# Frequency Dependence of Superfluid Onset in Thin $He^4$ Films*

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Superfluid onset has been measured at several frequencies by a quartz microbalance method. We find that onset occurs earlier (i.e., at smaller thickness and/or higher temperature) when measured at higher frequencies. The results are in agreement with a prediction based upon surface heterogeneity and superfluid connectivity.

Chester, Yang, and Stephens[1] (CYS) have employed a quartz microbalance to study the onset of superfluidity. In contrast to third-sound and persistent-current[3] results where the superfluid density $\sigma_S$ appears to be nonzero at onset, CYS interpret their measurements as indicating that $\sigma_S$ increases rapidly but continuously from zero as the thickness $d$ is increased slightly beyond the critical value $d_c(T_0)$ at the onset temperature $T_0$. The rapid increase of $\sigma_S$ near $d_c$ means that the normal-fluid density is decreasing; thus, if the film is entirely normal at onset, it seems that the addition of more helium causes a significant portion of the original film to convert to superfluid. This implies that contrary to the conventional microscopic theory[4-6] the healing length of the superfluid order parameter at the solid boundary is strongly reduced as the free-surface boundary recedes.

A model involving surface irregularities has been proposed to explain these results.[7,8] It views the adsorbed film as varying in thickness and density as a result of variations in surface binding energy. Near onset superfluidity does not appear uniformly but occurs first in regions of thickness $d \geq d_c$, so that the film forms superfluid regions of various sizes connected by thinner nonsuperfluid film. Under certain conditions some superfluid may be present in the film but not experimentally visible. Different experiments will present different criteria for superfluidity to be observed. In the microbalance experiment, a superfluid region appears normal if its lateral size $l$ is smaller than $\sim C_3/2f$, where $f$ is the microbalance frequency and $C_3$ is the velocity of a superfluid surface wave (third sound) excited by the motion of the superfluid boundaries. The model thus predicts a dependence of the experimental $d_c$ on the frequency of the microbalance: that onset would appear "earlier" (i.e., at higher temperatures or smaller thicknesses) at higher frequencies. We report here on a successful test of this prediction.

The quartz-microbalance technique relies on the decrease in shear-mode resonant frequency, $f_0$, of a quartz crystal due to the mass loading when a thin film is adsorbed on its surface. Under the conditions obtaining in our experiment, the change in frequency $\Delta f$ is given by

$$\Delta f = -2(2f_0^2/C_t\rho_q)\sigma. \quad (1)$$

Here $\sigma$ is the adsorbed mass per unit area which remains rigidly coupled to the surface, $\rho_q$ is the crystal density, and $C_t$ is the shear-wave velocity in the crystal. A factor of 2 occurs to include both crystal faces. The applicability of this method to studies of superfluid onset requires that the viscous wavelength $\lambda = (\eta/\rho_n f)^{1/2}$, using normal-fluid viscosity $\eta$ and density $\rho_n$, must be much larger than the film thickness (as is the case in our experiment). This ensures that the normal fluid moves with the surface and contributes to $\Delta f$. Under these circumstances, one expects that $\Delta f$ at $T < T_0$ will be less than for $T > T_0$. But edge effects and heterogeneity complicate the behavior in the following way. Near onset isolated regions containing finite superfluid fractions appear, whose boundaries are constrained to move with the surface. If the regions are small enough or $f$ low enough, they contribute to $\Delta f$ as if they were completely normal.[9] Such regions remain locked to the shear motion unless they grow to such a size that $l \gtrsim C_3/2f$, at which point the superfluid begins to slip and hence contributes less to the effective shear mass.

To measure any frequency dependence of $T_0$ in an unambiguous manner, the crystal was operated at series resonance in a circuit driven by a stable external oscillator and the complex impedance of the crystal measured by detecting the phase shift of the transmitted signal. When operated near $f_0$, the phase shift is directly proportional to the resonant frequency if $\Delta f/f_0 \ll 1$. Adding mass to the crystal surface changes $f_0$ by an amount $\Delta f$, but the crystal must continue to oscillate at the driving frequency; thus the signal

