[5]J. V. Sengers, Ber. Bunsenges. Phys. Chem. 76, 234 (1972).

[6]In any actual system Eq. (2) would include a background term as well as the divergent term shown; this would change the conclusions in the body of the paper quantitatively but not qualitatively.

Domain of Two-Dimensional Excitations in Superfluid Helium Films*

T. E. Washburn, J. E. Rutledge, and J. M. Mochel†
Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61820
(Received 31 July 1974)

Using third-sound resonance, we find for helium film thickness less than 2.8 atomic layers that there exist excitations which are phononlike and two dimensional, obeying a T^3 law, that their areal density is independent of film thickness, and that their velocity (using Landau theory) is 76 ± 2 m/sec. This velocity is slightly above the maximum third-sound velocity, 69 m/sec, that can be attained by ^{4}He on argon. For thicker films the areal density of these excitations becomes smaller.

A central problem in the study of thin ^{4}He films is whether "two-dimensional superfluidity" is simply a limiting case approached as film thickness and hence T_c vanish[1] or a domain of film thickness with finite T_c's.[2] Our interpretation of measurements given below is that there does exist such a domain.

The measurements of Pobell *et al.*[3] have demonstrated that reducing the dimensionality by confining helium in small pores has a profound effect on the magnitude and temperature dependence of the excitation density. Using the Landau quasiparticle theory for reduced dimensions, Padmore[4] and Haug[5] have provided a framework to understand this class of experiments. We believe that the measurements presented below provide an accurate characterization of these size effects for two dimensions.

In the first part of the analysis we will characterize our helium films by a superfluid film thickness through an analysis similar to the work of Scholtz, McLean, and Rudnick.[6] Next, we will analyze the temperature dependence of the third-sound velocities to produce an areal excitation density. Finally we will try to fit these results with the various Landau quasiparticle models.

Third-sound resonance[7,8] can produce measurements of third-sound velocity C with 1 part in 10^5 resolution. Fortunately, for the films described here, the linewidths are often less than 10^{-4} of the resonant frequencies (which range from 400 to 1800 Hz). The resonator consists of two thin sheets of quartz flame-sealed together. The substrate is a 15-atomic-layer film of crystallized argon which has sublimed just below liquid-nitrogen temperature. The argon gas is permanently sealed within the quartz resonator. Any amount of ^{4}He may be diffused into the resonator at room temperature with an 8-h time constant. Capacitor plates within the resonator provide a means of measuring the gas pressure, to a resolution of 2×10^{-4} Torr, through the flexing of the resonator walls. Total film thicknesses are established by comparing the chemical potentials, as measured by gas pressure, of various film thicknesses instead of by using the vapor pressure of saturated ^{4}He:

$$T \ln\left(\frac{P'}{P}\right) = \frac{\Gamma(d)}{d^3} - \frac{\Gamma(d')}{d'^3}. \quad (1)$$

P is the pressure well above a film of thickness d and P' is the pressure well above a film of thickness d'. In addition we use conservation of atoms since we are dealing with a sealed system. This method of analysis minimizes errors due to temperature scale and pressure calibration. The Van der Waals coefficient $\Gamma(0)$ has been calculated by Sabisky and Anderson[9] to be 14.5 K (atomic layers)^3 for a helium film adsorbed on an argon substrate. As the adsorbed film thickness increases, $\Gamma(d)$ decreases slowly. These calculations have been checked against direct measurements on CaF_2 substrates by Sabisky and Anderson.

Two-fluid hydrodynamics permits us to use measured values of C to determine the density of the excitations that comprise the normal part of the fluid. The equation for velocity V of surface waves of wavelength λ on a liquid with surface

tension σ and density ρ is

$$V^2 = \left(\frac{F\lambda}{2\pi} + \frac{2\pi\sigma}{\lambda\rho}\right)\tanh\left(\frac{2\pi d}{\lambda}\right), \quad (2)$$

where F is the force at the surface of the liquid and d is the average depth of the liquid. In two-fluid hydrodynamics[10] and for $\lambda \gg d$ this result is modified to

$$C^2 = [3k\Gamma(d)/md^4]\langle\rho_s/\rho\rangle d_l . \quad (3)$$

The term in square brackets is the Van der Waals force, with k Boltzmann's constant and m the mass of a ^{4}He atom, $\langle\rho_s/\rho\rangle$ is the superfluid fraction, and d_l is the thickness of the superfluid part of the film. Reference 6 chooses to include the solid layer, d_s, and the healing length, a, in the film thickness over which the superfluid fraction is averaged. Thus at $T=0$ they find $\langle\rho_s/\rho\rangle < 1$. In fact, there is no clear basis upon which to separate $\langle\rho_s/\rho\rangle d_l$ at low temperatures. In our case, we choose $\langle\rho_s/\rho\rangle = 1$ at $T=0$ which permits a direct comparison of $\langle\rho_s\rangle$ with the Landau quasiparticle model. This choice fixes the "superfluid film thickness" d_l at $T=0$. We have also omitted from Eq. (3) their correction for compressibility which has no significant effect on our analysis.

