## SUPERFLUIDITY IN FINITE SYSTEMS: PROPAGATION OF FOURTH SOUND AND THE DENSITY OF SUPERFLUID\*†

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Measurements have been made of the fractional density of superfluid,  $\rho_s/\rho$ , in helium II contained in micropores (average radius ~31 Å), as calculated from fourth-sound measurements between 1.2 and 2°K. Since the lambda transition is greatly modified in this system, the temperature variation of  $\rho_s/\rho$  may generally be expected to differ from that in "infinite" bulk liquid, at least near the transition temperature. The results lead to the conclusion that the critical velocity in such channels decreases rapidly with increasing temperature above 1.2°K.

The possibility of phase transitions, and in particular of superfluidity, in systems of fewer than three dimensions (for example, onedimensional superconductors and two-dimensional helium) is of considerable current interest. An experimental approximation to such systems in the case of helium is the adsorbed film. which can vary in thickness from one atom ( $\sim 3$  Å) up to (in practice) about 100 atoms. The present measurements arise out of a systematic investigation of helium adsorbed in microporous glass (Vycor), chosen because of its high porosity which makes possible a large sample of film in a reasonably small space. With this system, measurements have been made of the absorption isotherms<sup>1</sup> (which are fundamental to a detailed interpretation of the observations), the specific heat,<sup>2</sup> and the nuclear magnetic properties<sup>3</sup>; the measurements have been reviewed recently.<sup>4</sup> For He<sup>4</sup>, a consistent model of the film is one in which the first statistical layer is of solid density, the second corresponds to liquid at about 35-atm pressure, and subsequent layers are of normal density, like bulk liquid under its saturated vapor pressure. Such a stepwise layer model can be only an approximation, but it fits the experimental results to within about 5%. It can only apply at temperatures not too close to the superfluid transition temperature, since the character of the transition is greatly modified in thin films.

Another type of experiment which yields information on this system is the propagation, in the helium-II region, of fourth sound, a density wave in the superfluid with the normal fluid locked in a stationary position by its viscosity. This type of wave, which was first proposed by Pellam<sup>5</sup> and later discussed by Atkins,<sup>6</sup> has a velocity,  $u_4$ , given with an accuracy of about 1% by the relation

$$u_4/u_1 = (\rho_s/\rho)^{1/2}, \tag{1}$$

where  $u_1$  is the velocity of first sound,  $\rho_s$  the superfluid density, and  $\rho$  the total density. Rudnick and Shapiro<sup>7,8</sup> have shown that fourth sound can be propagated in channels which are narrow enough to lock the normal fluid but wide enough for the properties to be essentially those of bulk liquid right up to the normal lambda temperature, and their later experiments gave values of  $\rho_s/\rho$  in excellent agreement with Andronikashvili's results<sup>9</sup> for bulk liquid. For helium in smaller channels, one cannot expect such a straightforward situation since the density varies considerably over an appreciable section of the channel close to the wall. However, we expect to derive an average value of  $\rho_S/\rho$ , which has not previously been measured except in bulk liquid.

In the present experiments we have used a rod of Vycor porous glass 4.68 cm long and 7.24 mm in diameter, taken from the same batch as that from which the specimens used for the other experiments were cut. It is reasonable to use as an approximate model for the pores a random cylindrical capillary system with the pore diameter sharply peaked around the value 62.4 Å, as concluded from our previous experiments. This Vycor rod was incorporated in a fourth-sound resonator very similar to that described by Shapiro and Rudnick,<sup>8</sup> with parallel-plate capacitors as trans-

mitter and receiver. The assembly was immersed in a liquid-helium bath, and the resonator filled through shallow scratches made with a razor blade on the faces of the transducer housing. Thus the fourth sound was propagated through Vycor channels of the above average diameter which must be extremely tortuous in character, so that the effective path length of the resonator cannot be assessed directly.

The first few experiments with this arrangement revealed a large number of unexplained low-frequency resonances whose frequencies bore no obvious relation to that of fourth sound or to each other. However, most of these disappeared after the apparatus had been under vacuum for some weeks, leaving only three fundamental resonances. One of these was fourth sound; the other two are not fully explained, but may be first and second sound, propagat ed through a hole about  $5\mu$  in diameter, which was subsequently found to be present on the axis of the Vycor rod. (It is not clear, however, how a second-sound mode could be detected with this arrangement.) We make the reasonable assumption that the presence of this very small hole does not materially affect the characteristics of the fourth-sound propagation. The resonant frequency was measured as a function of temperature from 1.2°K up to about 1.95°K, where the attenuation, which increases with temperature, was large enough to make the measurement inaccurate. In addition, it suffered interference with the other low-frequency resonance near this temperature.