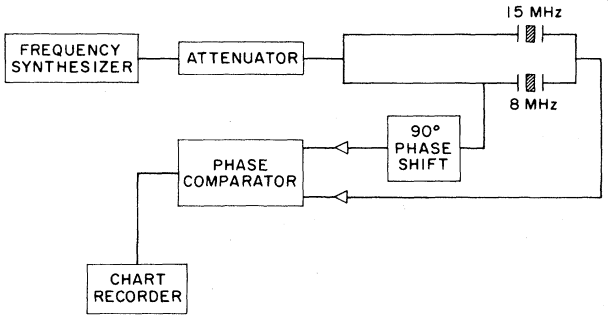


FIG. 1. A schematic diagram of the electrical system used to detect phase shifts across the crystal.

passing through the crystal will be phase shifted by an amount proportional to $\Delta f$. Using this technique, data were taken at different frequencies on the same crystal surface and under the same conditions by simply operating the oscillator at various overtone frequencies of the crystal.

Figure 1 shows a schematic diagram of the phase-measuring apparatus. A frequency synthesizer[10] with a range from 0 to 50 MHz provided a constant-amplitude signal of stability better than 1 part in $10^8$ per hour (at 8 MHz) while the variable attenuator permitted control of the power level reaching the crystal. The crystal was an 8-MHz fundamental mode, commercial, $AT$-cut quartz crystal[11] approximately 0.21 mm thick and 8.0 mm in diameter. The signal from the synthesizer was sampled immediately before and after passing through the crystal and analyzed by the phase comparator to determine the phase shift.

A typical experiment was carried out in the following manner. A known amount of $He^4$ was measured into the crystal chamber and allowed to equilibrate at a temperature slightly higher than $T_0$. The temperature was then reduced slowly while $\Delta f$, $T$, and the film vapor pressure $P$ were recorded. Cooling rates were typically 2 to 8 mK/min. When the temperature was about 0.1 K below $T_0$ the crystal was warmed above the transition and allowed to equilibrate again. The synthesizer frequency was changed to a different crystal overtone and the cooling repeated.

The upper part of Fig. 2 shows schematically a typical frequency trace. As the temperature is lowered, helium condenses out of the vapor onto the crystal causing a decrease in $f_0$ consistent with Eq. (1). Below a certain temperature, $f_0$ begins to increase with decreasing $T$ until a maximum is reached, beyond which $f_0$ decreases again. The curve is retraced when the crystal is

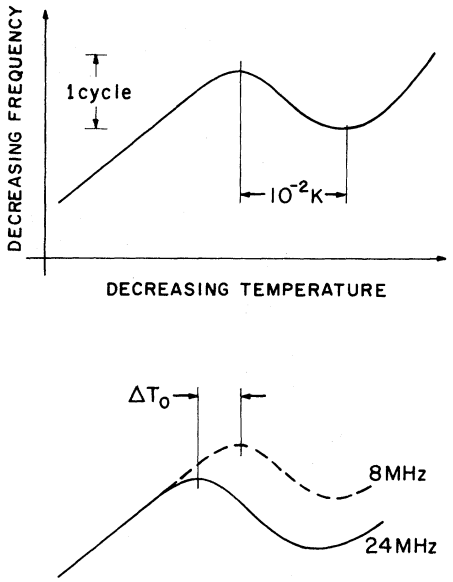


FIG. 2. The upper diagram depicts a typical trace obtained by cooling the film through the superfluid transition. The onset temperature $T_0$ is chosen to be the temperature of the local maximum. The lower diagram shows a superposition of two traces taken under the same conditions but at the fundamental and third resonant shear modes of the crystal.

allowed to warm. We believe the increase in $f_0$ to be due to the appearance of superfluidity in the film and define the onset temperature $T_0$ to be the temperature of the local maximum in the $f$-$T$ trace. This assignment essentially corresponds to that used by CYS; in their study the local maximum was established by varying the thickness at constant $T$. We emphasize that our choice of $T_0$ is made because the maximum is an easily identified and reproducible feature of our data associated with onset, and not because we believe this point on the trace to be the "true" onset of superfludity. In keeping with the discussions of Refs. 7 and 8, we take $T_0$ to be the temperature at which superfluidity is experimentally observable whereas "intrinsic" onset occurs at $T > T_0$.

