

Some interesting possibilities concerning future experiments present themselves. If x_2 is large enough and the temperature low enough, the Fermi fluid of the second layer may approach degeneracy which will limit the scattering from \vec{k} to \vec{k}' and may give a characteristic temperature dependence to the process. However, for large x_2 , effects of order higher in x_2 than the linear ones considered here may also become important. T_2 may also show the effects of second-layer solidification as the third layer begins to form. These effects will be studied theoretically in future work.

This research was supported in part by the National Science Foundation under Grant No. DMR-7614447.

¹M. Bretz, J. G. Dash, D. C. Hickernell, E. O. McLean, and O. E. Vilches, *Phys. Rev. A* **8**, 1589 (1973).

²B. P. Cowan, M. G. Richards, A. L. Thomson, and W. J. Mullin, *Phys. Rev. Lett.* **38**, 165 (1977).

³J. R. Owers-Bradley, B. P. Cowan, and M. G. Richards, in *Proceedings of the Workshop on Physical Adsorption*, Brookhaven National Laboratory, Upton, New York, December 1976 (unpublished).

⁴D. Thouless, *Proc. Phys. Soc. (London)* **86**, 893 (1965).

⁵L. H. Nosanow and C. M. Varma, *Phys. Rev.* **187**, 660 (1969), and *Phys. Rev. A* **1**, 133 (1970).

⁶R. A. Guyer, *Phys. Rev. A* **10**, 1785 (1974).

⁷A. K. McMahan, *J. Low Temp. Phys.* **8**, 115 (1972).

⁸W. J. Mullin, *Phys. Rev. B* **12**, 3718 (1975).

⁹This form is arrived at (Refs. 4–8) by considering matrix elements of the Hamiltonian in an antisymmetrized basis. By expansion in permutations, an effective Hamiltonian is then derived which, when acting on the space of unsymmetrized space-spin “home-base states” (Ref. 7), has the same eigenvalues. Equation (1) is the lowest-order term in the series of multiparticle permutations.

¹⁰What is needed here is the second time derivative of the spin-torque correlation function. I have been able to show that the derivative is given by Eq. (4) and is equal to the fourth moment of the resonance line. Equation (4) is an extension of the results given by R. C. Richardson, A. Landesman, E. Hunt, and H. Meyer [*Phys. Rev.* **146**, 244 (1966)], and by A. Landesman [*Ann. Phys. (Paris)* **8**, 53 (1973)].

¹¹W. J. Mullin, D. J. Creswell, and B. P. Cowan, *J. Low Temp. Phys.* **25**, 247 (1976).

¹²Cf. Ref. 7, Appendix A.

¹³Exchange-hopping operators have been used in the theory of itinerant-electron magnetism by T. W. Ruijgrok, *Physica (Utrecht)* **28**, 877 (1962). Also, see C. Herring, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1966), Vol. IV, p. 249.

¹⁴Noncorrelated wave functions for the z motion of particles on the first and second layers have been obtained by A. D. Novaco, *Phys. Rev. B* **13**, 3194 (1976). Because they cannot overlap, they are useless for computation of exchange integrals.

¹⁵Novaco, Ref. 14.

Superfluid Transition of ^4He Films Adsorbed on Porous Vycor Glass

J. E. Berthold, D. J. Bishop, and J. D. Reppy

Laboratory of Atomic and Solid State Physics, and The Materials Science Center, Cornell University, Ithaca, New York 14853

(Received 18 April 1977)

The temperature variation of the superfluid density has been measured for ^4He films adsorbed on porous Vycor glass. Films with transition temperatures ranging from 0.1 to 1.5 K were studied. Apart from a small rounding on the order of $5 \times 10^{-3} T_c$, the superfluid density of these films follows a power law near the transition similar to bulk helium.

Experimental studies of the superfluid transition in thin films of adsorbed ^4He atoms have promised to provide an excellent opportunity to observe the influence of size and dimensionality on a phase transition. The ease with which the film thickness can be altered and the wide range of substrates of varying topology and surface condition should make the helium film an extremely versatile system. Unfortunately, until recently there have been few experiments with the necessary precision required for critical-point studies.

