

Critical behavior of superfluid ^4He films adsorbed in aerogel glass

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We present measurements of the superfluid density and heat capacity of thin films of ^4He adsorbed in aerogel glass. We find that the critical behavior of the superfluid density for films with transition temperatures between 50 mK and 1 K is similar to that seen for films adsorbed on Vycor. Power-law behavior is observed over at least one order of magnitude in reduced temperature with an exponent $\zeta = 0.63 \pm 0.03$. This exponent is much smaller than the value 0.811 ± 0.004 found when the pores were filled with helium. We also find that the exponent is unchanged by the addition of small amounts of ^3He . These results are consistent with the existence of a correlation length that diverges at the superfluid transition. Unlike for full-pore ^4He aerogel, the correlation length is long compared with all structural length scales throughout the superfluid density power-law regime, thus leading to the more bulklike critical behavior. The heat capacity of ^4He films in aerogel shows no singularity like that seen for full-pore ^4He aerogel or for thin films adsorbed in Vycor. We show that this result is consistent with the predictions of hyperuniversality given the long correlation lengths implied by the superfluid density measurements.

I. INTRODUCTION

Experiments on superfluid ^4He films adsorbed in porous media address a variety of topics in the field of phase transitions and critical phenomena. Among these are the importance of topology in determining the effective dimensionality near a phase transition,^{1,2} the role of finite-size effects³ in a two-dimensional system, and the possible realization of a dilute Bose gas.⁴ Although various porous media have been used in these studies, a large fraction of the experimental effort has been devoted to the ^4He -Vycor system.^{1,4-6} Vycor⁷ offers several advantages over other porous media. It is well characterized⁸ and provides a large specific surface area, outstanding structural integrity, and good homogeneity on macroscopic length scales. Given these benefits and its availability, the widespread use of Vycor is not surprising. In many cases, however, the interpretation of the large body of data on the ^4He -Vycor system has been handicapped by the absence of a similarly comprehensive set of data for another substrate.

In this paper, we review our measurements of the superfluid density and heat capacity of thin films of ^4He adsorbed on aerogel glass for films with transition temperatures between 50 mK and 1 K. Aerogel comprises a network of silica strands 50–100 Å in diameter without any characteristic pore size.⁹ It is therefore very different from Vycor, which has a spongelike structure with a characteristic pore size of 70–80 Å.⁸ Nevertheless, we

find that the temperature dependence of the superfluid density ρ_s near the transition temperature T_c in thin ^4He films adsorbed on aerogel is very similar to that seen for films adsorbed on Vycor. In contrast, the superfluid densities measured near T_c when the two porous media are filled with ^4He are very different.² We find that the heat capacity of thin films of ^4He on aerogel is also similar to that of thin-film ^4He -Vycor, except that the small peak at T_c seen in the latter system does not appear. The contrast with the full-pore results is particularly strong in this case, since the heat capacity of full-pore ^4He -aerogel is marked by a singularity at T_c (Refs. 10–12) which has not been observed for full-pore ^4He -Vycor.^{6,13}

This paper will discuss the results on thin films of ^4He adsorbed in aerogel in the context of full-pore aerogel as well as the Vycor measurements. We will therefore refer frequently to any one of four systems: full-pore aerogel, thin-film aerogel, full-pore Vycor, and thin-film Vycor. Our results for thin-film ^4He -aerogel are consistent with the existence of a three dimensional correlation length ξ that diverges at the superfluid transition. The data allow for at least two tests of this interpretation, which we have adopted previously for the other three systems.^{11,14,15} First, we find that the fundamental length scale ξ_0 for the correlation length is much larger for thin films in aerogel than for full-pore aerogel. This implies that the Harris criterion,¹⁶ which appears to fail for the full-pore case, is more likely to apply to thin films. The measured critical behavior of the films is indeed closer to that of pure ^4He than that of full-pore aerogel, in agree-

ment with this argument. Second, the large value of ξ_0 for thin-film ^4He -aerogel implies that the critical amplitude of the heat capacity should be at least an order of magnitude smaller than for Vycor films of a comparable transition temperature. The experimental data are also in agreement with this prediction, which follows from the hyperuniversality argument¹⁷ applied previously to the other systems.^{11,18,19} We conclude that the critical behavior of thin films of ^4He in aerogel is three dimensional (3D) in character and closer to that of bulk ^4He than that of full-pore aerogel.

The remainder of this paper is divided into six sections. First, we present a brief review of the Vycor measurements and some of the ideas proposed about that system. Second, we will review some of the relevant material from the theory of critical phenomena, with emphasis on the Harris criterion and the hyperuniversality hypothesis. We will then discuss experimental details and our results. In the subsequent discussion section, we consider our data in the context of the Harris criterion and hyperuniversality. Finally, we conclude by noting some of the limitations of the current experiments and how they might be addressed in future work.

We dispense with two other points of organization at this time. First, this paper includes some data on mixture films and an accompanying discussion about the role of annealed impurities. Second, in organizing the paper we have excluded some topics of current interest. The first of these concerns the superfluid transition of very thin films ($T_c < 50$ mK) and the accompanying issues of the dilute Bose gas^{4,20} and boson localization.²¹ These subjects will be covered in a separate paper.²² We have also chosen not to include a detailed discussion of various extensions of the Kosterlitz-Thouless theory of the 2D superfluid transition to multiply connected substrates.²³⁻²⁵ Close to T_c , these theories predict a crossover to three dimensional critical behavior, in agreement with our conclusions about the ^4He -aerogel system. The details of this crossover, however, remain a point of debate and are beyond the scope of this paper, which focuses on the critical region.