For obvious reasons, we cannot transform the frequency measurements directly into values of  $\rho_S/\rho$  using Eq. (1). However, we can make an informed estimate of a proper normalization factor at the lowest temperature,  $1.2^{\circ}$ K, making use of the adsorbed layer structure outlined in the first paragraph. We calculate an average value of  $\rho_S/\rho$  for this system of 0.93 at that temperature (this is not very different from  $\rho_S/\rho = 0.97$  for liquid at its normal density). Hence, we derive the values of  $\rho_S/\rho$ shown in Fig. 1, together with Andronikashvili's values for bulk liquid at its saturated vapor pressure.

We first note in Fig. 1 that the temperature variation of  $\rho_s/\rho$  follows roughly that of the bulk liquid up to 1.9°K, consistent with the approximate layer model outlined above. The only theoretical prediction with which this can be compared is that of a finite noninteracting



FIG. 1. Values of  $\rho_S/\rho$  for the present Vycor specimen, normalized to the value 0.93 at 1.2°K. The solid curve gives Andronikashvili's values of  $\rho_S/\rho$  for the bulk liquid.

Bose-Einstein gas. Goble and Trainor<sup>10</sup> have computed the thermodynamic properties of such a gas in which one dimension is very much smaller than the other two (corresponding to a film rather than a channel) and they find that the fractional ground-state occupation drops off with increasing temperature more rapidly than in bulk liquid. In the present system,  $\rho_s/\rho_s$ changes rather less rapidly with temperature up to 1.9°K than in the bulk liquid, and actually becomes greater at some temperatures.<sup>11</sup> In view of the assumptions made in deriving the values in Fig. 1 one should not perhaps attach too much significance at present to their absolute magnitude, although the normalizing factor seems to be a rather plausible one.

Secondly, at temperatures above about 1.9°K,  $\rho_S/\rho$  drops very sharply below the bulk-liquid value. If we assume that this drop is due to the fact that a relevant correlation length  $\xi$  is becoming comparable with the pore diameter, then our experiment provides an estimate of the constant coefficient in the relationship between correlation length and temperature,<sup>12</sup>  $\xi = A/(T_{\lambda} - T)^{2/3}$ . With the assumption that the finite size effect becomes appreciable when  $\xi \approx \frac{1}{10}$  the pore diameter, one finds that  $A \approx 1.3$  Å °K<sup>2/3</sup>.

Third, we note that although our results do not extend to low enough frequencies to define accurately the transition temperature where  $\rho_S/\rho = 0$ , it is consistent with the temperature of onset of superflow,  $T_0 \approx 2.05^{\circ}$ K, determined<sup>13</sup> for Vycor glass with a pore radius similar to ours (35.4 Å, compared with our 31.2 Å). It has been known for some time<sup>4</sup> that  $T_0$  for a given thin film is appreciably below  $T_C$ , the temperature of the specific-heat maximum, which for the present Vycor speciments is nearly 2.1°K.

We can now use these new results to make a more specific comparison with a simple twofluid model, in which all the entropy is carried by the normal fluid  $[S_n(T) \text{ per gram}]$ , and the specific heat is given by

$$C = T \frac{d}{dT} \left( \frac{\rho_n}{\rho} S_n(T) \right). \tag{2}$$

It is well known that if the bulk-liquid results are fitted to this equation,  $S_n$  is found to be approximately independent of temperature and approximately equal to  $S_{\lambda}$ , the entropy per gram at the lambda point. In our system the behavior is quite different since a constant value of  $S_n$  in Eq. (2) implies that the specific-heat maximum would occur where  $\rho_n/\rho$  is changing fastest, i.e., at or below the temperature where  $\rho_s/\rho$  becomes zero, whereas experimentally it is found to occur at a higher temperature. Using again  $S_n = S_\lambda$  and the  $\rho_S / \rho$  values of Fig. 1 in Eq. (2), values of C can be calculated which are about 30% below the measured values at 1.2°K, but rise to a sharp peak which is a factor of about 4 too high at just over 1.9°K. Such a result can be explained if the total amount of liquid taking part in the fourth-sound phenomenon decreases as the transition temperature is approached. This could occur if a region of helium-I-type adsorbate ( $\rho_S/\rho=0$ ) grew out from the walls, leaving a progressively smaller filament of helium II adsorbate near the center of the channels, with  $\rho_s/\rho$  values given in Fig. 1. However, such a model is not in agreement with the analysis of Kiknadze, Mamaladze, and Cheishvili,<sup>14</sup> from which we would infer that a system like ours should have one sharp transition temperature from the helium-II to the helium-I state.

