The parameter σn_a used to best fit theory with experiment yields $\sigma \approx 5\times 10^{-20}$ m^2, a value of the order of the elastic ion-atom cross section but at least an order of magnitude smaller than the average charge-transfer cross section.[9] This is not yet understood fully. Possibly the introduction of the ion kinetic pressure term in the afterglow theory will clarify this point.

Using this theory with experimentally determined initial electron temperatures, we were able to obtain excellent agreement with electron density measurements made on four discharge tubes of radii 0.55, 0.85, 1.13, and 1.85 cm. More complete results will be presented elsewhere. This work tends to support the validity of using fluid theory for this sort of problem. It also predicts supercooling of the electrons[10]; whether or not this is real or a phenomenon introduced by approximations in the theory will be explored.

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SUPERFLOW IN HELIUM FILMS: THIRD-SOUND MEASUREMENTS*

R. S. Kagiwada, J. C. Fraser, and I. Rudnick
Department of Physics, University of California, Los Angeles, California 90024

and

D. Bergman†
Institute for Pure and Applied Physical Sciences, University of California, San Diego, California 92037
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The results of measurements of third-sound velocity in adsorbed superfluid helium films (1.12-1.85°K) are presented. It is found that the average superfluid fraction is nonvanishing at the superflow onset thickness and has a value about $\frac{3}{8}$ its bulk value. Possible reasons for a nonvanishing value are discussed. Healing lengths based on best fitting Ginzburg-Pitaevskii calculations are determined.

A new method of generating and detecting third sound was recently reported[1] which was particularly effective in unsaturated superfluid helium films. The purpose of this Letter is to present the results of some rather extensive measurements of the velocity of third sound and to discuss their implications for an understanding of the superfluid properties of such films.

The velocity of third sound, C_3, at low frequencies, is given by[1,2]

$$C_3^{\,2} = (\langle\rho_s\rangle/\rho)3\mu(1+TS/L), \qquad (1)$$

where $\langle\rho_s\rangle/\rho$ is the average superfluid fraction in the film, T is the temperature, S is the specific entropy,[3] and L is the specific latent heat. The quantity μ is the amount by which the chemical potential of the film differs from that of the bulk liquid at the same temperature and is given by

$$\mu = \frac{\alpha}{h^3} = \frac{kT}{m}\ln\frac{P_0}{P}. \qquad (2)$$

The middle term is the van der Waals potential at the surface of a film of thickness h (α is a constant), k is Boltzmann's constant, m is the mass of a helium atom, P_0 is saturated vapor pressure, and P the pressure of the vapor in equilibrium with the film.

Figure 1 gives some of the results for temperatures in the range 1.12-1.85°K. The vertical