II. BACKGROUND: ^4He FILMS ADSORBED IN VYCOR

In 1977, Berthold *et al.*¹ reported on measurements of the superfluid density ρ_s of ^4He films adsorbed in Vycor. They found that ρ_s decreased to zero continuously, vanishing at the superfluid transition temperature T_c . This observation was in sharp contrast to the Kosterlitz-Thouless²⁶ transition observed on planar substrates such as Mylar, at which ρ_s jumps discontinuously to zero.²⁷ Berthold *et al.* also found that ρ_s conformed to a power law

$$\rho_s(t) = \rho_{s_0} t^\zeta \quad (1)$$

for reduced temperatures $t = 1 - T/T_c$ between 10^{-2} and 10^{-1} . They found that the exponent $\zeta = 0.64 \pm 0.05$ for films with transition temperatures between 120 mK and 1 K. This exponent agreed within error limits with

the value 0.65 ± 0.03 measured for the case of *full-pore* Vycor²⁸ as well as the value 0.674 ± 0.001 (Ref. 29) measured for bulk ^4He . The agreement among these exponents supported the claim that the superfluid transition in the ^4He -Vycor system is a second-order phase transition in the same universality class as that of bulk helium. The central physical statement of this argument is that the system is effectively three dimensional near the superfluid transition. This follows from the assumption of a diverging correlation length ξ that becomes larger than any of the structural length scales as $T \rightarrow T_c$. Reppy and co-workers^{2,15} have argued that the multiple connectivity of the substrate at long length scales thus leads to the observed 3D behavior.

The apparent irrelevance of the disorder imposed by the substrate has proven to be a more subtle issue. The structure factor of Vycor glass has a pronounced peak at a wave vector of 0.022 \AA^{-1} with a half-width of about 0.015 \AA^{-1} ,⁸ corresponding to a physical length scale of $300 \pm 100 \text{ \AA}$. The superfluid correlation length ξ inferred from the Josephson relation,^{30,31} which will be discussed below, is on the order of 250 \AA for full-pore Vycor at a reduced temperature of 10^{-1} (Ref. 2) and increases as the system moves to the thin-film limit.³² ξ is therefore greater than the *structural* correlation length over much of the observed power-law regime for full-pore Vycor and all of the power-law regime for the thinnest films. This suggests that the correlated disorder of the Vycor is irrelevant in determining the superfluid critical behavior. The irrelevance of the *uncorrelated* disorder at longer length scales follows from the Harris criterion,¹⁶ which will be discussed in more detail below.

The interpretation of the early Vycor measurements was challenged by Kotsubo and Williams,³ who argued that the superfluid transition in ^4He films adsorbed on Vycor is a finite-size Kosterlitz-Thouless transition. This argument was inspired by their own work on the finite-size Kosterlitz-Thouless (KT) transition in packed powders with grain sizes greater than 500 \AA . Kotsubo and Williams derived modified KT recursion relations for a thin film confined to a spherical surface and showed that the jump³³ in the superfluid density ρ_s that occurs on a flat substrate disappears; instead, ρ_s decreases to zero continuously, and the broadening of the transition is more pronounced for smaller spheres. They argued that the superfluid transitions observed for films adsorbed on Vycor were qualitatively consistent with a broadened KT transition and demonstrated that the transition temperature scaled with the superfluid signal according to the Nelson-Kosterlitz universal jump line.³³ These authors also noted that the argument of Reppy and co-workers for a diverging correlation length was only self-consistent, and that the superfluid density measurements alone could not prove its existence.

Several aspects of the argument of Kotsubo and Williams have been addressed in subsequent experiments. First, heat capacity experiments have established that a sharp heat capacity anomaly exists at the superfluid transition in thin ^4He films adsorbed in Vycor^{6,18,34} for transition temperatures down to 20 mK. These results confirm that the phase transition is three dimensional

in character since only an unmeasurable essential singularity in the specific heat is expected at the 2D KT transition.³⁵ The Vycor data show a heat capacity peak at the transition superimposed on a simple cusp. The cusp is due to the different excitation spectra of the superfluid and normal films, and the peak is a signature of a diverging correlation length. The size of the peak is consistent with the superfluid density data and the predictions of hyperuniversality.^{17–19,34}

Although a diverging correlation length is not expected in the original model of Kotsubo and Williams, several extensions of the finite-size KT theory have been proposed which include a crossover to 3D behavior near the superfluid transition.^{23–25} These theories share a common approach in that each incorporates vortexlike excitations which are associated with the topology of the substrate. For example, Machta and Guyer²³ use pore vortices in which the superfluid circulation surrounds either a hole in the substrate or a “trunk” of the substrate. In either case, the vortex cannot be shrunk continuously to a point. Gallet and Williams²⁵ select as their fundamental excitation a string linking many Machta-Guyer pore vortices. Both of these models lead to three dimensional critical behavior, and Machta and Guyer demonstrate explicitly that their Hamiltonian is in the same universality class as bulk ^4He .

Given the apparent relevance of substrate topology and the success of the finite-size KT theory in the case of large-diameter powders like those used by Kotsubo and Williams, Shirahama *et al.*³⁶ chose to study a series of porous glasses with pore sizes extending from 50 Å (about the diameter used in the work of Reppy and co-workers) up to 10 000 Å. Each of their samples was prepared in a manner similar to that used in the manufacture of Vycor. They found a pore-size dependence of the transition temperature in films of the same chemical potential as well as a dissipation peak at the transition for films adsorbed in the larger pore-size glasses. Shirahama *et al.* argued that these features were evidence of the unbinding of vortex pairs. The size of the dissipation peaks, for example, increased with pore size as expected from the arguments of Kotsubo and Williams. Although this work did not include detailed measurements near the transition temperatures, it suggested that the observed T_c 's were determined in large part by the physics of a finite-size Kosterlitz-Thouless transition.