The lower part of Fig. 2 shows a superposition of two traces taken at different frequencies but with all other conditions the same.[12] A small but significant difference $\Delta T_0$ exists between the values of $T_0$ at the fundamental, 8 MHz, and the third overtone, 24 MHz. This is displayed more clearly in Fig. 3 where $\Delta T_0$ is plotted as a function of $T_0$ for 8 MHz. Except at the highest temperatures the 8-MHz onset always occurred at a lower temperature than that of the 24-MHz onset. In addition, data taken at the fifth overtone, 40

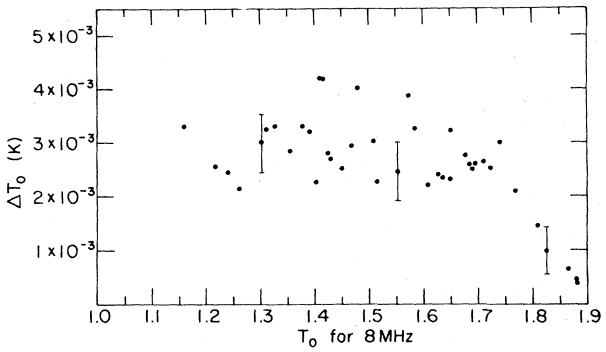


FIG. 3. The difference in onset temperatures $\Delta T_0$. The error bars indicate the uncertainty due to graphically identifying $T_0$ (see Fig. 2).

MHz, showed a $T_0$ higher than that at 24 MHz. However, increased electrical noise prevented much reliable data from being taken at 40 MHz.

Unfortunately, we are unable to draw any quantitative conclusions about the surface morphology based on the data of Figs. 2 and 3, nor do we have sufficient knowledge of the adsorption surface to which we can compare our results. Any conclusions based on Figs. 2 and 3 would require additional measurements at several other frequencies so that homogeneous regions of varying size could be probed. The problem further requires a knowledge of the dependence of third-sound speeds near onset, which is experimentally controversial.[2,13] However, $\Delta T_0$ is expected to go to zero as $T_0 \to T_\lambda$ since in the bulk there is a single transition temperature independent of frequency.[14] Also, as the film becomes thicker it may have smoothed out some of the surface roughness so that the homogeneous regions are large enough to fulfill the onset criteria at both frequencies.

We have considered as a possible alternative explanation of the data the effects of superfluid fluctuations. This can be illustrated by considering the theory of Langer and Fisher.[15] They show that fluctuations which carry the superfluid from one state of superfluid velocity $v_s$ to another of lower $v_s$ occur at a rate given by

$$R = \nu_0 \exp[-E_a(v_s, T)/k_B T], \qquad (2)$$

where $\nu_0$ is some fundamental frequency, $T$ the temperature, $k_B$ Boltzmann's constant, and $E_a(v_s, T)$ the energy required for the fluctuation. For a given experiment, onset will occur at some number of fluctuations depending on the type and sensitivity of the experiment. In our case, the sensitivity to onset is unchanged when the $f_0$ is varied (i.e., the number of dissipative events per cycle is constant); thus $R$ at 24 MHz is three times the rate at 8 MHz. Using this and the form of $E_a$ for a pair of vortex lines perpendicular to the film,[15] we can derive as an expression for $\Delta T_0 = T_{24} - T_8$, the difference in onset temperatures at the two frequencies,

$$\Delta T_0 = [1.024\rho_{24}/\rho_8 - 1]T_8 - 0.543\rho_{24} n, \qquad (3)$$

where $\rho_{24}$ and $\rho_8$ are the onset superfluid densities (in grams per cubic centimeter) measured at 24 and 8 MHz, respectively, and $n$ is the onset film thickness in layers at temperature $T_8$.[17] Substituting into Eq. (3) the results of third-sound experiments[12] for $\rho_{24}$ and our measured values of $T$ and $n$, we find $\Delta T_0 < 0$ over the entire experimental temperature range. In contrast, Fig. 3 shows $\Delta T_0 \geq 0$.[18] If we assume $\rho_{24} = \rho_8$, Eq. (3) also predicts that $n = 1.06 T_8$, when $\Delta T_0 = 0$. This disagrees with the observed values of $T = 1.9$ K and $n = 7.5$ layers.