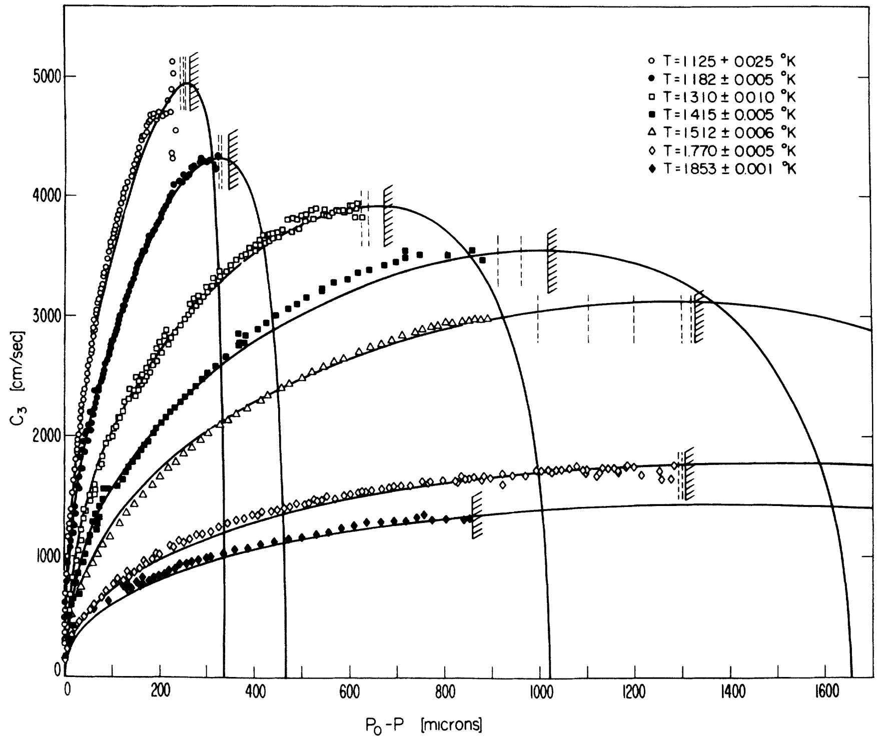

FIG. 1. The third-sound velocity versus P_0-P. The points are experimental. The hatched vertical line indicates where all signs of third sound disappear. These are taken to be the onset points for superflow and are plotted in Fig. 2. (See column 2 of Table I for film thickness at these points.) Between this hatching and the last data points, the dashed lines indicate that signals were seen, but not measured, because of their poor quality. An important feature of the data is that the velocity shows no sign of falling to zero near onset as it would if the onset were characterized by a vanishing of the superfluid fraction. The curves are calculated from the Ginzburg-Pitaevskii Ansatz assuming one solid layer of He at the substrate and the best-fitting healing lengths listed in Table I, column 4.

hatched line to the right of each set of data points indicates where we cease to see any signal even with advanced signal-averaging devices. We interpret this to be the superflow onset point and this interpretation is supported by the results shown in Fig. 2, where points so determined by third sound are compared with those obtained by other methods. Attention is directed to the fact that the film thicknesses at onset are quite small. They are shown in column 2 of Table I.

A striking feature of the results is that there is no sign of a significant decrease in C_3 just before superflow is lost as the film is thinned—in fact, in most cases, the evidence is that superflow disappears when the third-sound velocity reaches its maximum value. Thus $\langle\rho_s\rangle/\rho$ is finite and large just before the disappearance of superflow. If beyond that point $\langle\rho_s\rangle/\rho$ is zero, as in the normal state, this can mean only one of two things: (1) There is a discontinuous jump in $\langle\rho_s\rangle/\rho$; or (2) $\langle\rho_s\rangle/\rho$ is an extremely strong function of film thickness. The two amount essentially to the same thing and require that the superflow onset point is a first-order transition and has a latent heat. A theory proposing a first-order phase transition in helium thin films has been proposed by Amit.[4] Elgin and Goodstein[5] on the basis of adsorption measurements are able to place an upper limit on the latent heat far smaller than that which would be implied for 5 layers. Moreover, specific heat measurements on thin films, although not designed for our purpose, give no hint of the existence of such a latent heat.[6,7] We must thus conclude that even after the disappearance of superflow, the average superfluid fraction $\langle\rho_s\rangle/\rho$ is finite.

Landau[8] argued forcibly that the existence of a condensed boson state alone does not ensure superflow and advanced the criterion that there is a critical velocity given by $v_c = \min[\epsilon(p)/p]$, below which superflow can occur. (Here ϵ and p are the energy and momentum of the elementary excitations.) The existence of the energy gap for the rotons then ensures that $v_c > 0$. Noting that

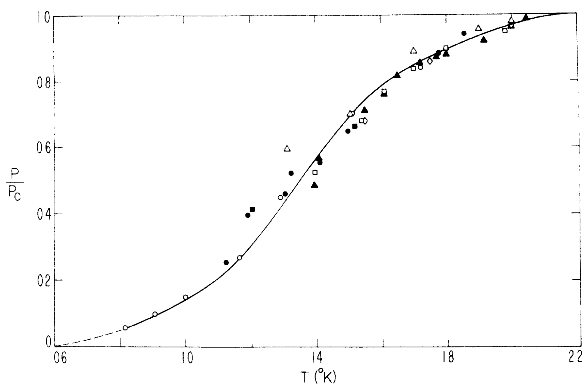

FIG. 2. The superflow onset parameter P/P_0, as determined by various experiments. Onsets of mass superflow from E. Long and L. Meyer [Phys. Rev. 85, 1030 (1952)] (solid triangles) and R. P. Henkel, G. Kukich, and J. D. Reppy [in Proceedings of the Eleventh International Conference on Low Energy Physics, St. Andrews, Scotland, August 1968 (to be published)] (open diamonds). Onsets determined by heat transport from E. Long and L. Meyer [Phys. Rev. 98, 1616 (1955)] (open triangles), D. F. Brewer and K. Mendelssohn [Proc. Roy. Soc. (London), Ser. A 260, 1 (1961)] (open squares) and K. Fokkens, W. K. Taconis, and R. De Bruyn Ouboter [Physica 32, 2129 (1966)] (open circles). Third-sound onsets (solid circles) are for data in Fig. 1; the solid squares are for data not shown in Fig. 1. The curve, taken from Fokkens, Taconis, and De Bruyn Ouboter, is a smoothed value for all heat transfer onsets and does not represent any theory.