In summarizing the above experimental and theoretical contributions, it is important to note that a vortex is an important excitation in the 3D XY model; therefore, the extensions of the finite-size KT theory to multiply connected systems may incorporate much of the essential physics of the ^4He -Vycor system. We emphasize that the existence of singular behavior in both ρ_s and the heat capacity C does not mean that vorticity is irrelevant. The singularities, however, establish that Vycor is distinct from two-dimensional substrates, packed powders, and even larger-diameter porous glasses such as those studied by Shirahama *et al.* We find that thin films of ^4He adsorbed on high-porosity aerogel glasses are very similar to those adsorbed on Vycor, although the respective systems are very different when the pores are filled

with helium.^{2,10,11} The next section reviews the theoretical background necessary to discuss the data.

III. CRITICAL PHENOMENA AND DISORDER

A. Superfluid transition in bulk ^4He

Excellent reviews of critical phenomena at the superfluid transition in ^4He have been given by Ahlers,³⁷ and some of the topics relevant to ^4He in porous media have been covered in recent papers by Reppy¹⁵ and Wong *et al.*¹¹ The focus here will be on the *correlation length* ξ , which is assumed to follow a power law near the superfluid transition:

$$\xi(t) \sim \xi_0 |t|^{-\nu}, \quad (2)$$

where $t = T/T_c - 1$ is the *reduced temperature*, which is the independent thermodynamic variable in all of the experiments discussed here. For ^4He , the correlation length as conventionally defined diverges for $T < T_c$ and is generally replaced by the *phase coherence length*, also denoted $\xi(t)$. The phase coherence length is related to the *superfluid density* ρ_s by the Josephson relation³⁰

$$\xi(t) = \left(\frac{k_B T m^2}{\hbar^2} \right) \frac{1}{\rho_s(t)}. \quad (3)$$

The relation stated here is for three dimensions, although it can be generalized for other dimensions and model systems using finite-size scaling arguments.³¹ We will follow conventional practice and refer to the phase coherence length as the superfluid correlation length, although we emphasize that further arguments are necessary to relate the length defined in Eq. (3) with the *physical* correlation length for $T > T_c$. The two lengths are related by a temperature-independent scale factor only in the asymptotic power-law regime.¹⁷ The Josephson relation allows one to infer ξ from measurements of ρ_s . In the case of bulk ^4He , ρ_s vanishes at the superfluid transition as

$$\rho_s \sim \rho_{s0} |t|^\zeta. \quad (4)$$

The parameters in this power law, determined from the most recent analysis of the data of Ahlers and co-workers,^{29,38} are $\rho_{s0} = 0.351 \text{ g/cm}^3$ and $\zeta = 0.6717 \pm 0.0004$. Note that by Eq. (3) the correlation length exponent ν is equal to the superfluid density exponent ζ . The deviations from Eq. (4) are less than 1% for reduced temperatures t between 10^{-2} and $10^{-4.5}$. Using Eq. (3), we find that the correlation lengths in this power-law regime vary from approximately 80 to 500 Å.

A series of high-resolution experiments³⁹ have determined that the specific heat $C(t)$ of ^4He at the saturated vapor pressure follows power laws

$$C(t) \sim \begin{cases} \frac{A}{\alpha} |t|^{-\alpha} + B, & t > 0, \\ \frac{A'}{\alpha'} |t|^{-\alpha'} + B', & t < 0, \end{cases} \quad (5)$$

for $10^{-7} < |t| < 10^{-3}$, with $\alpha = \alpha' = -0.013 \pm 0.003$. The exponent α is related to the correlation length exponent

ν by the hyperscaling relation³⁰

$$\nu = \frac{2 - \alpha}{d}. \quad (6)$$

This relation is satisfied by the above values of α and ν .

B. Introduction of disorder

We now consider the behavior of the various critical exponents upon the introduction of disorder into the system. In the case of ${}^4\text{He}$, this can be achieved by allowing it to fill the pores of a porous medium. The disorder in this case is *quenched* in that the positions of the impurities are fixed in time. The structure of the porous medium can be characterized by a density-density correlation function

$$g(\vec{r}) = \langle n(\vec{r})n(0) \rangle - \langle n \rangle^2, \quad (7)$$

where $n(\vec{r})$ is the impurity density at \vec{r} and the averages are over the volume of the porous medium. For an isotropic system such as Vycor or aerogel, $g(\vec{r}) = g(r)$. The effect of quenched disorder on the critical behavior of a system has been examined on a rigorous level only for a few special cases in which a solvable Hamiltonian could be constructed.⁴⁰ This is not possible in the case of ${}^4\text{He}$, and one is able to construct general arguments only after making very strong assumptions. In particular, we assume, following the traditional approach for magnetic systems,¹⁶ that there exists a distribution of transition temperatures $T_c(\vec{r})$ which mimics the impurity density $n(\vec{r})$. This identification breaks down on length scales smaller than the *coarse-graining length* l characteristic of the porous medium. In the case of Vycor, l is expected to be of the order of the pore size (70–80 Å). In the following discussion of disordered systems, all physical quantities are assumed to be *coarse-grain averaged*. For example, $n(\vec{r})$ is the impurity density averaged over a sphere of radius l about \vec{r} .

Although it is not possible to justify rigorously the assumption of a T_c distribution and the adoption of the coarse-graining approach, evidence from full-pore experiments on various porous media suggests that the model is at least self-consistent. First, studies of ${}^4\text{He}$ -filled aerogels have established that the superfluid transition temperature T_c decreases as the aerogel density increases,^{11,41} indicating a correlation between the average impurity density $\langle n(\vec{r}) \rangle$ and the postulated average transition temperature, which is simply the observed T_c . Second, a reasonable choice for the coarse-graining length l in aerogel is the size of the silica grains⁴² (~ 100 Å) of which it is composed. The correlation lengths in the experiments described here range from 100–50000 Å, and so the inferred l is a reasonable unit of length near the superfluid transition.

