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Direct Observation of Third-Sound Mass Displacement Waves in Unsaturated Superfluid ^4He Films

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We report direct observation of pulsed third-sound film thickness oscillations in unsaturated superfluid ^4He films using a capacitance technique. Simultaneous measurements of accompanying temperature oscillations allow the first study of the relative amplitudes of these oscillations and the results are in reasonable agreement with the theory of Bergman although some discrepancies are noted.

Third sound in superfluid ^4He films was first observed by Everitt, Atkins, and Denenstien¹ through the use of an optical technique. Unsaturated film films have been studied extensively through the use of superconducting thin film bolometers pioneered by Rudnick *et al.*² The bolometer records the passage of a third-sound wave by allowing one to observe the associated temperature oscillation. We have developed a capacitance technique which allows us to directly measure the film thickness oscillations³ associated with pulsed third sound in unsaturated superfluid ^4He films. Simultaneous measurements of both the thickness and temperature oscillations allow the first comparisons of the ratio of the amplitude of these oscillations to theoretical predictions. Our observations are in reasonable agreement with the theory of Bergman.⁴

A schematic representation of the apparatus is presented in Fig. 1. Plate A has evaporated on it four aluminum superconducting bolometers, Nos. 1-4, ($250\ \mu\text{m} \times 2.0\ \text{cm} \times \sim 50\ \text{nm}$) and capacitor plates, ($640\ \mu\text{m} \times 2.0\ \text{cm} \times \sim 100\ \text{nm}$). The other plate, B, has a silver capacitor ground plane and a silver strip heater, H, evaporated on it. The separation between the two glass plates is $7\ \mu\text{m}$ and this is maintained constant by means of tiny Stycast 2850GT epoxy beads at the corners. Electrical contact is made to the various elements by means of silver contact strips evaporated from the ends of the elements in question, around the edges of the glass plates and onto the back surface. Fine copper wires are then attached to these back surface contact strips using pure indium solder. The entire assembly is mounted at the end of the cryostat in a chamber (which can be sealed by means of a superfluid valve⁵) which is in turn in a second chamber.

The sensitivity of the bolometers to temperature changes is calibrated at the particular operating bias current and magnetic field by measuring the resistance versus temperature. The re-

sulting curve is smooth in the region of our temperature oscillation measurements with a typical slope $\Delta R/\Delta T \approx 3000\ \Omega/\text{K}$. The capacitors are each part of the tank circuit of two separate tunnel diode oscillators which operate near 20 MHz. The capacitor sensitivity is calibrated to be $\sim 50\ \text{Hz}/\text{nm}$ by filling the gap with pure ^4He . By use of a phase lock loop arrangement we have utilized this sensitivity in the study of third-sound pulses of period, τ , as small as $5 \times 10^{-5}\ \text{sec}$.

Third-sound pulses are generated in the apparatus by application of a single-cycle, positive-going, offset sine wave, voltage pulse of frequency ν , between 0.5 and 5.0 kHz to either the heater, H, or one of the bolometers (typically No. 4). Drive power is typically $2\ \mu\text{W}$ at a repetition rate which can be varied between 30 and 100 Hz. Signals detected on either the bolometers or the capacitors are then signal averaged to improve the signal-to-noise ratio.

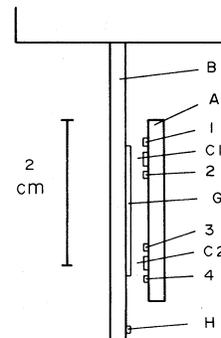


FIG. 1. Schematic representation of the apparatus. The glass plates A and B extend 2.5 cm into the page. The plate spacing and the size of the evaporated strips has been exaggerated for clarity. Strips 1-4 are superconducting bolometers, H is a heater and G a ground plane which serves as one plate of capacitors C1 and C2. Not shown are the epoxy beads which maintain the separation. The assembly is suspended from the base of a recirculating ^3He refrigerator which was not used for the measurements described here.

