Superfluidity of Thin ^{4}He Films*

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(Received 4 March 1974)

Superfluid persistent currents have been studied in ^{4}He films adsorbed on a porous Vycor glass substrate at temperatures down to 150 mK. Stable superflow was observed in films as thin as 2.4 atomic layers at the lowest temperatures. Critical-velocity and superfluid-mass data were obtained as a function of film thickness. These data indicate that on a Vycor substrate superflow is only possible for films greater than 2 atomic layers in thickness and, further, that only the coverage above the first two layers contributes to the superflow.

In 1956, Penrose and Onsager[1] showed that Bose-Einstein condensation can occur in an infinite system of interacting Bose particles, a finding which strengthened the notion that superfluidity in liquid helium is associated with Bose-Einstein condensation. Subsequently, however, Hohenberg[2] and Chester, Fisher, and Mermin[3] proved that Bose-Einstein condensation in the sense of Penrose and Onsager does not exist in a two-dimensional helium system or, indeed, in any helium film of finite thickness. Nevertheless, superfluidity is observed in the laboratory.[4]

Kosterlitz and Thouless[5] and more recently Penrose[6] have reexamined this problem in the context of thin helium films. They conclude that metastable superflow should be possible provided the temperature is sufficiently low. At higher temperatures vortices will occur even under conditions of thermal equilibrium and any persistent superflow will decay. The temperature interval for stable superflow is expected to be a linear function of the number of particles per unit area participating in the superflow.

The aim of the work reported here has been to examine the stability of superflow in helium films at the lowest practical temperatures and coverages. The existence of persistent currents is taken as a criterion of superfluidity in these films.

^{4}He films of various coverages adsorbed on a substrate of porous Vycor glass are examined for superfluidity in a range from slightly below 0.2 K to above 1 K. Critical-velocity and superfluid-mass data are obtained as a function of temperature and coverage for those films which exhibit superflow. For the flow velocities in these experiments, the temperature at which the superflow becomes unstable is a linear function of film coverage with a zero-temperature intercept at slightly more than two layers coverage. As the coverage is increased, the superfluid mass involved in the flow increases in proportion to the coverage above the first two layers. These results indicate that the coverage up through the first two layers is bound to the Vycor and does not participate in the superflow, but rather acts as a favorable substrate. Interestingly, persistent currents are observed for fractional coverages of the third layer. This suggests the possibility of superfluidity in a dilute two-dimensional system.

The persistent currents are observed by direct measurement of angular momentum using a gyroscopic apparatus cooled by a ^{3}He-^{4}He dilution refrigerator.[7] The currents are formed in a ring of porous Vycor.[8] Vycor is particularly well suited for film experiments because it provides the large continuous surface necessary to obtain a measurable angular momentum for very thin films. The ring used in this experiment was only 1.6 cm long with an inner diameter of 4.2 cm and an outer diameter of 4.4 cm, yet its surface area estimated by nitrogen adsorption was 982 m^2.[9] Another important feature of porous Vycor, in contrast to packed powders which might provide similar surface areas, is the existence of a minimum pore of approximately 15 Å.[10] In the present experiment we restrict ourselves to films considerably thinner than this minimum.

The experimental procedure consists of introducing a measured quantity of helium gas into the experimental chamber containing the Vycor ring. At temperatures below 1 K, most of the gas is adsorbed onto the surface of the Vycor pores. An attempt is made to form a persistent current in the adsorbed film by rotating at a standard speed. The speed chosen gives a peripheral velocity of about 20 cm sec^{-1} while cool-

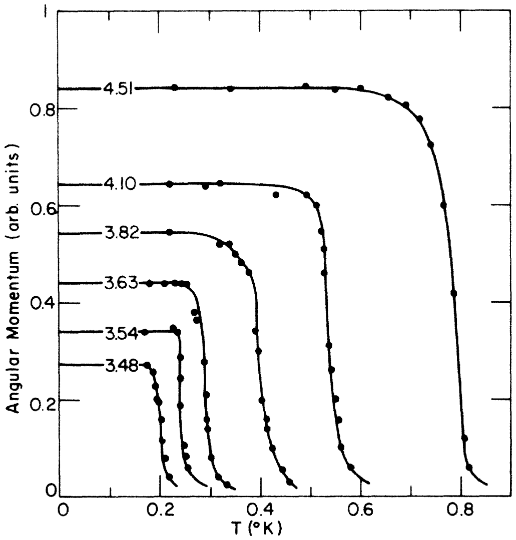

FIG. 1. The temperature dependence of the persistent current angular momentum is shown for films of various coverages. Coverage is indicated in units of 10^{-5} mole m^{-2}.