C. Harris criterion

Following our assumption regarding the existence of a local transition temperature $T_c(\vec{r})$, we define the correla-

tion function

$$\tilde{g}(\vec{r}) = \langle T_c(\vec{r})T_c(0) \rangle - \langle T_c \rangle^2. \quad (8)$$

In the picture adopted here, this is proportional to the density-density correlation function $g(\vec{r})$ defined in Eq. (7). A corresponding correlation function can be defined for the local reduced temperature $t(\vec{r}) = [T - T_c(\vec{r})]/T_c$, where $T_c = \langle T_c \rangle$ is the average transition temperature.

Following Harris¹⁶ and Weinrib and Halperin,⁴³ we make the following assumption about the stability of the pure system critical behavior upon the introduction of disorder: *If the fluctuations in the reduced temperature vanish as $T \rightarrow T_c$, the asymptotic critical behavior will be the same as in the pure system.* In order to apply this criterion, we need a working definition of the fluctuations in the reduced temperature t . A physically intuitive choice, adopted by Weinrib and Halperin, is the statistical variance Δ^2 in t over a correlation volume ξ^d . They show that

$$\Delta^2 \sim \xi^{-d} \int_0^\xi x^{d-1} \tilde{g}(x) dx \quad (9)$$

as $T \rightarrow T_c$. As discussed above, we will assume that the correlation function $\tilde{g}(x)$ is proportional to the density-density correlation function $g(x)$ for the quenched disorder. For the case $g(x) \sim x^{-a}$, it is easy⁴³ to reduce Eq. (9) to

$$\Delta^2 \sim \xi^{-d} \begin{cases} \text{const}, & a > d, \\ \ln \xi, & a = d, \\ \xi^{d-a}, & a < d. \end{cases} \quad (10)$$

Using the asymptotic relation $\xi \sim \xi_0 t^{-\nu}$, Weinrib and Halperin obtain

$$\frac{\Delta^2}{t^2} \sim \begin{cases} t^{d\nu-2}, & a > d, \\ t^{d\nu-2} \ln t^{-\nu}, & a = d, \\ t^{a\nu-2}, & a < d. \end{cases} \quad (11)$$

The condition for the stability of the pure critical behavior requires $\Delta^2/t^2 \rightarrow 0$ as $t \rightarrow 0$. For the case $a \geq d$, commonly referred to as *uncorrelated disorder*, we thus require

$$d\nu - 2 = -\alpha > 0, \quad a \geq d, \quad (12)$$

where we have used the hyperscaling relation of Eq. (6). Eq. (12) is known as the Harris criterion¹⁶ and predicts that the critical behavior of the pure system will be stable if the specific heat exponent α is negative. The case $a < d$, known as *correlated disorder*, leads to the stability criterion

$$a\nu - 2 > 0, \quad a < d. \quad (13)$$

This is the extended Harris criterion of Weinrib and Halperin,⁴³ who showed that a new fixed point with its own set of critical exponents appears when this criterion is violated.

All ^4He -porous media systems realized to date are technically cases of uncorrelated disorder since the density-density correlation function decays exponentially at sufficiently long length scales. One would thus expect the usual Harris criterion to hold. Nonetheless, the observed critical behavior in the case of ^4He -filled aerogels is very different from the pure system for reduced temperatures down to 10^{-4} .^{2,10,11} We will return to this observation in the discussion section, but we note at this point two features that make the ^4He -porous media problem subtle. First, the Harris criterion is only barely satisfied for ^4He since the specific heat exponent $\alpha = -0.013 \pm 0.003$ (Ref. 39) is so close to zero. As a result, the variance in t approaches zero very slowly. The consequences of the marginality of the Harris criterion have been considered at a rigorous level by Narayan and Fisher,⁴⁴ who have calculated corrections to the pure exponents and critical amplitudes. These corrections decay logarithmically and hence slowly as $T \rightarrow T_c$. The second point is that the superfluid correlation length, which determines the volume over which the T_c fluctuations are averaged, is not always large compared to the length scales characterizing the disorder. For example, in the full-pore experiments of Wong *et al.*,¹¹ the correlation lengths varied from 10 to 10 000 Å in the range over which the critical behavior was characterized. Structural correlations in aerogels persist out to about 1000 Å, and so the traditional Harris criterion, Eq. (12), does not apply over much of the experimental range. In fact, the more stringent case, Eq. (13), applies over part of the power-law regime, and this stability criterion actually fails.¹¹

D. Annealed impurities

In the previous section, we considered the case of a quenched impurity, which is not in thermodynamic equilibrium with the ^4He system. In contrast, the ^3He in a ^3He - ^4He mixture is an *annealed* impurity: The density $n_3(\vec{r})$ of ^3He atoms is a thermodynamic variable which fluctuates about some equilibrium value n_3 . (The presence of a substrate in the case of a film results in a spatial variation in the equilibrium value of n_3 , but we shall ignore this complication here.) Annealed impurities were considered in a systematic way by Fisher,⁴⁵ who considered the case $\alpha \geq 0$ under the reasonable assumption that the critical behavior is unchanged as long as the system follows a thermodynamic path in which the “field” conjugate to the impurity density is held constant. For the case of a ^3He - ^4He mixture, this field is simply the chemical potential difference $\Phi = \mu_3 - \mu_4$. Since the convenient experimental path follows a line of constant impurity density n_3 , the constant-field condition is generally not satisfied, and Fisher’s argument would lead one to expect renormalized critical exponents if α were greater than zero. As noted above, α is actually slightly less than zero, which leads us to expect corrections to the ideal critical behavior that vanish very slowly as T_c is approached.