in thin films the onset temperature for superflow is below that of the specific-heat maximum (which falls below the lambda temperature), Brewer, Symonds, and Thomson[9] with support from specific-heat data argue for the existence of gapless surface excitations which result in the critical velocity's being zero between the superflow and the specific-heat maximum temperature. Thus the ordered state is presumed to exist between these two temperatures but superflow does not. Most recently Henkel, Kukich, and Reppy,[10] using a persistent-current technique for helium films adsorbed on filter material, found that as the temperature was increased, the critical velocity approached zero more rapidly than the apparent superfluid density. Thus the determination in our work of the disappearance of superflow at a finite superfluid density is not without precedent, although the average superfluid densities at which this occurs are determined here for the first time. They are listed in the last column of Table I as fractions of the bulk values and are obtained by extrapolating the data to the onset points with the help of the curves of Fig. 1. The value of 0.44 at 1.853°K is higher than any of the others. When the onset parameter, P/P_0, is taken to be that falling on the curve of Fig. 2, then the onset point falls nearer the maximum of the third-sound curve and the value 0.37 (designated by an asterisk in Table I) is obtained. It appears that the superflow onset value of $\langle\rho_S(\text{film})\rangle/\rho_S(\text{bulk})$ is relatively temperature independent and is approximately $\frac{3}{8}$.

Since only the superfluid component moves in a third-sound wave and carries no entropy, it is clear that when an element of film thins as a result of this motion the sign of the temperature change is (1) positive if the entropy per unit area decreases with decreasing thickness, and (2) negative in the reverse case. Moreover, the wave is isothermal if the unit-area entropy is a stationary function of thickness. The first alternative is the normal one and applies in all cases to thick helium films. However, there is some evidence that below 1.5°K in films at approximately onset thickness the unit-area entropy becomes stationary and (especially at lower temperatures) may even increase with decreasing thickness before again decreasing as the film approaches vanishing thickness.[11] Goodstein and Elgin,[12] on the basis of an analysis of adsorption isostere data of much higher resolution, report

Table. I. Superfluid parameters of the helium films of Fig. 1.

T (°K)	Onset thickness (atomic layers)	Healing lengths (atomic layers) Equation (4)	This experiment ($h_s = 1$)	$\langle\rho_s(\text{film})\rangle/\rho_s(\text{bulk})$ at onset
1.125	4.0	0.76	0.64	0.38
1.182	4.3	0.78	0.72	0.39
1.310	4.4	0.86	0.80	0.35
1.415	4.7	0.94	0.85	0.36
1.512	5.2	1.02	0.94	0.36
1.770	7.4	1.43	1.41	0.36
1.853	9.3	1.67	1.63	0.44 (*0.37)

evidence that for films 4.5 and 5 layers thick, the unit-area entropy becomes stationary at the corresponding superflow onset temperature (~1.5°K). This kind of condition would in itself suffice to explain our results (the existence of a thermal third-sound wave is necessary for our experimental method), but it would leave unexplained the apparent agreement of mass-flow and thermal-current determinations of the onset parameters.[13] There remains a possibility that the regime in which the unit-area entropy increases as the thickness decreases is an unstable regime for two-fluid hydrodynamics.

What is not apparent in Fig. 1 is that as the films get thinner, the measured values of C_3 deviate more and more from those calculated from Eq. (1) using bulk values of $\langle \rho_S \rangle / \rho$.[1]

In attempting to understand these experimental results we first consider the possibility that not all of the helium film is superfluid. In fact, the van der Waals attraction exerted by atoms of the substrate on the adsorbed helium atoms leads to an increasing hydrostatic pressure in the film as the surface of the substrate is approached.[14] Thus, about 2 atomic layers away from it the pressure reaches the bulk-solidification value, and one may expect to find a thin layer of solid helium right at the substrate surface. Measurements of the adsorption isotherms for helium indicate that the first 1 or 2 atomic layers are much denser than the rest, while specific-heat data indicate that the first layer corresponds to a two-dimensional solid.[7,15] The hydrostatic pressure decreases rapidly as one moves away from the substrate, and at temperatures below 1.8°K we expect the rest of the film, which is not solid, to be superfluid.