An example of an event typical of some of our observations is shown in Fig. 2 for a helium film at 1.30 K with a thickness⁶ of 13 atomic layers (1 layer = 0.36 nm). The application of a drive pulse is observed to cause an increase (decrease) in temperature (thickness) followed by a decrease (increase) in temperature (thickness) to be propagated to the bolometer (capacitor) by the film. This observation is consistent with previous *temperature* oscillation measurements by others^{7,8} on films of similar temperature and thickness. The velocity of propagation can be determined from the time of flight of either the thickness oscillation or the temperature oscillation. The third-sound velocity deduced from these two separate time-of-flight measurements is the same and hence, the observed phase difference between the temperature and thickness oscillation is $\sim \pi$ in agreement with predictions based on the detailed theory of Bergman⁴ applied to this case. Measurements of the third-sound velocity as a function of film thickness⁶ at 1.69 K (for example) yield third-sound velocity values which average 2% (thin films) to 10% (thick films) below expectations.⁷ We do not consider this deviation significant.

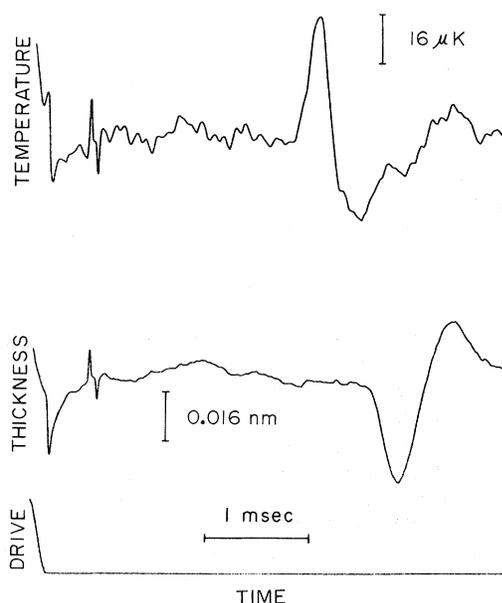


FIG. 2. Bolometer (top) and capacitor (middle) response to a $\nu = 3$ kHz drive pulse (only half of which is displayed). The capacitor signal is displaced to the right because the capacitor is farther downstream from the driver than is the bolometer. Here $T = 1.30$ K and the film thickness $d \approx 13$ layers.

The presence of detectors sensitive to both temperature and thickness changes allows us to measure the relative amplitude, $|\Delta T/\Delta d|$, of these oscillations for the first time. Consistent $|\Delta T/\Delta d|$ measurements can be made when either bolometer No. 4 or heater H is used as a driver. This is because a clear third-sound signal can be seen on bolometers No. 1 and No. 2 in each case. We thus observe the presence of essentially equal amplitude third sound on the substrate opposite the one on which the third sound is generated. The thermal mean free path in the vapor is about $35 \mu\text{m}$ under our typical operating conditions and since the plate spacing is $7 \mu\text{m}$ we interpret our observations to mean that the existence of a third-sound wave on one substrate induces a twin wave on the other substrate.⁹ As a consequence the capacitor responds to two waves rather than one. We have used this interpretation in the analysis leading to the values of $|\Delta T/\Delta d|$ we now present.

In Fig. 3 we show the results of $|\Delta T/\Delta d|$ measurements as a function of the square of the ob-

