THIRD SOUND IN ADSORBED SUPERFLUID FILMS*

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It is the purpose of this Letter to report a new method of generating and detecting third sound and to present some results for unsaturated superfluid films.

Third sound is a surface wave on a thin superfluid helium film whose existence was first pointed out by Atkins.[1] For temperatures well away from the lambda point its velocity, C_3, is given by[2]

$$C_3 = [fd\rho_S/\rho]^{1/2}, \qquad (1)$$

where ρ_S/ρ is the superfluid fraction, f is the van der Waals force per unit mass, and d is the thickness of the film.[3] The superfluid fraction appears in Eq. (1) because only the superfluid moves, the normal fluid being locked by viscous forces.[4]

Previous third-sound measurements,[5] which have been restricted to saturated films, 200-900 Å thick, utilized a thermo-optical source and an optical interference system which was sensitive to the height changes in the wave. The system reported here utilizes an electrical current in a deposited metallic film as a thermal source and the receiver, a thin aluminum film, responds to the temperature changes in the wave.

The source-receiver combination is a pair of identical parallel (about 1 cm apart) films of aluminum deposited on a glass plate, which are sufficiently thin that their superconducting transition temperature significantly exceeds the bulk value.[6] The approximate dimensions of each film were a few hundred angstroms thick, 0.02 cm wide, 2 cm long. Current of the desired wave form is fed into the source strip. The resulting heat input causes a flow of superfluid to the source thus generating a surface wave. The receiver strip is operated in its superconducting-transition region with a proper amount of dc current. Characteristically the V-I curves have regions where the slope is abnormally great, and these are chosen for the operating points since it is essential that the effective resistance be a very strong function of temperature. The variations in voltage across this strip are then indications of the temperature variations.

Velocity determinations are based on the time of flight between source and receiver of signals generated by single-cycle sine wave bursts. A characteristic oscilloscope display is shown in Fig. 1, where it is seen that the difference in wave form between the input signal (upper trace) and the received signal (lower trace) reflects the fact that the superfluid flow toward the source is determined by the square of the input current. The time of flight is taken to be the time difference between the initial peaks in both waves.

Figure 2 shows the third-sound velocity at 1.5°K as a function of P_0-P, where P_0 is the saturated vapor pressure and P is the pressure of the gas in the closed can containing the third-sound unit. At the lowest pressures (thinnest films), signals could be seen but could not be

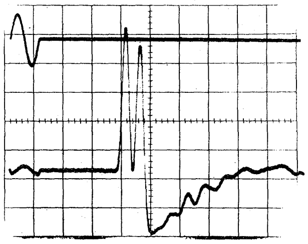

FIG. 1. Top trace is voltage across metallic source film; single cycle period 0.2 msec. Bottom trace is voltage across superconductor receiver film. Temperature increase is up. Source-receiver distance is 1.1 cm. $C_3 = 1445$ cm/sec. An interesting subsidiary feature of the received signal is the fact that the temperature peaks are followed by a time interval in which the temperature is less than ambient. [Second-sound pulses are almost identical in appearance. The fact that cooling occurs in the trailing end of the pulse is an analog of the rarefaction, in ordinary sound, which develops in the trailing end of an initially wholly positive pressure pulse. See Lord Rayleigh, Theory of Sound (The Macmillan Company, Ltd., London, England, 1929), Vol. 2, p. 101.]

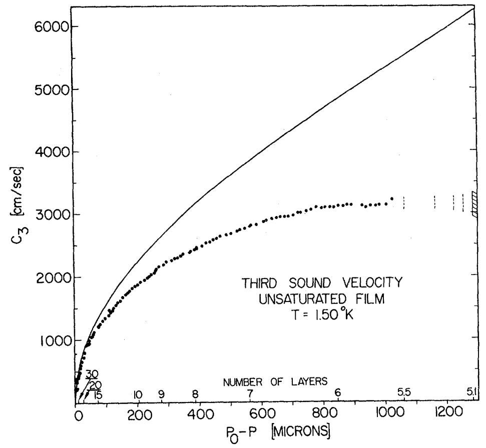

FIG. 2. The points are experimental determinations of C_3 obtained by measured time delays as in Fig. 1. The short dashed lines indicate that signals were visible but could not be measured accurately. The dashed area at $P_0-P = 1290\ \mu$ indicates where all signals disappeared and is taken to be the point at which superfluidity disappears. P_0-P is the difference between vapor pressure at 1.5°K and the pressure of the adsorbed gas. Also shown is the thickness of the adsorbed film in atomic layers.

