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SUPERFLUIDITY WITHOUT SUPERFLOW IN UNSATURATED HELIUM FILMS

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It is shown experimentally that the superfluid fraction ρ_s/ρ is continuous and finite at the point at which superflow vanishes in unsaturated helium films. It follows that there is a region of superfluidity without superflow. In addition it is shown that the behavior of the partial molar entropy may account for the disappearance of superflow without requiring that ρ_s/ρ vanish.

The onset of superfluidity in unsaturated helium films has long been the subject of study and controversy. At each temperature below the bulk lambda point, superfluid properties are observed to vanish at some pressure below the bulk vapor pressure. The transition curve from various measurements is shown in Fig. 1. The question of the nature of the transition has been thrown into new perspective by recent measurements of the velocity of third sound[1] and of the critical velocity of persistent currents[2] in unsaturated films. In these studies values of the superfluid fraction ρ_s/ρ are deduced and in both measurements found to be finite at the onset.

If one supposes the onset curve in Fig. 1 to separate the normal and superfluid phases in the film, a finite value of ρ_s/ρ at the superfluid side implies a first-order phase transition. A first-order phase transition has also been predicted by Amit[3] on independent theoretical grounds. Assuming a first-order phase transition it can be shown[4] that

$$\left[\frac{\partial T\ln(P/P_0)}{\partial T}\right]_{\mathrm{eq}}+\frac{S_{\mathrm{I}}-S_0}{R}=\frac{(S_{\mathrm{I}}-S_{\mathrm{II}})}{R}\frac{N_{\mathrm{II}}}{(N_{\mathrm{II}}-N_{\mathrm{I}})},\qquad(1)$$

where T is temperature, P pressure, S molar entropy, R the gas constant, N the amount adsorbed per unit area, and the subscripts I, II, and 0 refer to normal film, superfluid film, and

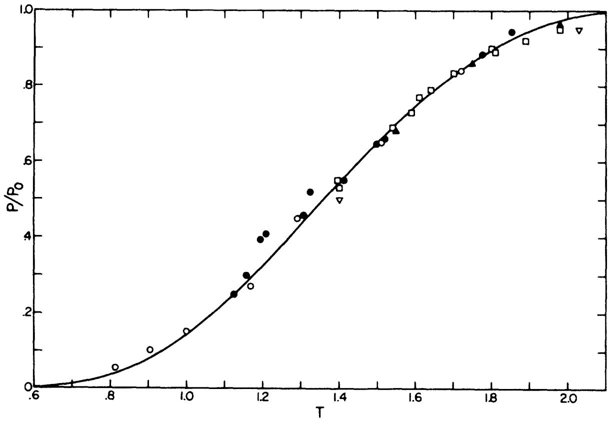

FIG. 1. The onset of superflow in unsaturated films. Inverted open triangles: mass transport, E. Long and L. Meyer, Phys. Rev. 79, 1031 (1950). Open squares: heat flow, D. F. Brewer and K. Mendelssohn, Proc. Roy. Soc. (London), Ser. A 260, 1 (1961). Open circles: heat flow, K. Fokkens, K. W. Taconis, and R. DeBruyn Ouboter, Physica, 32, 2129 (1966). Closed circles: third sound, Rudnick et al. (Ref. 1). Closed triangles: persistent currents, Henkel, Kukichi, and Reppy (Ref. 2).

bulk liquid, respectively. The temperature derivative is to be taken along the phase equilibrium curve. This equation requires that a measurement of $P(N)$ at constant T, called an adsorption isotherm, will show a discontinuity on crossing the transition curve.

If one assumes that, as in the bulk superfluid, the entropy is proportional to $(\rho-\rho_s)/\rho$, and takes ρ_s/ρ at the transition from the published third-sound data at 1.5°K,[1] one expects at this temperature a discontinuity of about 6% in N. An unrealistically extreme assumption that the film entropy is no higher than the bulk entropy[5] gives a jump of about 1%. Amit's theory predicts a jump of about 5% at this temperature.

We have measured an adsorption isotherm at 1.48° on an argon-coated copper sponge.[6] The result is shown in Fig. 2. N is continuous with an uncertainty of about 0.1%.[7] We believe this rules out a first-order phase transition. The meaning of ρ_s/ρ taken from the third-sound data is the fraction of the film that can flow parallel to the surface and which does not transport entropy.[8] That fraction is evidently still finite when the onset curve has been crossed.

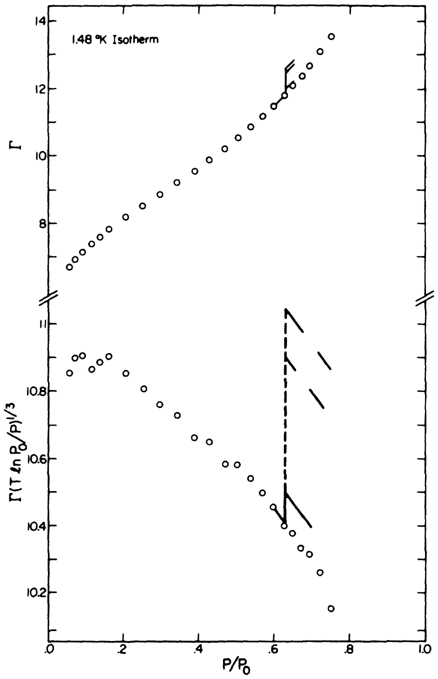

FIG. 2. Adsorption isotherm at 1.48°K. The coverage in arbitrary units, Γ, is directly proportional to N. The lower plot allows a much expanded scale. On both plots are shown hypothetical discontinuities in N of 6%, 5%, and 1%.