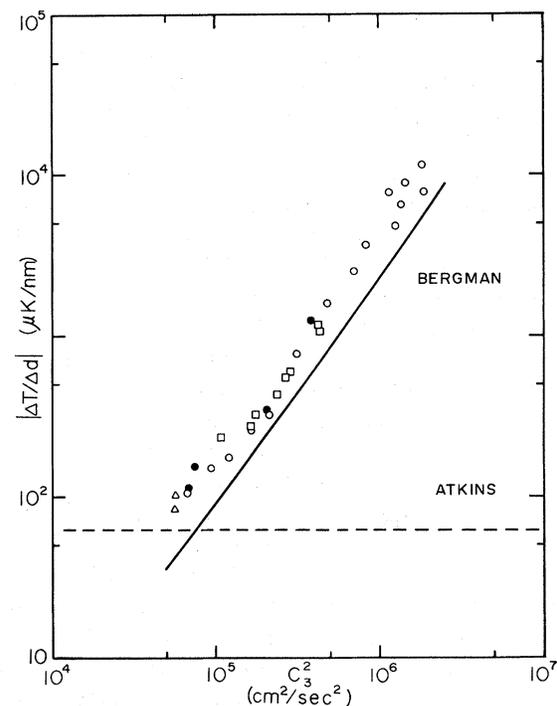


FIG. 3. Magnitude of the relative amplitude of the temperature oscillation to the thickness oscillation at 1.35 K vs the square of the observed third-sound velocity. The various symbols correspond to various experimental runs with $\nu = 1$ kHz. Near $|\Delta T/\Delta d|$ values of $10^3 \mu\text{K}/\text{nm}$ we expect that our calibration errors are about $\pm 20\%$. At the extremes of the data these errors may be as much as a factor of 2 larger.

served third-sound velocity at 1.35 K. Bergman's theory predicts that

$$\Delta T/\Delta d = -(fT/L)(1 - i2\pi\nu\xi), \quad (1)$$

where f is the van der Waals force, T the temperature, L the latent heat, ν the third-sound frequency, and ξ a complicated temperature- and thickness-dependent parameter. For the measurements we report here $2\pi\nu\xi \ll 1$. To obtain the solid curve on the figure we have computed the coordinate pairs $(C_3^2, |\Delta T/\Delta d|)$ for numerous arbitrary values of the film thickness, d , by use of Eq. (1) and^{4,10}

$$C_3^2 = (\langle \rho_s \rangle / \rho) f d [1 + TS/L]^2, \quad (2)$$

where⁷ $f = \alpha\beta(3\beta + 4d)/[d^4(d + \beta)^2]$. Here $\beta \cong 41.7$ layers,⁷ α is the van der Waals force constant and the reduced superfluid fraction $\langle \rho_s \rangle / \rho = (\rho_s / \rho)(d - D)/d$, where¹¹ $D = 0.5 + 1.13(T/T_\lambda)(\rho/\rho_s)$ and d is the film thickness. For the dashed line we have taken $|\Delta T/\Delta d| = 2\pi\rho S\nu T/KL$ from the Atkins¹² theory where $K = \gamma[M/(2\pi k_B N_0 T)]^{1/2} dP/dT$. Here S is the entropy, $\gamma \cong 1$, M is the molecular weight of helium, k_B is the Boltzmann constant, N_0 is Avogadro's number, and dP/dT is the slope of the saturated vapor pressure curve. The Atkins' expression is independent of d and hence for a measurement at fixed temperature also independent of C_3^2 .

Our measurements are seen to have the general dependence on C_3^2 predicted by Bergman⁴ although they are larger by a factor of approximately 1.9. A number of possibilities might account for this discrepancy. The Bergman theory predicts that for the temperature and film thickness range of our measurements $|\Delta T/\Delta d|$ should be independent of the third-sound frequency. We observe instead a frequency dependence with $|\Delta T/\Delta d|$ rising by a factor of 1.8 as the third-sound frequency is varied from 500 Hz to 5 kHz at 1.3 K. Similar behavior is observed at other temperatures. It is also possible that the observed twin wave is somewhat smaller than the identical twin used for the analysis; however, our detailed measurements argue against a large difference in amplitude between the waves. In any comparison between the Bergman theory and our measurements it should be kept in mind that the prediction for $|\Delta T/\Delta d|$ is based on the consideration of a helium film on a single-material substrate which is bounded above by an infinite vapor. This is substantially different from the geometry used for the present measurements.

Finally, we have observed that in thicker films

and at lower frequencies the third-sound pulse shape is different from the shape (e.g., Fig. 2) observed in thinner films at higher frequencies. This difference is observed to be a reduction (enhancement) of the leading (trailing) temperature crest (trough) with equivalent differences observed in the thickness pulse shapes. This qualitative difference is unaffected by changes in the drive power from 0.5 to 10.0 μ W.

In summary, we have used capacitive detection of third-sound pulses in conjunction with bolometer detection to observe for the first time the relative amplitudes of the temperature and thickness oscillations which are third sound. General agreement with the predictions of Bergman is found although discrepancies exist. The advantages of the capacitance detection of third sound include the complete freedom of the choice of the temperature at which the experiments are carried out. Thus, for example, the technique should be useful at extremely low temperatures where the more usual bolometer techniques fail. In such a case a capacitive drive rather than a thermal one may be desirable.