accurately measured, and this situation is denoted by the dotted lines in the region $1000\ \mu < P_0-P < 1250\ \mu$. Finally, at $P_0-P = 1290\ \mu$ there is no evidence of any signal and this is indicated by the lined area. The signal disappearance is interpreted as due to the disappearance of superfluidity; thus, one obtains a superfluid onset temperature of 1.5°K for $P/P_0 = 0.65$ which is in very good agreement with values obtained by other methods.[7]

If the van der Waals potential is given by $-\alpha d^{-n}$, then the requirement that the thermodynamic potentials in the film and vapor are equal gives

$$\alpha d^{-n} = \frac{RT}{M} \ln \frac{P_0}{P}, \tag{2}$$

where R is the gas constant and M the molecular weight. Substitution in (1) gives[8]

$$C_3{}^2 = \frac{\rho_s}{\rho} \frac{nRT}{M} \ln \frac{P_0}{P}. \tag{3}$$

The weight of experimental evidence and theory indicates that $n = 3$ for thin films, and the value of C_3 calculated from Eq. (3) using this value is shown by the curve of Fig. 2. Also shown on the abscissa are the film thicknesses in number of atomic layers, x, calculated from Eq. (2). Recent data[9] give an assignment of α and single layer thickness which, when put in Eq. (2), leads to the expression $x^3 = 87/T \ln(P_0/P)$.

The reduction of the superfluid fraction due to size effects has been measured in packed powders using fourth sound.[10] The increasing discrepancy between the observed and theoretical values of C_3 shown in Fig. 2 is believed to be due to the continued reduction in superfluid fraction as the film thickness decreases.[11] On the basis of limited knowledge of the pore size in the packed powder, the reductions in the superfluid fraction are comparable in the two cases when the average pore radius and the film thickness are comparable.

Measurements have been made at frequencies up to 30 kHz, this limit occurring because the wavelength becomes approximately equal to the width of the aluminum strips.

Attenuation coefficients have also been measured. As representative values we can quote values ranging from 0.3 cm^{-1} at 1 kHz to 1.8 cm^{-1} at 20 kHz for a film thickness of six atomic layers.

Measurements have been made at several temperatures close to 1.5°K by properly adjusting the operating current to get the required transition temperature. In addition we have made extensive measurements in saturated films at approximately 1.5°K which are in substantial agreement with those of Everitt, Atkins, and Denenstein.[5]

It is clear that third-sound measurements can be useful in measuring the magnitude of size effects in superfluidity and superfluid film thicknesses. There is promise in its use for determinations of critical velocities, persistent currents, and quantum coherence effects in thin helium films.

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[1] K. R. Atkins, Phys. Rev. 113, 962 (1959).

[2] A more complete expression is

$$C_3 = \left[\frac{\rho_s}{\rho} f d \left(1 + \frac{TS}{L}\right)\right]^{1/2},$$

where T = temperature, S = entropy, and L = latent heat. At 1.5°K, $TS/L = 0.013$ and may be neglected for present purposes.

[3] The ordinary-fluid analog of third sound is the shallow water wave whose velocity is $(gd)^{1/2}$, the body force in this case being the gravitational force per unit mass, g.

[4] The thickest films of interest are 900 Å. At 1.5°K the viscous diffusion length exceeds this thickness, and the condition for locking is met for frequencies less than 3×10^6 Hz.

[5] C. W. F. Everitt, K. R. Atkins, and A. Denenstein, Phys. Rev. **136**, A1494 (1964).

[6] M. Strongin, O. F. Kammerer, and A. Paskin, Phys. Rev. Letters **14**, 949 (1965).