In fact, the nature of the superfluid phase transition in thin ^4He films has remained an open and controversial question.¹⁻³

The principal difficulty has been the lack of experimental probes of sufficient sensitivity to examine the superfluid properties in the immediate vicinity of the superfluid onset or transition temperature. Prior to the present work, the only method which provided a measure of the superfluid mass through the region of the transition was the quartz microbalance technique developed

by Chester and Yang⁴ at the University of California at Los Angeles. Other approaches utilizing heat transfer, mobility, third sound, or measurement of persistent-current angular momentum suffered from either ambiguity in the interpretation or loss of signal as the onset temperature was approached.²

We have developed a method which permits a high-resolution measurement of the superfluid mass for helium films adsorbed on a substrate of porous Vycor glass. Vycor glass is useful in this type of experiment because it provides a large (on the order of 100 m²/g) continuous surface area. Furthermore, the interior channels of the glass on which the helium films are adsorbed form a highly interconnected three-dimensional network.

We find that the character of the superfluid transition for the films studied is independent of thickness, and that, except for a small rounding feature, the superfluid mass follows a power law in the vicinity of the transition temperature similar to that found for bulk ⁴He. We believe that this similarity to bulk helium is a consequence of the three-dimensional topology of the substrate.

The classic technique of Andronikashvili⁵ for the measurement of the superfluid density in liquid helium has been adapted for use in the present experiment. In our case a cylinder of porous Vycor glass with length 1.4 cm, diameter 0.9 cm, and with a mean channel diameter of approximately 80 Å is incorporated in the bob of a torsional pendulum with a resonant frequency near 1570 Hz. The torsion rod is hollow and provides a means for admitting helium to the Vycor. In normal operation, the pendulum is the frequency-determining element of an oscillator circuit; the drive and detection are both electrostatic. With proper temperature control the frequency stability is one part in 10⁸ for many hours. The fractional filling of the Vycor pores is monitored by noting the frequency shift which occurs as helium is admitted; the total fractional shift in frequency between filled and empty pores is 3.06 × 10⁻³. The *Q* of the oscillator is in excess of 5 × 10⁵.

When the oscillator is cooled below the transition temperature, *T*_c, superfluid begins to form, and the previously constant period, *P*(*T*), of the oscillator decreases with temperature. Since the observed fractional changes in period are small, the difference in period, $\Delta P(T) = P(T_c) - P(T)$, is a measure of the superfluid mass. We have examined the temperature variation of $\Delta P(T)$ for films with a range of transition temperatures ex-

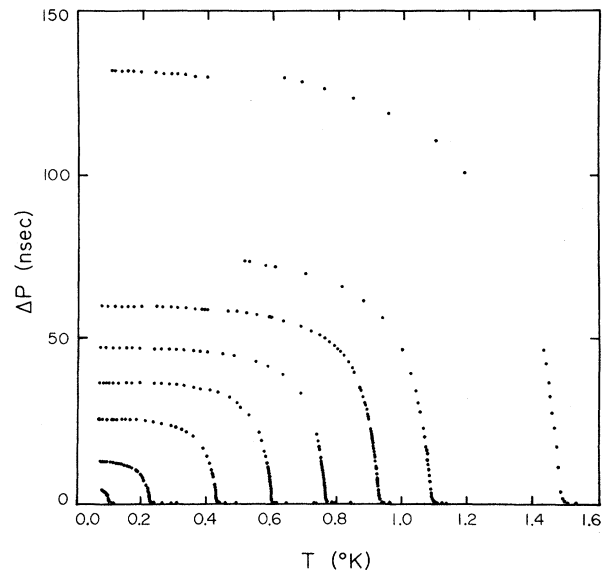


FIG. 1. The difference between the period at the transition temperature, *T*_c, and the period at a temperature *T*, $\Delta P(T)$, is plotted against *T* for a number of different film thicknesses.

tending from above 1.5 K to near 100 mK, and the results are plotted in Fig. 1.