For the case of dilute mixtures, Ahlers⁴⁶ has noted that there is a large difference between the paths of con-

stant Φ and constant n_3 , but the resulting corrections depend only weakly on reduced temperature. This suggests that the effect on the critical exponents should be much less significant than the renormalization of the critical amplitudes. At higher concentrations, the analysis is complicated by the proximity of the tricritical point.⁴⁷ The experimental results for bulk mixtures partially confirm the view presented here. Ahlers⁴⁶ analyzed several second-sound experiments⁴⁸ and found a ρ_s exponent $\zeta = 0.68 \pm 0.01$ for ^3He concentrations less than 0.4, in agreement with the value measured for pure ^4He . The observed exponent increases for higher ^3He concentrations as the superfluid transition approaches the tricritical point. In contrast, the critical exponent for the specific heat is observed to depend on n_3 even for small concentrations.⁴⁹

E. Hyperuniversality and the heat capacity

One of the advantages of working with thin films is the ability to vary the density of ^4He and hence the critical amplitude ρ_{s_0} of Eq. (4). The procedure for deducing ρ_{s_0} will be discussed below. Although ρ_{s_0} is not necessarily proportional to the density, it does decrease as the density decreases. The Josephson relation [Eq. (3)] thus implies that the correlation length ξ_0 should change as well, although ξ_0 depends in turn on the transition temperature T_c . We shall demonstrate in this paper that the correlation lengths probed in thin-film experiments on aerogels are much longer than in the case of full pores. In addition to having important implications for the stability criteria discussed above, the longer correlation lengths should have a measurable effect on other thermodynamic quantities besides the superfluid density. We consider here the specific heat of the system.

The hyperuniversality hypothesis,⁵⁰ applied to superfluid ^4He by Hohenberg *et al.*,¹⁷ provides one means of connecting the correlation length $\xi(t)$ in the asymptotic regime with the singular part of the heat capacity $C_s(t)$. The hypothesis states that once normalized by the temperature T , the singular part of the free energy F_s in a correlation volume ξ^d is a constant for all systems in a given universality class. This assertion can be tested in the laboratory since $TC(T)$ is a second derivative of F with respect to T . Given that the heat capacity follows a power-law form in the transition region, a convenient form of the hyperuniversality hypothesis is

$$\frac{t^2 \alpha C_s(t) \xi(t)^d}{k_B} = X, \quad (14)$$

where C_s is the singular contribution to the heat capacity per unit volume and X is the universal constant of interest. In the case of ^4He , Eq. (14) can be rewritten using the Josephson relation, Eq. (3), giving

$$X = \frac{1}{k_B} \alpha t^2 C_s(t) \left(\frac{k_B T m^2}{\hbar^2 \rho_s(t)} \right)^3. \quad (15)$$

The value of X is 0.62 for bulk superfluid ^4He . Perhaps

the most useful corollary of Eq. (14) follows if we compare two systems, denoted primed and unprimed, within a given universality class. The ratio of the singular heat capacities at a particular reduced temperature t is then

$$\frac{C'_s(t)}{C_s(t)} = \frac{\xi(t)^d}{\xi'(t)^d}. \quad (16)$$

The hyperuniversality argument is constructed around the physical correlation length for $T > T_c$, which is proportional to the superfluid correlation length of Eq. (3) only within the asymptotic regime.¹⁷ This condition cannot be established in the case of the heat capacity measurements, and so the theory may break down, in which case the ratio X of Eq. (15) will be temperature dependent. For two different full-pore aerogels, Wong *et al.*¹¹ did observe that X was temperature dependent, changing by a factor of 2 over two orders of magnitude in reduced temperature. Since we are interested only in order of magnitude estimates of C_s , breakdowns of hyperuniversality at this level should not affect our conclusions.

In principle, precision measurements of the superfluid density and heat capacity allow for a stringent test of the hyperuniversality argument. The analysis, however, is complicated by the difficulty in determining the *singular* part of the heat capacity. A simple cusp in the heat capacity may occur at the superfluid transition due to a change in the excitation spectrum of the system. The relevant singular contribution will be a peak superimposed on this cusp. The heat capacity data for ⁴He films on Vycor seem to fit this description. The dominant temperature dependence shifts abruptly from $C(T) \propto T^2$ to $C(T) \propto AT + BT^2$ as T increases through T_c . A small peak, identified as the singular contribution, remains after this background is subtracted.^{18,19,34} The magnitude of this peak scales reasonably well with the correlation length in the manner predicted by Eq. (16).^{18,19}

IV. EXPERIMENTAL DETAILS

A. Substrates

The substrates used in these experiments were base-catalyzed aerogels prepared using the methanol process.^{51,52} The porosity of the sample used in the superfluid density experiments was 91% while that of the sample used in the heat capacity experiments was 95%. A detailed description of aerogels, including a comparison with Vycor glass, can be found in Ref. 11. We characterized our samples using transmission electron microscopy and found the typical cross-linked chain structure for silica aerogels. The chains are composed of silica spheres 50–150 Å in diameter. Small-angle x-ray-scattering data for samples from the same blocks as those used in the current experiments are shown in Fig. 1. The data are typical of base-catalyzed aerogels,^{53,54} and the measured structure factor $S(q)$ in both cases indicates that correlations exist for $q > 0.01 \text{ \AA}^{-1}$, corresponding to lengths less than about 600 Å. There is, however, a significant contribution from incoherent scattering at small q , leading to

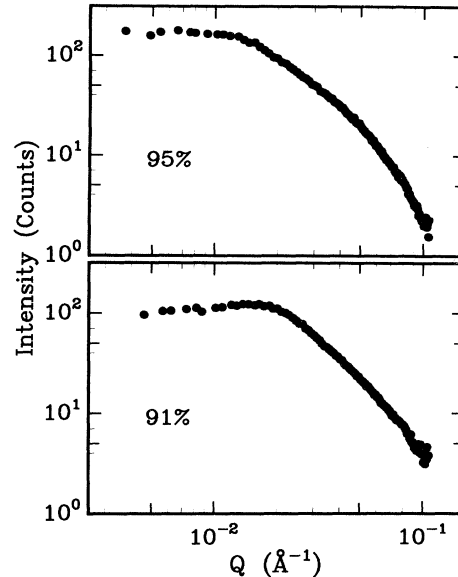


FIG. 1. Small-angle x-ray scattering data for the 95% (top panel) and 91% (bottom panel) porosity aerogels. The flat regions for momentum transfers $Q < 10^{-2}$ correspond to the uncorrelated regime.

the tails seen in Fig. 1. Aerogel thus lacks a characteristic length scale like that observed for Vycor.