As a further test, we changed the crystal surface velocity at fixed frequency. It is the nature of Langer-Fisher theory that the onset of detectable dissipation is very sensitive to velocity, so that many experiments appear to detect a critical velocity. Thus $T_0$ should be decidedly more sensitive to velocity than frequency. The crystal power level was changed a factor of 8 above and below its normal operating point of approximately 1 $\mu$W, corresponding to nearly a threefold change in velocity about its usual value.[19] No change in $T_0$ was observed at either frequency. We feel that these results rule out superfluid fluctuations as an explanation.[20]

Conventional explanations of superfluid onset have not considered the possible influence of substrate heterogeneity. This experiment, in confirming the prediction of a frequency-dependent onset, supports the assumptions of the model, i.e., that substrate heterogeneity can play an important role in the manifestations of superfluid onset in many experimental studies.

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[9]When $l \approx \lambda_3$, the third-sound wavelength, the superfluid may oscillate as a standing wave with its center of mass moving further than that of the crystal. In this case, the contained fluid can contribute more (or less) to $\Delta f$ than just its normal mass. The average effect over the entire crystal depends on the distribution of the sizes and shapes of the irregularities and the third-sound attenuation. This nonlinear contribution could affect the shapes and magnitudes of the curves in Figs. 2 and 3, but its contribution depends on little-known parameters and is very difficult to calculate.

[10]Hewlett Packard 5100A Frequency Synthesizer driven by a Hewlett Packard 5110A Synthesizer Driver.

[11]International Crystal Mfg. Co., 10 North Lee, Oklahoma City, Okla. 73102.

[12]Although the crystal environment was the same at 8 and 24 MHz, the film thicknesses were not identical. Since onset at 8 MHz occurred at lower $T$, the pressure was correspondingly lower and the film thicker which enhances the frequency shift.

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[14]We estimate $\Delta T_0 = 0$ at about $n = 7.5$ layers from the Frenkel-Halsey-Hill expression $n = [\alpha\,(T \ln p_0/p)^{-1}]^{1/3}$ where $p/p_0$ is the relative vapor pressure at temperature $T$ and $\alpha$ is a measure of Van der Waals' attraction between the helium and the substrate. See D. M. Young and A. D. Crowell, *Physical Adsorption of Gases* (Butterworths, London, 1962). Since $\alpha$ could not be experimentally determined for our substrate, we employed $\alpha = 39$ (layer)$^3$ K from Ref. 1.

[15]J. S. Langer and M. E. Fisher, Phys. Rev. Lett. 19, 560 (1967). See also J. S. Langer and J. D. Reppy, in *Progress in Low Temperature Physics,* edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Vol. 6, Chap. 1.

[16]In this experiment, the superfluid velocity is taken to be the relative velocity between the superfluid and the crystal surface and hence the velocity of the surface due to the applied electric field. Using the work of C. D. Stockbridge, in *Vacuum Microbalance Techniques,* edited by K. H. Behrndt (Plenum, New York, 1966), Vol. 5, pp. 161–167, we estimate the surface velocity of the crystal at the third overtone to be approximately twice that of the fundamental.

[17]The constants in Eq. (3) depend only logarithmically on the important parameters. Thus any reasonable choice of the parameters changes the constants very little.

[18]Similar conclusions are obtained if other expressions for $E_a(\nu_s, T)$ cited in Ref. 15 are used.

[19]Although the crystal displacement also increases by 3 the change is very much smaller than the lengths, $C_3/2f$, being probed.

[20]The lack of a power-level dependence in $T_0$ also rules out crystal heating as an explanation.

# New Microscopic Approach to the Phonon Dispersion in Ionic Crystals

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A microscopic theory of lattice dynamics of ionic crystals is developed in terms of nonorthogonal highly localized wave functions. It is shown that this approach is in close correspondence to some shell models and leads to a microscopic understanding of the model parameters. The phonon dispersion of LiD has been calculated without using any adjustable parameters and it agrees with experiment to within 10% on the average.

The standard approach to microscopic lattice dynamics[1-5] describes the electronic response to core displacements in terms of equilibrium wave functions. As a consequence the dynamical matrix is obtained as a small difference of two large contributions, namely the electrostatic interaction between bare cores and the electronic contribution of the valence electrons. From a computational point of