In the interpretation of these results we attribute the entire period shift to changes in the superfluid mass. The high *Q* of the oscillator makes the amplitude of oscillation a sensitive measure of the total dissipation in the system. We have seen no degradation of the *Q* in the critical region such as would accompany the drag of even a few percent of the superfluid mass through critical velocity or other dissipative mechanisms.

In contrast to the Andronikashvili experiment performed with perfect parallel plates, the superfluid part of the film adsorbed on the Vycor must execute a complex potential flow as the pendulum oscillates. As a result, in Vycor a fraction, χ , of the superfluid moment of inertia, *I*_s, remains effectively locked to the pendulum⁶ and the observed period shift is proportional to the unlocked portion, $(1 - \chi)I_s$. This geometric flow factor can be eliminated by normalization at zero temperature, and an effective superfluid mass ratio defined by

$$m_s/m \cong \Delta P(T)/\Delta P(0), \quad (1)$$

where $\Delta P(0)$ is the zero-temperature value of $\Delta P(T)$ obtained by extrapolation for each film coverage.

In Fig. 2 we plot $\Delta P(0)$ (closed circles) as a

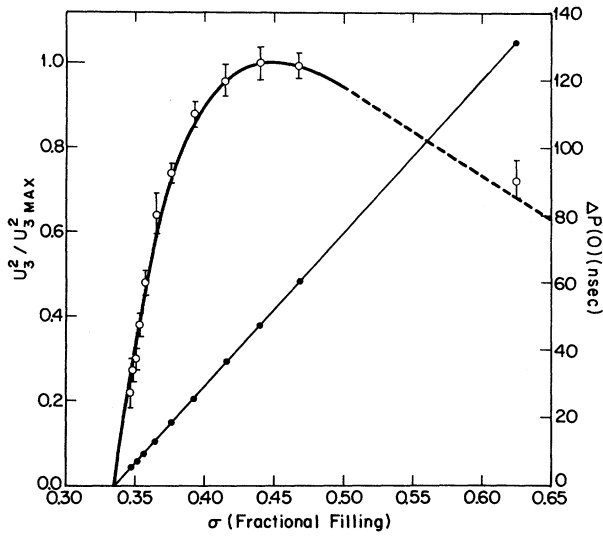


FIG. 2. The period shift observed between T_c and zero temperature, $\Delta P(0)$, is plotted (closed circles) against σ , the fractional filling of the Vycor pores. The open circles are the experimentally determined values of $[U_3(0, \sigma)/U_{3 \max}]^2$. The curve passing through the third-sound data is obtained from Eq. (4) with $\sigma_0 = 0.337$.

function of the fractional filling, σ , of the Vycor; it is a linear relationship with a nonzero intercept, σ_0 . We estimate the film thickness at σ_0 to be 1.8 atomic layers, a value which is in satisfactory agreement with the value of 2.0 layers obtained from the persistent-current measurements of Chan, Yanof, and Reppy⁷ on bare Vycor. The difference between the intercept σ_0 and the lowest coverage for which we observed superflow corresponds to a thickness of 0.06 atomic layers. The linear dependence of $\Delta P(0)$ on fractional filling implies that the connectivity of the system is independent of coverage. Thus we see no evidence for a percolation-type transition¹ in helium films, even when the amount of active superfluid is as little as 0.06 atomic layers.

By assuming that all the helium above σ_0 is superfluid at zero temperature, the slope of the line drawn through the $\Delta P(0)$ data in Fig. 2 can be used to determine the quantity χ discussed earlier. The value of $\chi = 0.77 \pm 0.01$ obtained in this way is in agreement with the value 0.76 ± 0.05 obtained from analysis of earlier third-sound measurements on Vycor.⁸