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⁴D. J. Bergman, *Phys. Rev. A* **3**, 2058 (1971), and **188**, 370 (1969).

⁵R. B. Hallock, *Rev. Sci. Instrum.* **43**, 1713 (1972).

⁶In principle the film thickness can be measured directly with the capacitors. We did not do this due to long-term drifts in the tank circuit resonance frequency. These long-term drifts had no effect on the transient behavior. The thickness values we report were determined from measurements of the relative pressure, ΔP , between the experiment chamber pressure, P , and the pressure, P_0 , of the saturated vapor at the chosen operating temperature through use of $\beta^{-1}d^4 + d^3 + 27T^{-1}/\ln(P/P_0) = 0$ where $\beta = 41.67$ layers and d is the film thickness.

⁷See, for example, S. J. Putterman, *Superfluid Hydro-*

dynamics (North-Holland, Amsterdam, 1974), Chap. 5.

⁸Ref. 1 and also K. R. Atkins and I. Rudnick, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Vol. 6.

⁹The epoxy beads used to maintain the capacitor gap could also couple the third sound on the two plates. Because of the small bead size and the large size of the

bolometer signals, we are convinced that this is not the primary cause of the twin wave.

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Dynamical Calculations of Low-Energy Electron Diffraction for Incommensurate Lattice Structures—Xe on Ag(111)

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Dynamical calculations of low-energy-electron-diffraction intensity-voltage curves are carried out for the first time for the case of an incommensurate overlayer-substrate system, Xe-Ag(111). A new scheme was developed to take into account that the substrate (or overlayer) has incident upon it electron beams at angles unrelated to those angles into which the substrate (overlayer) itself diffracts beams. Comparing the calculated curves with experiment, a Xe-Ag(111) spacing of $3.55 \pm 0.1 \text{ \AA}$ is determined.

Dynamical calculations of low-energy electron diffraction (LEED) has, in the past, succeeded in determining the surface structures of a number of metals, semiconductors, and ordered overlayer systems.^{1,2} However, up to now, no successful dynamical calculation has been made on systems where an ordered overlayer is out of registry with the substrate lattice, i.e., the overlayer and substrate lattices are incommensurate with each other. A system of this kind of particular interest is the Xe monolayer on Ag(111).³⁻⁵ The system involves the physisorption of a noble gas on a transition metal substrate. Many studies have been made on surface interaction forces of such systems,^{6,7} but there is little detailed knowledge of atomic spacings at the surface.

Extensive LEED experimental data have been gathered on the Xe-Ag(111) system. Chesters, Hussain, and Pritchard⁵ studied this system at liquid nitrogen temperatures. They established that the Xe monolayer forms a hcp lattice on the Ag(111) substrate. Webb and co-workers^{3,4} recently made extensive measurements of intensity-voltage (*IV*) curves at 25 K. From the two-dimensional spot pattern, they determined that the Xe lattice is out of registry with the Ag substrate, requiring either a contraction of 2.8% or an expansion of 12.3% (plus a 30° rotation) to become

integrally related to the substrate.³

An important structural parameter is the inter-layer Xe-Ag distance, a distance determined by adsorbate-substrate interaction forces. From LEED, this distance can only be determined by analyzing intensity-voltage data.^{1,2} We have carried out a dynamical calculation of *IV* curves for the Xe-Ag(111) system. Because the lattice is incommensurate, Xe atoms take a continuum of lateral positions relative to the Ag unit mesh and this causes a number of problems which require special treatment in the theoretical method. In a LEED calculation of integrally related overlayer-substrate systems, the beam set of the substrate is always a subset of that of the overlayer. Multiple scatterings between the overlayer and substrate simply redistribute electron amplitudes among a finite set of beams.⁸ In the case of an incommensurate overlayer-substrate system, there are two entirely independent, unrelated beam sets. The fact that the overlayer beams, \vec{g} , are unrelated to the substrate beams, \vec{G} , [except, of course, for the (00) beam], implies that the substrate has incident on its surface beams at angles *unrelated* to those angles into which the substrate itself diffracts beams. The layer scattering matrices normally calculated in LEED theory⁸ do not include the proper matrix elements