When a solid (or normal fluid) layer of thickness h_S is present, the third-sound velocity is given by the revised expression

$$C_3^2 = \frac{\langle \rho_S \rangle}{\rho} 3\mu \frac{h-h_S}{h}\left(1+\frac{TS}{L}\right). \tag{3}$$

This decreases the expected value of C_3^2 by the factor $(h-h_S)/h$ which is smallest at the onset thickness. At 1.77°K, where the onset thickness is 7.4 atomic layers, the smallest value one might reasonably expect for this factor is 0.73 (assuming the extreme value of two atomic layers for h_S). We find however that C_3^2 is only 0.31 of its bulk value at that point. In other words, we would need a solid layer that is 5.0 atoms thick in order to explain the results in this way. This exceeds the onset thickness at the four lowest temperatures (see Table I). Thus this cannot be the sole cause of reduced values of C_3^2.

An attempt to explain the decrease of C_3 by the dependence of $\langle \rho_S \rangle$ on pressure[16] also failed, mainly because (1) the variation of $\langle \rho_S \rangle$ with pressure is too weak, (2) below 1.5°K it is in the wrong direction (ρ_S increases with increasing pressure), and (3) over most of the liquid film the pressure is nearly constant.

Clearly, it is impossible to understand these experiments within the framework of the thermodynamic properties of bulk helium. The effects of the finite size of the film have to be taken into account explicitly.

In the theories of Ginzburg and Pitaevskii[17] (GP) and Gross,[18] modified by Mamaladze[19] and Josephson[20] to conform correctly to the observed temperature dependence of the superfluid density,[21] the superfluid state is assigned a complex order parameter ψ which, in nonuniform situations, must satisfy a differential equation in space. Finite size has a special effect on the order parameter, independent of its effects on other variables of the system, that is due to the boundary conditions the order parameter must satisfy. The theory predicts a lowered value for the average superfluid density in a finite sample of helium, if the order parameter is required to vanish at least at one of the boundaries. Two different sets of boundary conditions have been used: (1) $\psi = 0$ at both surfaces and (2) $\psi = 0$ at the substrate and the normal gradient of $\psi = 0$ at the free surface. It is impossible to decide which of these is correct within the framework of this theory. We have assumed the first set of boundary conditions. The healing length l, near T_λ, is given by

$$l = \left[\frac{\hbar^2 T_\lambda \rho_{S\ \mathrm{bulk}}}{2m^2 \Delta C_p (T_\lambda - T)^2}\right]^{1/2} \propto (T_\lambda - T)^{-2/3}. \tag{4}$$

ΔC_p is the jump in specific heat superposed on the logarithmic singularity at T_λ.

Using $\langle \rho_S \rangle$ values from GP to substitute into Eq. (3), and treating the healing length as a free parameter, we can get a reasonably good fit to the experimental data as seen in Fig. 1. The fit is obtained by adjusting l to give the correct maximum value of C_3 (except at 1.853°K where we judged that the maximum had not been reached). If one assumes the possibility of a solid layer of thickness h_S next to the substrate, then it can be shown that our method of fitting the data yields a value of $h_S + 2\sqrt{2}l$. Listed in column 4 of Table I are the values of l with an assumed value of $h_S = 1$ atomic layer. Also listed are the values of l obtained from Eq. (4). While there is no reason to expect this equation to be applicable further away than about 25 mdeg from T_λ, it is interesting to note the agreement both in trend and, at higher temperatures, in absolute magnitude. This agreement deteriorates if $h_S = 0$ or 2 atomic layers is assumed.[22] We emphasize that the data do not uniquely determine h_S nor for that matter do they uniquely establish the correctness of the GP Ansatz; rather, the situation is that with a value $h_S = 1$ atomic layer and the assumed boundary conditions, the Ansatz is in good agreement with our results. It is appropriate to point out here that the agreement between the curves and the data far from onset is in large measure independent of the GP Ansatz since most of the variation is directly connected with the thickness, through μ in Eq. (1), rather than the dependence of the superfluid density on thickness. However, this is not true near onset and the method of fitting the curves to the data leaves very little latitude in the choice of $h_S + 2\sqrt{2}l$.

Finally, we return to the question of the vanishing of superflow at a finite average superfluid density. The region between this point and the presumed point at which the average superfluid density vanishes is reminiscent of the phenomenon in superconducting thin films where, in a finite temperature interval, the conductivity changes from that characterizing a superconductor to that characterizing the normal state.[23,24] In this case the explanation is in terms of fluctuations, and perhaps that is what is required in the present instance.[25]

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