Using our measurements of C extrapolated to $T=0$ and Eq. (3), we obtain Fig. 1. If the inert layer of adsorbed helium (that portion not superfluid) is independent of film thickness, then the points for various film thicknesses will fall on a straight line at 45°. The thick-film deviations can be explained by a 4% error in film thickness, Γ being 16% too small, or the thickness of the inert layer increasing for thicker films. In fact, we have some evidence in our pressure measurements that there is a frozen layer of helium at the substrate which increases in thickness for thicker films. At 3.4 ± 0.05 K an abrupt change in the temperature dependence of the pressure within the resonator may indicate the completion of a solid layer. Note the intercept D_1 of 1.30 $\pm$ 0.05 atomic layers. Since the points fall close to the 45° line in Fig. 1, we can establish d_l at $T=0$ as the total film thickness less this intercept. This intercept compares well with that obtained by Scholtz, McLean, and Rudnick[6] of 1.47 atomic layers. In their case, with a more intense Van der Waals attraction, a thicker inert layer should be expected. In this paper, as in Ref. 6, there are no adjustable constants.

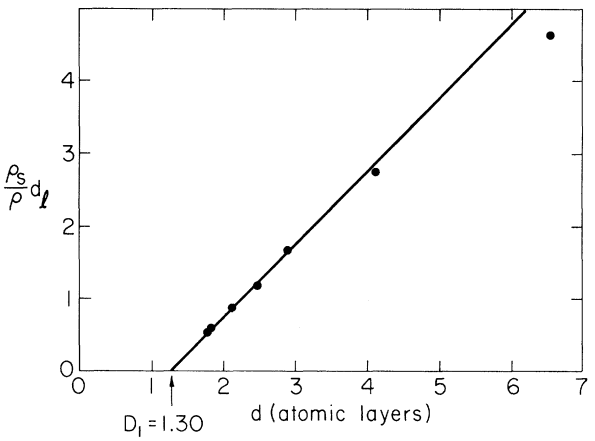

FIG. 1. $\langle\rho_s/\rho\rangle d_l$ is calculated from the third-sound velocity C (extrapolated to $T=0$) from resonance measurements. The helium film thickness d at $T=0$ is established from pressure measurements and the Van der Waals constant of 14.5 K (atomic layers)^3 given by Sabisky and Anderson (Ref. 6). The solid line is drawn at 45°. Because the experimental points lie close to this line, the concept of an inert film thickness (the intercept D_1) and a superfluid film thickness (the remainder) can be used.

Because of the great resolution of the resonance technique, we are able to examine the detailed temperature dependence of third-sound velocities. We can calculate how the density of excitations ρ_n depends on temperature, using Eq. (3):

$$\rho_n = \rho[1 - C^2(T)/C^2(0)], \quad (4)$$

where $C(0)$ is the extrapolation of our measured third-sound velocity to $T=0$ and $\rho_n = \rho - \rho_s$. The result is close to a T^3 dependence of $1 - C^2(T)/C^2(0)$ for each film thickness studied as shown in Fig. 2.

We can summarize this result by writing

$$1 - C^2(T)/C^2(0) = \rho_n/\rho = \alpha T^3. \quad (5)$$

Note that α depends on film thickness. In Fig. 3 we have plotted α^{-1} versus the total film thickness. The result is a straight line with an intercept $D_2 = 1.35$ atomic layers. If we again associate the total film thickness less this intercept with the thickness of the mobile part of the film, we see that

$$\alpha \propto 1/d_l , \quad (6)$$

and we can rewrite (5) as

$$\sigma_n \equiv \rho_n d_l \propto T^3. \quad (7)$$

Thus the areal density of the excitations, σ_n, is independent of film thickness. The inverse of the slope of the straight line in Fig. 3 gives an areal