An interesting feature of these experiments has been the observation of a set of third-sound resonances which occur when the frequency of certain third-sound modes matches the resonant

frequency of the torsional oscillator. In the low-coverage regime, we expect the film thickness to be proportional to σ . In this case the temperature and coverage variation of the third-sound velocity $U_3(T, \sigma)$ is given by the expression⁹

$$U_3^2(T, \sigma) \propto (3\alpha/\sigma^4)(\rho_s/\rho)(\sigma - \sigma_0), \quad (2)$$

where α is the coefficient of the van der Waals interaction. Since the superfluid mass term, $(\rho_s/\rho)(\sigma - \sigma_0)$, in the third-sound expression is proportional to $\Delta P(T, \sigma)$ we obtain the zero-temperature value of the third-sound velocity from the relation

$$U_3^2(0, \sigma) = U_3^2(T, \sigma) [\Delta P(0, \sigma) / \Delta P(T, \sigma)]. \quad (3)$$

Since $\Delta P(0, \sigma)$ is proportional to $(\sigma - \sigma_0)$, $U_3^2(0, \sigma)$ will vary as $(\sigma - \sigma_0)\sigma^{-4}$. Thus $U_3^2(0, \sigma)$ will be zero for $\sigma = \sigma_0$, increase to a maximum value, $U_{3 \max}^2$, at $\sigma = 4\sigma_0/3$, and then decrease continuously for larger coverages. Normalizing with the maximum third-sound velocity, we then have

$$\frac{U_3^2(0, \sigma)}{U_{3 \max}^2} = \left(\frac{256}{27} \sigma_0^3 \right) \left(\frac{\sigma - \sigma_0}{\sigma^4} \right). \quad (4)$$

The quantity $[U_3(0, \sigma)/U_{3 \max}]^2$ (plotted in Fig. 2 as open circles) is calculated from the observed third-sound resonances through the use of Eq. (3) and the $\Delta P(T, \sigma)$ data. The data points represent an average value obtained from the observation of several different cavity modes at each coverage. For comparison we have also plotted the function given by Eq. (4) for $\sigma_0 = 0.337$, the value determined by the $\Delta P(0)$ vs coverage data. The dashed section of the line indicates the region where we expect the assumption of linearity between σ and film thickness to begin to fail because of the substrate curvature.

The maximum value of the third-sound velocity used for normalization in Fig. 2 may be expressed as an equivalent velocity for third sound propagating over a flat surface. In making this conversion we must identify a specific cavity mode (the longitudinal fundamental) and make use of the relation $n^{-2} = 1 - \chi$, where n is the index of refraction. We then obtain the value of 79 m/sec for the maximum third-sound velocity on a flat Vycor surface.

The most important aspect of the observation of third-sound cavity modes is the implication that superfluidity exists throughout the entire cell and not just in isolated regions, even when the active superfluid mass is as little as only 0.06 of a monolayer. In this regime we picture the superfluid as a dilute surface gas adsorbed

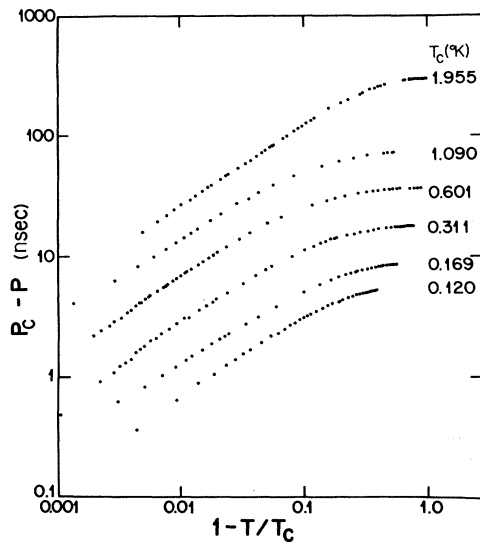


FIG. 3. The change in period $P_c - P(T) = \Delta F(T)$ is plotted against the reduced temperature, $1 - T/T_c$, for films with transition temperatures ranging from 1.090 to 0.120 K. Full-pore fourth-sound data with a transition of 1.955 K are shown for comparison (Ref. 13).

on a substrate of localized ^4He atoms.