B. Superfluid density measurements

The superfluid density was measured using the low-frequency torsional oscillator technique, which has been reviewed in detail by Wong *et al.*¹¹ The experimental cell in the experiments discussed here was used previously for the full-pore measurements on sample F of Ref. 11. The 91% porosity sample was a cylinder of diameter 0.56 cm and length 0.52 cm. The surface area, determined from a N₂ Brunauer-Emmett-Teller (B.E.T.) isotherm,⁵⁵ was $9.2 \pm 0.9 \text{ m}^2$. The aerogel was glued in place inside a small brass can on top of a BeCu torsion rod. The original resonant frequency of the cell was 294 Hz, and it was lowered to 250 Hz after the first experimental run. The oscillator was mounted in a massive brass annular container, which was in turn attached to the low-temperature stage of a dilution refrigerator by means of a second torsion rod. The resonant system comprising the brass annulus and the second torsion rod served as a vibration isolator.

The oscillator motion was driven and detected electrostatically using either the scheme described by Agnolet *et al.*⁵⁶ or that of Wong *et al.*¹¹ The oscillator could be run in either a constant amplitude or a constant drive mode. The constant drive mode was preferred because the intrinsic nonlinearity of the cell was small and the time constant was long enough (30 min at 10 mK) to make implementation of a stable feedback scheme, which is required for constant amplitude operation, very diffi-

cult. As a result, we only used the constant amplitude mode for the thicker films, where the dissipation swings were largest. Due to the long time constant at low temperatures, we did not always wait for the amplitude to ring in to its equilibrium value before proceeding with the measurement. This choice, along with the existence of a long-term drift in the amplitude background, meant that the dissipation data were only qualitative. The error introduced in the resonant period data, however, was negligible because of the small nonlinearity of the oscillator.

A torsional oscillator functions as a mass balance, with a shift in the resonant period (for fixed temperature T) that is proportional to the amount of ^4He locked to the substrate. The number of atoms in the vapor phase is negligible for the experiments reported here, and so we assume that any mass not locked to the substrate is superfluid. As the mass is increased from zero, the period increases linearly with a slope $X_n = (\partial P/\partial m)$ that corresponds to all of the adsorbed helium being locked to the substrate. X_n is the mass sensitivity of the oscillator. A graph of the period versus coverage at 10 mK is shown in Fig. 2. The data follow a line up to $35 \mu\text{mol}/\text{m}^2$. The slope of this line should be independent of temperature, which is easily checked by comparing the resonant period P of the empty cell as a function of temperature T with that measured for one of the nonsuperfluid coverages. As shown in Fig. 3, the two sets of data are offset by a constant, indicating that our interpretation of the nonsuperfluid film is correct.

As the coverage is increased above $35 \mu\text{mol}/\text{m}^2$, the period begins to fall below the extrapolation of the nonsuperfluid data in Fig. 2, indicating unlocked mass and hence the presence of a superfluid film. The resonant period $P(T)$ is shown for a superfluid film close to T_c in the top panel of Fig. 4. In this case, the data follow the empty-cell background down to 116 mK, below which

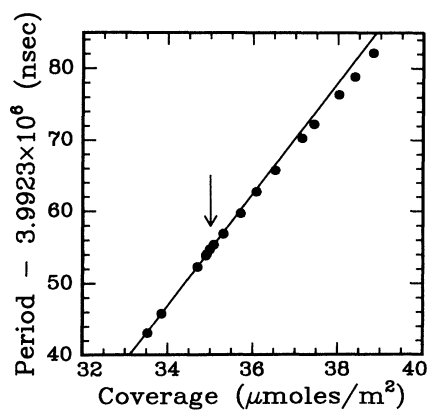


FIG. 2. The resonant period of the torsional oscillator is shown as a function of coverage at a temperature of 10 mK. The arrow marks the onset of superfluidity. The line is a fit of the data below the onset of superfluidity, and its slope determines the mass sensitivity X_n defined in the text.

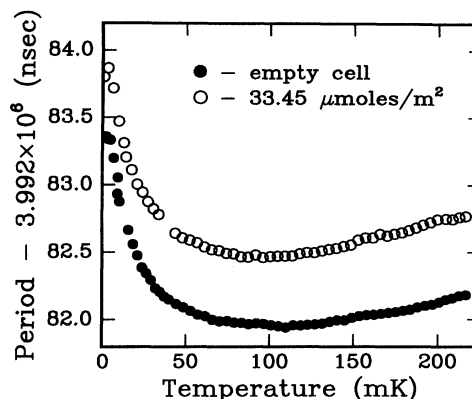


FIG. 3. The period of the empty cell for $T < 200$ mK compared with that measured for a coverage of $33.45 \mu\text{mol}/\text{m}^2$. The ordinate for the latter data has been shifted downward by 259.6 nsec. The two sets of data are offset by a constant within the limits imposed by the long-term period stability of the oscillator.

$P(T)$ drops. The superfluid period shift $\Delta P(T)$, indicated schematically in Fig. 4, is the difference between the nonsuperfluid background, adjusted for the total ^4He mass, and the measured data.