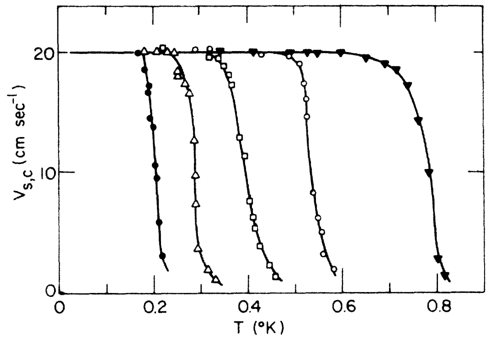

FIG. 2. The superfluid flow velocity is shown for the different film coverages. The data for 3.54×10^{-5} mole m^{-2} is omitted for clarity.

ing. The temperature is monitored by a carbon thermometer mounted directly on the Vycor ring. Vibration associated with the rotation prevents cooling much below 0.2 K. When the lowest temperature is reached, the rotation is slowly stopped and the angular momentum associated with the persistent current is measured. In the temperature range of this experiment, the smallest coverage for which a stable persistent current is observed is 35.4 μmole m^{-2}. Once a persistent current is detected, the temperature is slowly increased until the current becomes unstable and disappears. The procedure is then repeated with increased helium coverage, taking care to form each persistent current at the same rotational speed.

Figure 1 shows the angular momentum as a function of temperature for a number of persistent currents in films of increasing thickness. The angular momentum curves have two distinct regions. At low temperatures the angular momentum is a reversible function of temperature and traces out the weak temperature dependence of the superfluid mass of the persistent current. At higher temperatures the angular momentum of the persistent current decreases rapidly and irreversibly as the temperature is raised. The boundary between the reversible and irreversible regions gives a measure of the temperature dependence of the critical velocity.

The persistent currents at each film thickness were formed with the same initial velocity. In the reversible portion of the angular momentum curves of Fig. 1, the flow velocity remains constant at the initial velocity of about 20 cm sec^{-1}. Thus the differences between the angular momentum curves in the reversible region reflect the variation of the superfluid mass with coverage. We can obtain an average superfluid flow velocity as a function of temperature for each coverage by removing the dependence of the angular momentum on superfluid mass.

A family of superfluid velocity curves for the different coverages is shown in Fig. 2. The steep portions of the velocity curves correspond to the irreversible region of angular momentum and give the temperature dependence of the superfluid critical velocity for each coverage. It is seen that a small addition of helium changes the critical velocity throughout the entire system. Also, as the coverage is reduced, the critical velocity curves lie closer to zero temperature and are progressively steeper.

We are interested in the dependence of the critical velocity curves on coverage. By choosing a fixed critical velocity $v_{s,c}$, we can plot the temperature $T_{v_{s,c}}$ at which instability occurs as a function of coverage. In Fig. 3, the values for $T_{v_{s,c}}$, where $v_{s,c}$ is 10 cm sec^{-1}, are plotted for films of various coverages. It is clear that the values of $T_{v_{s,c}}$ are a linear function of the coverage with a zero-temperature intercept of 31 μmole m^{-2}. We have made similar plots for other values of the critical velocity. In each case a linear dependence on coverage is found. An important feature is that the zero-temperature intercept is the same, independent of the value of the critical velocity. This implies that on a bare Vycor sub-

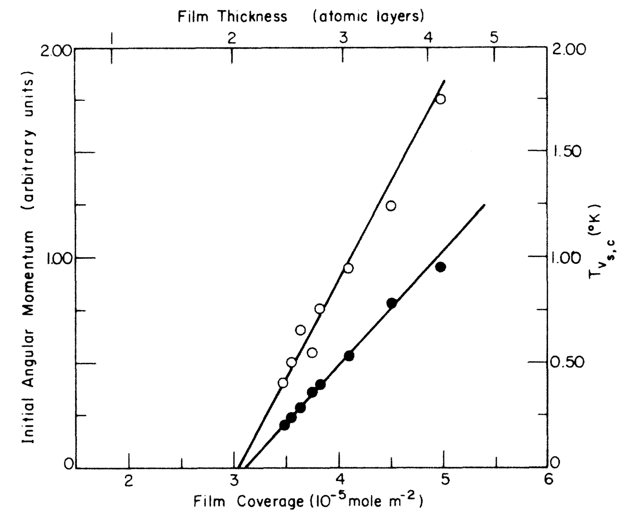

FIG. 3. The temperatures $T_{v_{s,c}}$ at which the flow-velocity curves of Fig. 2 intersect a constant critical-velocity curve, $v_{s,c}=10$ cm sec^{-1}, are plotted as closed circles against film coverage. The initial values of angular momentum for each coverage are shown as open circles.

strate a coverage of at least 31 μmole m^{-2} of He^4 is needed to sustain stable superflow at any nonzero temperature.

In Fig. 3 we also plot the initial values of angular momentum from Fig. 1 (open circles). These values are a measure of the actual mass participating in the superflow since each current is formed at the same angular velocity. The mass of flow is a linear function of coverage, with a zero intercept near that of the critical-velocity data. The difference in intercepts is probably not significant because of the size of scatter in the initial angular momentum data. It appears that only the mass above 31 μmole m^{-2} is contributing to superflow.