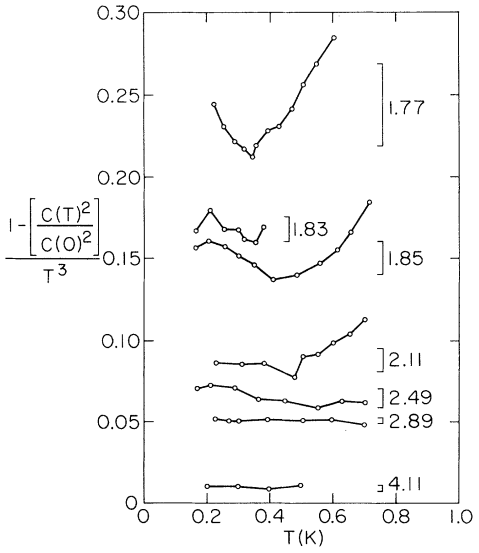

FIG. 2. The quantity $1-C^2(T)/C^2(0)$ corresponds to the thermally excited normal fraction and is calculated from resonance measurements with errors of less than 10^{-4}. The T^3 dependence indicates two-dimensional, phononlike excitations. We can express this dependence as $1-C^2(T)/C^2(0)=\alpha T^3$ for each film thickness. The brackets indicate the range of α used in Fig. 3. Next to the brackets are the corresponding helium film thicknesses in atomic layers. The thinnest film has the maximum deviation from a T^3 dependence of 10% in the exponent if the portion between 0.35 and 0.7 K is considered.

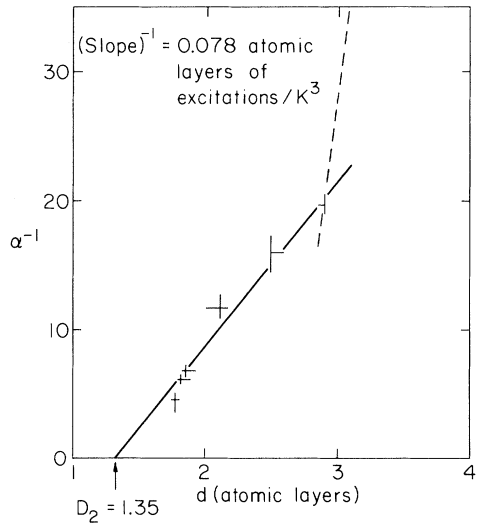

FIG. 3. The inverse of the average T^3 coefficient α as determined from Fig. 2 versus total film thickness in atomic layers. A straight line through the points indicates an areal excitation density independent of film thickness if film thickness is measured from the intercept D_2. Note that D_2 is close to D_1 (independently established in Fig. 1). The dashed line is drawn through points from thicker films not shown in the figure and indicates the end of the domain of simple two dimensionality.

excitation density of

$$\frac{1}{T^3}\frac{\sigma_n}{\rho}\left(\frac{\rho N_a}{M}\right)^{1/3}=\alpha d_1=0.078\pm0.006 \text{ (atomic layers)/K}^3, \quad (8)$$

and

$$\sigma_n=(4.05\pm0.3)\times10^{-10}\ \text{g/cm}^2\ \text{K}^3,$$

which is independent of film thickness up to about 1.5 atomic layers of superfluid (or a total film thickness of 2.8 atomic layers). N_a is Avogadro's number and M is the molar mass of ^{4}He. For thicker films (4.11 and 3.31 atomic layers in our case), although a T^3 dependence of ρ_n remains, the magnitude of α drops precipitously and the points fall well above the solid line and along the dashed line in Fig. 3. This is in qualitative agreement with Padmore's[4] comments on the effects of dimensionality on the Landau quasiparticle density. For increased dimensionality one expects decreased excitation density. A final experimental result we should mention comes from these thicker films where we see spontaneous jumps in resonant frequency of as much as 0.2% from time to time. We conjecture that we have trapped macroscopic vortices in these films which shift the resonances through a second-order Doppler effect.

The contribution to the surface normal-fluid density from thermally excited surface waves may be calculated in the same way one calculates the contribution to the bulk normal-fluid density from phonons and rotons.[11] This has been done by Kuper[12] for waves corresponding to the second term in Eq. (2) (wavelength $\lambda \ll d$, the liquid depth). The result is a $T^{5/3}$ temperature dependence of the areal excitation density. Padmore[4] has calculated σ_n for density waves with linear dispersion in two dimensions (no free surface) in the limit $\lambda \ll d$ and finds a T^3 dependence:

$$\sigma_n=[3\zeta(3)/2\pi](kT)^3/h^2C^4, \quad (9)$$

where $\zeta(3)\approx1.20$, k is Boltzmann's constant, and C is the velocity of the excitations. Although we have a free surface, we expect the same result in the long-wavelength limit as Padmore since the first term in Eq. (2) gives a linear dispersion and the second term becomes negligible in this limit. We have calculated the Landau quasipar-

ticle density in two dimensions using the complete dispersion relation for surface waves [Eq. (2)]. There are two temperature regimes for a given thickness, one corresponding to each term in the dispersion relation, and in these two limits our calculation agrees with those of Kuper and Padmore. For all the film thicknesses and temperatures realized in our experiments, this calculation produces Kuper's $T^{5/3}$ dependence as opposed to the observed T^3 dependence.