[7] See K. Fokkens, K. W. Taconis, and R. de Bruyn Ouboter, Physica **32**, 2129 (1966), esp. p. 2136; or in *Progress in Low Temperature Physics*, edited by C. J. Gorter (John Wiley & Sons, New York, 1967), Vol. 5, p. 63.

[8] It is interesting that the third-sound velocity depends only on the exponent of the thickness in the van der Waals potential and not on the quantity α. A similar situation exists for saturated films (Ref. 5).

[9] W. D. McCormick, D. L. Goodstein, and J. G. Dash, "Adsorption and Specific Heat Studies of Monolayer and Sub-Monolayer Films of He^3 and He^4," to be published. This is based on adsorption isotherms obtained on an argon-plated copper substrate. The results for an aluminum substrate give $x^3=98/T\ln(P_0/P)$; see R. Bowers, Phil. Mag. **44**, 485 (1953).

[10] I. Rudnick, E. Guyon, K. A. Shapiro, and S. A. Scott, Phys. Rev. Letters **19**, 488 (1967); D. F. Brewer, G. W. Leppelmeier, C. C. Lim, D. O. Edwards, and J. Landau, Phys. Rev. Letters **19**, 491 (1967).

[11] Much of the impetus for this work comes from the fact that thin films have a well-defined geometry and accordingly are free of the defect possessed by helium-filled small pores that size effects are statistical.

APPARENT VIOLATION OF GRIFFITHS' INEQUALITY FOR He^4

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A reanalysis of the data used to determine the critical exponent δ given by Roach and Douglass in a recent Letter shows that the value of this exponent is significantly larger than the value they obtained and satisfies Griffiths' inequality.

Recently, Roach and Douglass[1] presented experimental measurements of the isotherms of He^4 near the critical point which purported to show that the critical exponent, δ, caused Griffiths' inequality[2] to go the wrong way. The purpose of this Letter is to point out that the value of δ which one obtains from these measurements is actually compatible with Griffiths' inequality when due allowance is made for the fact that none of the isotherms measured was precisely the critical isotherm. This experimental limitation was pointed out by Roach and Douglass in their Letter but, as we shall show, it makes the determination of δ more difficult and the data less reliable than they originally appeared to be.

The key to the difficulty of interpreting the data lies in the (incorrect) characterization of isotherms above the critical point by the equation

$$\Delta P = A|\Delta\rho|^{\delta}\,\mathrm{sgn}(\Delta\rho) + B(T)\Delta\rho. \qquad (1)$$

Such an equation implies that there is a line of singularities in the pressure-density plane above the critical point. This is in disagreement with the observed qualitative behavior of thermodynamic functions in the neighborhood of the critical point as well as with the theories of Griffiths and others[3] which explain these qualitative features. According to these theories, the appropriate thermodynamic potential (in this case, the Helmholtz free energy) is an analytic function of both its arguments except on the coexistence curve and at the critical point. Thus, one should write ΔP as a double Taylor series expansion in $\Delta\rho$ and ΔT ($=T-T_c$). Such an expansion would be very unwieldy as the critical isotherm was approached since all the terms become comparable in size because of the singularity at the critical point. However, one *can* write[4]

$$\Delta\mu = v_0\Delta P + (\Delta P)^2 f(\Delta P) + C|\Delta P|^{1/\delta+1} + D|\Delta P|^{\epsilon}(\Delta T) + \cdots, \qquad (2)$$

where $\Delta\mu$ is the deviation of the chemical potential from its value at the inflection point of the P-ρ isotherm and $f(x)$ is analytic at $x=0$. C and D are constants and δ and ϵ are the critical exponents associated with the critical isotherm. From Eq. (2), we immediately get

$$|v-v_0| \equiv |\Delta v| = \left|\Delta P g(\Delta P) + E|\Delta P|^{1/\delta} + F|\Delta P|^{\epsilon-1}(\Delta T) + \cdots\right|, \qquad (3)$$

where E and F are constants equal to $(1/\delta+1)C$ and ϵD, respectively, and again $g(x)$ is regu-