In interpreting these results it is useful to consider a layer model for the helium adsorbed on the surface of the Vycor glass. It is expected that the effective density of the first few layers will be enhanced by strong substrate attraction. Symond and co-workers[11] find that the first and second layers are complete at total coverages of 18.5 and 31.0 μmole m^{-2}, respectively. Recent heat-capacity measurements[12] of helium films adsorbed on Vycor show well-defined signatures, which may be identified with the completion of the first and second layers. The heat-capacity results indicate that at zero temperature a coverage of 29 μmole m^{-2} is sufficient to complete the second layer. This value is in reasonable agreement with that found by Symond and co-workers[11] and is close to the zero-temperature extrapolations of the $T_{v_{s,c}}$ and superfluid-mass data shown in Fig. 3.

The data lead us to conclude that the first two layers of helium adsorbed on Vycor are strongly bound to the substrate and do not contribute to the superflow at any temperature. However, only a fraction of the third layer is required before stable superflow is possible. In this experiment a persistent current was observed when the third layer was only 40% complete. We believe that stable superflow at even smaller coverages in the third layer would be possible if the gyroscope could be cooled to a lower temperature.

Our results are consistent with those found in the recent film experiments of Chester and Yang[4] and Scholtz, McLean, and Rudnick.[4] These investigators find also that above a certain minimum coverage the superfluid mass increases linearly with coverage. The values obtained for the minimum coverage range from 1.5 to 2 atomic layers; since the experiments involve different techniques and substrate materials[13] a more precise quantitative comparison is not possible.

The authors gratefully acknowledge the contributions of Dr. F. D. M. Pobell to the success of this experiment.

*Work supported in part by the National Science Foundation through Grant No. GH-37658 and also under the Cornell Materials Science Center, Grant No. GH-33637.

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Spin Flop, Supersolids, and Bicritical and Tetracritical Points

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A scaling theory is introduced for *bicritical* points, such as antiferromagnetic spin-flop points (with analogies to the upper λ point in ^{4}He), where two distinct critical lines meet. Experimentally testable predictions follow from renormalization-group calculations which indicate that the bicritical exponents should be Heisenberg like for systems with $n \leq 3$ components; the crossover exponent φ ($\simeq 1.25$ for $n=3$) is directly observable. For $n>3$ an *intermediate* ("supersolid") low-temperature phase may appear, the bicritical point then becoming *tetracritical*.

The Hamiltonian of a uniaxially anisotropic antiferromagnet of n-component spins $\vec{S}(\vec{R}) = \{S_1(\vec{R}) \equiv S_\parallel(\vec{R}); \vec{S}_\perp(\vec{R})\}$ at the sites $\vec{R}$ of a d-dimensional lattice may be written

$$\mathcal{H}_{\text{int}} = -\sum_{\vec{R},\vec{R}'} [J(\vec{R}-\vec{R}')\vec{S}(\vec{R})\cdot\vec{S}(\vec{R}') + D(\vec{R}-\vec{R}')S_\parallel(\vec{R})S_\parallel(\vec{R}')] - \sum_{\vec{R}} [H_\parallel S_\parallel(\vec{R}) + \vec{H}_\perp\cdot\vec{S}_\perp(\vec{R})] - \sum_{\vec{R}} \exp(i\vec{k}_0\cdot\vec{R})\vec{H}^\dagger\cdot\vec{S}(\vec{R}), \qquad (1)$$

where $J(\vec{R})$ is the isotropic exchange coupling which leads to antiferromagnetic ordering on two interpenetrating but equivalent sublattices A and B (with superlattice reciprocal vector $\vec{k}_0$), while $D(\vec{R})$ (which might well vanish for $\vec{R}\neq 0$) represents the anisotropy energy aligning the spins along the "easy" or "parallel" axis. The staggered, ordering field $\vec{H}^\dagger = (H_\parallel^\dagger, \vec{H}_\perp^\dagger)$ is physically inaccessible, so that $\vec{H}^\dagger \equiv 0$, although its response may be studied by neutron diffraction. (In a real *bi*axial Heisenberg system, with one perpendicular "hard" axis, we may,[1] in the critical region, neglect the corresponding spin components and so take $n=2$ in place of $n=3$.)

In zero uniform external field ($\vec{H}=0$) the spins align parallel and antiparallel to the easy axis, and the critical point, at T_{c0}, should have Ising-like ($n=1$) character with corresponding Ising exponents visible for T very close to T_{c0}.[1,2] If the uniform field $H_\parallel$ is now imposed (with $\vec{H}_\perp \equiv 0$, which in practice demands careful crystal alignment), one expects, as first pointed out by Néel,[3] that for $H_\parallel = H_\sigma(T) \sim DS$ the spin system should "flop" over, via a first-order transition, into an alignment that is predominantly perpendicular to the easy axis (see Fig. 1). The corresponding phase boundary in the $(H_\parallel, T)$ plane ends at what