Bergmann has pointed out[9] that the dependence of S on N in the film may lead to a situation in which sound cannot be propagated. Since entropy transport takes place only by evaporation and recondensation, one has for the time rate of change of entropy per square centimeter in the film

$$\partial(NS)/\partial t = (T-T_a)\alpha, \qquad (2)$$

where the subscript a means average value for the film and $\alpha = \alpha(T_a, P_a, N_a)$. Putting in $S = S(T, N)$ one obtains

$$\bar{S}\frac{\partial N}{\partial t} + N\frac{C_N}{T}\frac{\partial T}{\partial t} = (T-T_a)\alpha, \qquad (3)$$

where $\bar{S}$ is the partial molar entropy and C_N the molar heat capacity at constant N. It can be shown[10] that

$$\bar{S} = \left(\frac{\partial NS}{\partial N}\right)_T = \frac{\partial}{\partial T}\Big[RT\ln(P_0/P) + \int_0^T S_0(\theta)d\theta\Big]_N, \qquad (4)$$

According to Eq. (3), when $\bar{S}$ becomes very small or equal to zero, N waves are no longer coupled to T waves, and a fluctuation in T would rapidly damp out, so that third sound cannot propagate. Physically, when $\bar{S} = 0$ the total entropy per unit area of film becomes independent of N (or film thickness) so that a local heat input cannot be absorbed at constant T by a change in N.

We have measured two adsorption isosteres [$P(T)$ at constant N] at values of N close to that for which the onset is observed at 1.5°K. The results are plotted in Fig. 3 as $[RT\ln(P_0/P)$

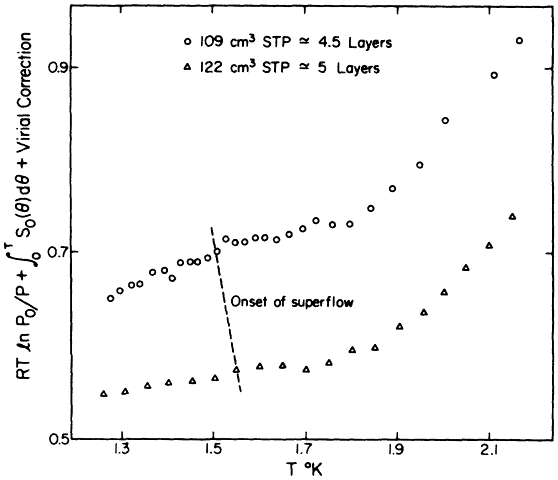

FIG. 3. Adsorption isosteres plotted so that the slope is equal to the partial molar entropy $\bar{S}$. A small virial correction is added to compensate for errors introduced by use of the ideal-gas approximation to derive Eq. (4) in the text.

$+\int_0^T S_0(\theta)d\theta]$ vs T, so that the slope of the curve is $\bar{S}$. We observe that $\bar{S}$ falls to a minimum which is equal to zero within experimental uncertainty at the onset in both of these curves. This behavior of $\bar{S}$ accounts for the disappearance of third sound and other thermally excited superflow at the transition, without requiring that ρ_S/ρ vanish.[11]

We believe these results throw new light on the nature of the film. For the first time the onset curve may be related directly to an independent measurement of a thermodynamic property of the film. On the other hand, there is apparently another transition curve to the right of the onset curve in Fig. 1 where ρ_S really goes to zero. The nature of the film between these curves, with finite ρ_S but without macroscopic flow, presents an interesting problem for the future.[12]

A more detailed report on the adsorption properties of the film is in preparation.

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[5] It is known that $S > S_0$ in these conditions. See E. Long and L. Meyer, Advan. Phys. 2, 1 (1953).

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[7] These results have been presented: R. L. Elgin and D. L. Goodstein, Bull. Am. Phys. Soc. 13, 12, 1669 (1968).

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

[9] David Bergmann, private communication. Atkins (Ref. 8) did not take into account the dependence of S on N.

[10] Hill, Ref. 4; Long and Meyer, Ref. 5.

[11] The connection of the present result to the disappearance of persistent currents remains obscure. A more complete discussion will be given by D. L. Goldstein, to be published.

[12] For an example of a model with some of these properties see D. F. Brewer, A. J. Symonds, and A. L Thomson, Phys. Rev. Letters 15, 182 (1965).

DENSITY-DEPENDENT POTENTIALS AND THE HARD-SPHERE MODEL FOR LIQUID METALS

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It has been found that various properties of liquid metals are interpretable in terms of a hard-sphere model. Since electronic screening of pair interactions in metals leads to density-dependent hard-sphere diameters, we have examined the effect that this has on the thermodynamic equations. The virial equation is shown to contain extra contributions to the pressure and compressiblity which are quite relevant for metallic systems. The behavior of alkali metals is discussed.

In many treatments of the condensed state the simplifying assumption often is made that the total N-particle interaction can be considered to be the sum of independent pair-wise contributions. At least for one class of systems, the rare gases, this approximation is known to be rather accurate.[1] For other cases important many-body effects arise and this assumption becomes questionable. However it is sometimes possible to include the most important part of the contribution of the many-body forces in a self-consistent treatment in which pair interactions alone are formally retained, but in which these become implicit functions of the thermodynamic variables.

In metals this reduction is also complicated by the two-component nature of the system of ions and electrons. Nevertheless pair potentials reflecting the screening of the ionic charges by the electron gas have been suggested and used successfully in a variety of problems.[2,3] These screened potentials are always implicit functions of the electron density, and by neutrality, of the ion number density. The temperature dependence of potentials which have been used is negligible.

The purpose of the present paper is to discuss the contributions to pressure and compressibility in liquid metals arising from the explicit density dependence of these effective pair interactions in the liquid state. We point out that the hard-sphere model of liquids can be encompassed within this