Finally, it is worth noting the implications of this temperature variation of  $\rho_S/\rho$  for the critical flow velocities in Vycor. The volume flow rate  $\vec{V}$  through unit cross-sectional area

of a Vycor superleak is given by

$$\dot{\mathbf{V}} = (\rho_{s} / \rho) v_{c}, \qquad (3)$$

where  $v_c$  is the critical velocity. Measurements<sup>13</sup> show that  $\dot{V}$  decreases almost linearly by a factor of about 4 between 1.2 and 1.7°K, whereas  $\rho_s/\rho$  decreases in the same temperature interval by only about 15%, so that  $v_c$  must decrease rapidly with increasing temperature. A similar conclusion would seem to hold for flow of the thin unsaturated film,<sup>4,15</sup> although no measurements of  $\rho_s/\rho$  for them exist. This is contrary to the results for the thick saturated film, in which  $v_c$  is nearly constant.

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<sup>11</sup>I. Rudnick, E. Guyon, K. A. Shapiro, and S. A. Scott, private communication, have recently observed fourth sound in a highly compressed powder system, and find

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## HYPERSONIC ATTENUATION IN SUPERCONDUCTING MERCURY AT 9 GHz †

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In the work reported here, measurements were made for the first time of the attenuation of longitudinal ultrasound in polycrystalline superconducting mercury at a frequency of about 9.3 GHz. Recent measurements of ultrasonic attenuation at lower frequencies in the strong-coupling superconductors lead and mercury showed an anomalous, or non-BCS, temperature dependence.<sup>1,2</sup> Mason pointed out that at least some of these data could be explained by assuming an interaction of the phonons with dislocations whose motion is in turn damped by the electrons.<sup>3</sup> He also showed that the ultrasonic attenuation due to electron-phonon interactions increases faster with frequency than that due to dislocations and will dominate at high frequencies. At 9.3 GHz the electronic attenuation in mercury is approximately 2000 dB/cm, which should be several orders of magnitude larger than the attenuation due to dislocation effects,<sup>3</sup> and indeed we observe a normal BCS temperature dependence.

The frequency used here is much smaller than the energy gap in mercury over essentially the entire superconducting region, and no "pair-breaking" effects are observed.<sup>4,5</sup> However, measurements using still higher frequencies or superconductors with smaller gaps are now feasible,<sup>6</sup> and the results should give a better test of the theories of attenuation.

The longitudinal acoustic pulse is generated and detected by surface excitation of a piezoelectric X-cut crystal in a re-entrant microwave cavity. The sample construction is shown in Fig. 1. Since the acoustic wavelength is of the order of a few thousand angstroms, the end surfaces of the quartz and the metal must be flat and parallel within approximately one optical wavelength. To accomplish this, a few small pieces of carefully rolled indium foil about  $3 \times 10^{-3}$  cm thick are placed between the quartz surfaces as spacers, and a drop of mercury is placed in the middle. The two pieces of quartz are pressed together, and the parallelism of the mercury is checked by examining the fringe pattern between the plates under monochromatic light. The small end of the quartz is placed in the microwave cavity and the entire assembly slowly cooled so that the mercury solidifies. While the state of the solid mercury is not known, it is believed to be polycrystalline with grain sizes of the order of the sample thickness.

A pulse-echo technique is used to measure the change in attenuation between the normal and superconducting states. The rf pulse of interest is detected by a superheterodyne receiver followed by an i.f. amplifier and video detector and displayed on an oscilloscope. A Hewlett-Packard display scanner alternately samples the signal and the noise, and the output is fed to a lock-in detector for averaging. Since the acoustic signal through the mercurv increases below the transition temperature by as much as 25 dB in some samples, a calibrated microwave attenuator is used to keep the detected signal at some reference lev-



FIG. 1. The construction of a metal sample for hypersonic-attenuation measurements.