We turn now to a discussion of our results near the superfluid transition. In this region the temperature dependence of the superfluid mass may be characterized by a power law,¹⁰

$$\Delta P(T) = A(1 - T/T_c)^\zeta.$$

Least-squares fits to the data were performed to choose the best value of T_c and the exponent ζ for each coverage. The values of ζ obtained in this way vary from 0.59 to 0.67 but are uncorrelated with the fractional filling. Therefore, we fix ζ at its average value of 0.635 ± 0.050 and choose T_c for each coverage to optimize the fit. The average value obtained for ζ is near the value of 0.674 measured in bulk ^4He .^{11,12} In Fig. 3 we show log-log plots of $\Delta P(T)$ as a function of the reduced temperature. The transition temperatures for the various coverages are shown at the right. The top set of data in this graph is from the fourth-sound measurements of Kieweit, Hall, and Reppy,¹³ properly scaled by our own measurements of $\Delta P(0)$ in the case of filled pores. The striking feature of this graph is that the nature of the transition is independent of fractional filling over the entire region studied. In fact, even when the helium film is considerably thinner than the minimum size of the Vycor pores, it still displays a three-dimensional character near the

transition.

These results for ^4He films adsorbed on porous Vycor are in marked contrast to the behavior seen by others for films on flat substrates, where the superfluid mass disappears rather abruptly at onset. We attribute this difference in behavior to the change from the three-dimensionally connected surface of Vycor to the essentially two-dimensional geometry used in the other investigations.^{4,9,14}

We would like to thank Dr. E. N. Smith for providing us with the Leiden-type ^3He - ^4He dilution refrigerator used in this work and J. H. Reppy for aid in data analysis.

This work has been supported by the National Science Foundation through Grant No. DMR75-08624 and by the National Science Foundation through the Cornell Materials Science Center Grant No. DMR76-01281, MSC Report No. 2795.

¹J. G. Dash and J. A. Herb, Phys. Rev. A **7**, 1427 (1973); M. W. Cole, J. G. Dash, and J. A. Herb, Phys. Rev. B **11**, 163 (1975); J. G. Dash, Phys. Rev. B **15**, 3136 (1977).

²D. Brewer, *The Physics of Liquid and Solid Helium*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, to be published), Pt. 2.

³A. Eggington, *The Helium Liquids*, edited by J. Armitage and I. Farquhar (Academic, New York, 1975).

⁴M. Chester and L. C. Yang, Phys. Rev. Lett. **31**, 1377 (1973).

⁵E. L. Andronikashvili, Zh. Eksp. Teor. Fiz. **16**, 780 (1946).

⁶J. B. Mehl and W. Zimmerman, Jr., Phys. Rev. **167**, 214 (1968).

⁷M. H. W. Chan, A. W. Yanof, and J. D. Reppy, Phys. Rev. Lett. **32**, 1347 (1974).

⁸D. J. Bishop, J. M. Parpia, and J. D. Reppy, in *Low Temperature Physics—LT 14*, edited by M. Krusius and M. Vuorio (American Elsevier, New York, 1975), and to be published.

⁹K. R. Atkins, Phys. Rev. **113**, 962 (1959); K. R. Atkins and I. Rudnick, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1970).

¹⁰For the purpose of the power-law fits we omitted a small region of obvious rounding at the transition. The extent of the rounded region depends only weakly on the coverage. The value of reduced temperature ($1 - T/T_c$) at which the data depart from a power-law behavior is 0.0015 for a transition at 1.0 K and increases to 0.005 for a transition at 0.1 K.

¹¹G. Ahlers, in *The Physics of Liquid and Solid Helium*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976), Pt. 1.

¹²Additional evidence for a power-law type of behavior in the ⁴He-Vycor system comes from the recent mass conductivity measurements of A. Tyler and M. Bagley, *J. Low. Temp. Phys.* **26**, 573 (1977).

¹³C. W. Kiewiet, H. E. Hall, and J. D. Reppy, *Phys. Rev. Lett.* **35**, 1286 (1975).

¹⁴B. Ratnam and J. Mochele, *J. Low Temp. Phys.* **3**, 239 (1970).