Although we will not focus on the dissipation in this paper, we show a representative set of data in the lower panel of Fig. 4. The most pronounced feature of these

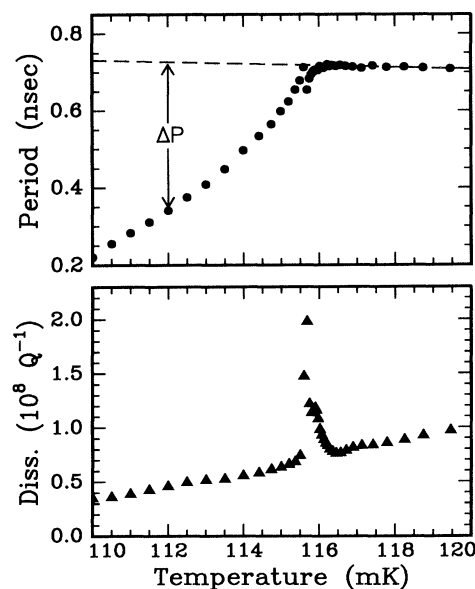


FIG. 4. Experimental data near the superfluid transition for a coverage of $37.4 \mu\text{mol}/\text{m}^2$. The resonant period, offset by a constant, is shown in the upper panel and the dissipation (also offset) in the lower panel. The dashed line is the nonsuperfluid background and the difference ΔP is the superfluid period shift.

data is the large peak near T_c . The accompanying distortion of the resonant period, which is measured at a fixed phase angle between drive and receive, indicates that the dissipation peak is due to a third-sound resonance. We do not know why this resonance is so pronounced, since a simple calculation shows that it is not a low-order mode for the boundary conditions of our cell. One of its important consequences was the limitation it imposed on the range of the power-law fits discussed below.

Several ^3He - ^4He mixture films were studied at the end of the second experimental run. After the introduction of the appropriate amount of each isotope, each film was raised to a temperature of 2–6 K to assure that the ^3He was distributed uniformly in the cell. The film was then cooled slowly to low temperature. The period shift was measured in the same manner as for the pure films.

C. Heat capacity measurements

The heat capacity measurements were conducted using a modification of the ac calorimetry technique of Sullivan and Seidel.⁵⁷ A sample of 95% porosity aerogel was held in place by compression inside a copper calorimeter. The sample diameter was 1.5 cm and the height was 0.025 cm. The surface area measured by a N_2 B.E.T. isotherm⁵⁵ was $2.0 \pm 0.2 \text{ m}^2$. The combination of the increased contact area resulting from the compression and the thin sample reduced the internal time constant of the calorimeter to less than 2 sec. The external time constant of approximately 100–250 sec for temperatures between 0.1 and 1 K was set by a 0.13-mm-diam wire linking the calorimeter to the mixing chamber of the dilution refrigerator. The frequency of oscillation for the ac calorimeter (0.1 Hz) fell within the window set by the internal and external time constants.

The measurements were conducted by applying a current to the calorimeter heater at a frequency of 0.1 Hz and measuring the resulting temperature oscillations using a lock-in referenced to the heater source. The temperature was measured using a ratio-transformer bridge operating at a much higher frequency (1 kHz). The power dissipated in the heater was typically 50 nW, much larger than the self-heating of the thermometer, which varied from 2 pW at 0.1 K up to 20 pW at 1 K. At these power levels, the resolution of the calorimeter was approximately 0.1% of the total heat capacity.

V. RESULTS

A. Superfluid density

1. Pure ^4He films

In Fig. 5 we show a composite plot of the period shift $\Delta P(T)$ for several coverages spanning transition temperatures between 50 mK and 1 K. These data are similar to those obtained for the ^4He -Vycor system.^{1,5,4} Both $\Delta P(0)$ and T_c decrease with decreasing coverage, but the overall shape of the curves is approximately the same for

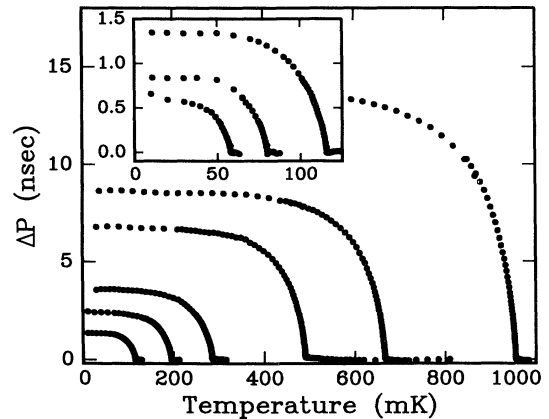


FIG. 5. Composite of the superfluid period shift ΔP for several coverages from both experimental runs. The data from the first run have been scaled by a constant to account for the change in the mass sensitivity of the oscillator between the runs.

all coverages with $T_c > 50$ mK. The noise in these data is most visible at the lowest temperatures, where the background corrections were largest. Other sources of noise, such as lingering effects of helium transfers, were irregular, affecting some coverages more than others. Finally, long-term period instability emerged as a problem for the lowest coverages, where the data accumulation period was several days. This may account for the difficulty in establishing a base line above the superfluid transition for coverages such as the lowest in Fig. 5.

The conversion from the period shift $\Delta P(T)$ to a superfluid density $\rho_s(T)$ relies on an important physical observation: ρ_s , which represents the stiffness of the superfluid order parameter, is a macroscopic quantity. In our experiment, we define ρ_s to be the mass density that is at rest in the laboratory frame of reference (or that moving at angular velocity $-\omega$ in the rest frame of a torsional oscillator). This approach is inspired by Josephson's definition of ρ_s as the proportionality factor relating the momentum density p_s of the superfluid to its velocity v_s .³⁰ In the current experiment, this factor is simply the "missing mass density" below T_c . We thus find

$$\rho_s(T) = \frac{\Delta P(T)}{V} \left(\frac{\partial P}{\partial m} \right)^{-1}, \quad (17)$$

where V is the *total* volume of the cell and $\partial P/\partial m$ is the mass sensitivity of the oscillator. The definition of Eq. (17) is sometimes referred to as the *coarse-grain averaged superfluid density*.