We have considered only gapless excitations in our calculation. Padmore[13] has done the Feynman-Cohen roton calculation for rotons in two-dimensional films and finds that the energy gap is reduced 2 to 3 deg below that for bulk rotons. However, we see no exponential contribution to ρ_n between 0.2 and 0.7 K for any of our films.

We are faced with two problems. One, what sort of excitation produces the observed T^3 temperature dependence in the areal excitation density? Two, why do we not see Kuper's surface-wave excitations? The T^3 behavior clearly indicates two-dimensional, phononlike excitations. In our case the natural velocity for these excitations in the long-wavelength limit is the measured third-sound velocity. However, this velocity depends strongly on film thickness [(Eq. (3)] while the excitation velocity we obtain from our measured values of α and Eq. (9) (76 ± 2 m/sec) is nearly independent of film thickness.

The films seem to behave as though their surface is rigid for short-wavelength (100 Å) thermal excitations while remaining mobile for the much longer-wavelength (1 cm) third-sound waves. If so, we must consider a two-dimensional phononlike excitation, perhaps a compressional wave between the surface and the substrate, with a velocity approximately $\frac{1}{3}$ the first-sound velocity for bulk liquid helium. This velocity is also above the maximum third-sound velocity which can be attained on argon. A partial answer to these puzzles is that the excitation we are seeing might be nonhydrodynamic.

We would like to thank Bill McMillan for his advice on calculating ripplon densities and Guenther Ahlers for pointing out to us his interest in understanding $\partial C/\partial T$ at low temperatures.

*Research supported in part by the National Science Foundation under Grants No. GH-33634 and No. GH-37892.

†Alfred P. Sloan Foundation Fellow.

[1]P. C. Hohenberg, Phys. Rev. 158, 383 (1967).
[2]J. M. Kosterlitz and D. J. Thouless, J. Phys. C: Proc. Phys. Soc., London 6, 1181 (1973).
[3]F. D. M. Pobell, H. W. Chan, L. R. Corruccini, R. P. Henkel, S. W. Schwenterly, and J. D. Reppy, Phys. Rev. Lett. 28, 542 (1972).
[4]T. C. Padmore, Phys. Rev. Lett. 28, 1512 (1972).
[5]H. Haug, J. Low Temp. Phys. 12, 479 (1973).
[6]J. H. Scholtz, E. O. McLean, and I. Rudnick, Phys. Rev. Lett. 32, 147 (1974).
[7]B. Ratnam and J. Mochel, J. Low Temp. Phys. 3, 239 (1970).
[8]B. Ratnam and J. M. Mochel, Phys. Rev. Lett. 25, 711 (1970).
[9]E. S. Sabisky and C. H. Anderson, Phys. Rev. A 7, 790 (1973).
[10]K. R. Atkins, Phys. Rev. 113, 962 (1959).
[11]K. R. Atkins, *Liquid Helium* (Cambridge Univ. Press, London, 1959), pp. 66–68.
[12]C. G. Kuper, Physica (Utrecht) 24, 1009 (1956).
[13]T. C. Padmore, Phys. Rev. Lett. 32, 826 (1974).

Frequency and Temperature Dependence of Sound Velocity in Liquid Helium-4: A Test of the Phonon Dispersion Relation*

W. R. Junker and C. Elbaum
Brown University, Providence, Rhode Island 02912
(Received 6 November 1974)

Changes $\Delta C/C$ in sound velocity C in liquid helium-4 over the frequency range 1 to 15 MHz were studied as a function of temperature for $0.45 \geq T \geq 0.1$ K. The experimental results are in qualitative agreement with predictions of positive phonon dispersion for small momentum values.

Earlier experiments by Whitney and Chase,[1] who measured the changes ΔC in the velocity C of sound in liquid He^4, indicated that at frequencies lower than 12 MHz, the velocity change $\Delta C/C$ as a function of temperature increases with increasing frequency over the temperature range