High-Resolution X-Ray Study of a Second-Order Nematic-Smectic-A Phase Transition

J. Als-Nielsen,^(a) R. J. Birgeneau, M. Kaplan, J. D. Litster, and C. R. Safinya

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 6 June 1977)

We have carried out a high-resolution x-ray-scattering study of the critical fluctuations in the nematic phase associated with the nematic-smectic-A transition in N-*p*-cyanobenzylidene-*p*-octyloxyaniline (CBOOA). It is shown that for $8 \times 10^{-3} \leq T/T_c - 1 \leq 2 \times 10^{-2}$ the transverse and longitudinal correlation lengths diverge with identical exponents, that is, $|\nu_L - \nu_T| < 0.03$. As one approaches T_c the system appears to crossover from helium- to mean-field critical behavior at a reduced temperature of $\sim 10^{-3}$.

Liquid crystals present a fascinating array of phase transitions associated with the orientational and spatial order of the rodlike liquid-crystal molecules.¹ Recently, considerable attention has been directed towards the nematic-smectic-A (*N-A*) transition in which, in the oriented liquid, a one-dimensional sinusoidal density wave is established. The general phenomenology of the behavior around the *N-A* transition seems to be well understood.¹ There are, however, a number of disturbing quantitative conflicts both within and between various theoretical models²⁻⁴ and experimental measurements⁵⁻⁸ of the critical behavior at the *N-A* transition. Most importantly, a number of experiments,⁶ especially those in N-*p*-cyanobenzylidene-*p*-octyloxyaniline (CBOOA), suggest that the *N-A* transition is characterized by *two*, rather than *one*, divergent lengths. This would seem to conflict with current ideas on scaling near second-order transitions. Further it greatly complicates any attempt at a coherent theoretical description of the *N-A* transition.

In this Letter we report a detailed high-resolution x-ray study of the *N-A* transition in CBOOA. As emphasized by McMillan,⁵ x rays couple directly to the smectic-A mass-density fluctuations so that one measures the relevant transverse and longitudinal correlation lengths, ξ_T and ξ_L , respectively, directly rather than indirectly as in most other measurements. Our salient result is that ξ_L and ξ_T diverge with identical exponents: More quantitatively, $|\nu_L - \nu_T| < 0.03$ for $8 \times 10^{-5} \leq T/T_c - 1 \leq 2 \times 10^{-2}$. This result thence represents a major simplification of the *N-A* problem. We believe that previous measurements have implied $\nu_L \neq \nu_T$ either because of limited accuracy

or because of difficulties in interpretation. We find in addition the following results which are important in understanding the physics of CBOOA itself. Firstly, there is an apparent crossover from helium- to mean-field-like behavior, at $T/T_c - 1 \approx 10^{-3}$. Secondly, critical-exponent- η effects are seen to be negligible throughout the critical region. Thirdly, in our sample the transition is second order to within $T/T_c - 1 \approx 3 \times 10^{-5}$.

The experiments were carried out on a two-crystal x-ray spectrometer with use of Cu- $K\alpha$ radiation from a Rigaku 12-kW rotating-anode source. The experimental setup is shown schematically in Fig. 1. We postpone any detailed discussion of the apparatus and the x-ray techniques to a later publication. Here it is sufficient to note that using germanium-single-crystal techniques it is possible to measure the liquid-crystal mass-density-fluctuation spectrum with a spatial resolution expressed as half-widths at half-maximum of $4.2 \times 10^{-4} \text{ \AA}^{-1}$ in the longitudinal direction (in this case, perpendicular to the smectic layers), of $< 2 \times 10^{-5} \text{ \AA}^{-1}$ in the transverse direction, and of $1.8 \times 10^{-2} \text{ \AA}^{-1}$ in the direction perpendicular to the scattering plane. The scattering diagram in reciprocal space together with the in-plane resolution ellipse and the critical fluctuation half-intensity contour at $t = T/T_c - 1 = 0.001$ are shown in the bottom half of Fig. 1. The liquid crystal was contained in a flat rectangular vessel $12 \times 12 \times 1.5 \text{ mm}^3$, with Kapton windows coated with SiO to provide planar alignment in the smectic phase. An applied magnetic field of 400 G provided alignment in the nematic phase. Only the center $1 \times 3 \text{ mm}^2$ was illuminated with x rays. The liquid-crystal holder was mounted in