the solid-liquid interface as a function of temperature. The curve represents a fit of the relaxation function, Eq. (3). See text.

been previously measured) and calculate the total relative energy dissipation ε at the interface as a function of temperature, we find that ε displays relaxational behavior (Fig. 3). We emphasize that the raw data for this energy dissipation are obtained directly from the experimental results using only an energy balance equation [Eq. (2)], without reference to any specific model. This is in contrast to the deduction of the interface mobility ξ discussed above.

In view of these observations, we offer the following interpretation.

The observed energy dissipation is related to a diffusion process at the interface. The activation energy of ~ 11.5 K can be interpreted as the sum of a vacancy formation energy¹⁵ of ~ 8 K and a barrier U_M to vacancy (or atom) motion of ~ 3.5 K. This barrier would result in a vacancy tunneling rate of $\omega_T = \omega_D e^{-\lambda}$, with $\lambda = (d/\hbar)(2mU_M)^{1/2}$ (where d is the barrier width and ω_D is the Debye frequency) and an activated jump rate of $\omega_A = \omega_D \exp(-U_M/T)$. The temperature T_{CR} at which the two rates become equal is ~ 1.3 K, where the mass of a helium atom is used for m , i.e., on the assumption that a vacancy “jump” involves the displacement of a single atom (while this may be an oversimplification for delocalized vacancies in a quantum solid, it should give a reasonable approximation to T_{CR}). The melting-freezing process is thus considered to be limited by the accommodation of thermally generated vacancies. At low temperatures below T_{CR} vacancy migration and rearrangements are governed by (temperature-independent) tunneling. The *decrease* in interface mobility with increasing temperature is thus determined by the increasing vacancy concentration alone, i.e., $\exp(-U_F/T)$, where U_F is the formation energy of ~ 8 K. Above T_{CR} vacancy (or atom) migration is thermally activated with an energy barrier $U_M \approx 3.5$ K. Thus, in this case the interface mobility is limited by concentration and migration of vacancies, with an effective activation energy $U_F + U_M \approx 11.5$ K. The observed relaxational behavior of the relative acoustic energy loss (Fig. 3) is consistent with this interpretation. For consistency with our as well as earlier^{2,4} results, we only have to assume that $T_{CR} \approx 1$ K, rather than ~ 1.3 K estimated above. In view of the simplification used in our estimate, this should certainly not be considered a serious discrepancy. We also note that the location and magnitude of the peak (Fig. 3) are determined primarily by values of ε above ~ 1 K and are insensitive to the values of ε below ~ 1 K, where a change to tunneling is expected.

In conclusion, from the first measurements of sound reflection, as well as transmission, at the solid-liquid interface of ^4He we determined the total, relative acoustic energy absorbed in the melting-freezing process as a function of temperature. On this basis we propose a new mechanism for limiting the interface mobility.

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