We emphasize that the above argument represents a macroscopic definition of ρ_s . The superfluid density is determined from a measurement of the angular momentum of the entire system, and the normalization factor for the density is the volume of the whole system. We shall show below that this definition of ρ_s is part of a

self-consistent description of the critical behavior of the system. An alternative definition of a superfluid density follows from the view that a torsional oscillator simply measures the hydrodynamic flow of a 2D superfluid film on the substrate. In this case, which is of interest to those studying the Kosterlitz-Thouless transition, the areal superfluid density at $T = 0$ is assumed to be $n - n_c$, where n is the areal density of the film and n_c is the critical coverage for the onset of superfluidity. At finite temperatures, this areal superfluid density is

$$\rho_{s_a}(T) = \frac{\Delta P(T)}{A} \left(\frac{\partial \Delta P(0)}{\partial m} \right)^{-1}, \quad (18)$$

where A is the surface area of the substrate. The scaling factor $X_s = \partial \Delta P(0) / \partial m$ is determined by plotting the period shift at $T = 0$ as a function of the coverage n as shown in Fig. 6. Except for a tail near the onset coverage of $34 \mu\text{mol}/\text{m}^2$, these data follow a line of slope X_s . For the aerogel substrate, the *superfluid mass sensitivity* X_s is much smaller than the total mass sensitivity X_n . The two sensitivities determine the χ factor

$$\chi = 1 - X_s / X_n, \quad (19)$$

which is discussed in detail by Wong *et al.*¹¹ For the aerogel used in the current experiment, $\chi = 0.90$ for thin films, much larger than the value of 0.16 measured when the pores were filled with ^4He .

If the correlation length were small compared with the aerogel microstructure, the appropriate superfluid density would be given by Eq. (18), and the χ factor would be the amount of superfluid mass dragged inertially by the substrate.¹¹ This hydrodynamic argument, however, breaks down for correlation lengths large compared to the structural length scales. In the thin-film ^4He -aerogel system, the correlation lengths in the critical region are at least an order of magnitude larger than the 100 \AA sil-

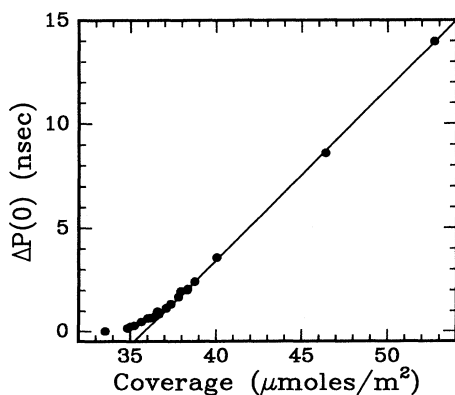


FIG. 6. The superfluid period shift at $T=0$, $\Delta P(0)$, is shown as function of coverage. The data from the first run have been scaled by the same constant as in Fig. 5. The slope of the line determines the superfluid mass sensitivity X_s defined in the text.

ica strands comprising the aerogel. The superfluid mass must be averaged over many correlation volumes, which is implicit in the macroscopic ρ_s defined in Eq. (17). Although we will be using the macroscopic ρ_s in this paper, we list both mass sensitivities as well as other calibration information in Table I.

We now consider the period shift data near T_c in the context of the preceding discussion. A subset of the data near T_c was selected for each coverage and fitted to a power law

$$\Delta P(t) = \Delta P_0 t^\zeta, \quad (20)$$

where $t = 1 - T/T_c$ is the reduced temperature. In each case, the subset spanned at least one order of magnitude in reduced temperature. The fitting procedure was a simplified version of the one described by Wong *et al.*¹¹ For each coverage, T_c was swept, and the data were transformed to $\log_{10}(\Delta P)$ vs $\log_{10}(t)$ for each value of T_c . Each data point was assigned a weight of $1/(\Delta P)^2$, corresponding to equal uncertainties in the raw period data, and a linear least-squares fit was then conducted for the given T_c . The procedure was repeated for each value of T_c , producing a set of triplets (T_c, χ^2, ζ) . The best-fit exponent was then determined by minimizing the normalized χ^2 with respect to ζ . Uncertainties in the various parameters were determined from the value of ζ at twice the minimum value of χ^2 . In cases where the normalized χ^2 was significantly greater than 1, the fitting range was modified and the above procedure was repeated. The results of the power-law fits for all of the coverages are compiled in Table II.

Figure 7 is a composite plot of $\log_{10}[\Delta P(t)]$ vs $\log_{10}(t)$

TABLE I. Information about the torsional oscillator for each of the experimental runs. The superfluid mass sensitivity defined in the text increased with coverage for run II, approaching the value listed in this table. The substrate velocity is estimated from the cell geometry and the various amplifier gains. The oscillator Q and substrate velocity are not available for run I.

	Run I	Run II
Frequency (Hz)	294	250
Moment of inertia (g cm ²)	0.25	0.34
Cell volume (cm ³)	0.13±0.01	0.13±0.01
N ₂ B.E.T. surface Area (m ²)	9.2±0.9	9.2±0.9
Mass Sensitivity (nsec/μg)	0.272	0.211
Superfluid mass Sensitivity (nsec/μg)	0.028	0.022 ^a
Q at 10 mK	NA ^b	1.6×10^6
Maximum substrate Velocity at 10 mK (cm/sec)	NA ^b	2×10^{-2}

^aLimiting value at high coverages.

^bNonapplicable (NA).

TABLE II. The results of the power-law fits for pure ^4He films adsorbed in 91% porosity aerogel glass. The three fitting parameters for each coverage are the transition temperature T_c , the critical amplitude ρ_{s_0} , and the critical exponent ζ . The critical amplitude for the correlation length, ξ_0 , is determined from ρ_{s_0} using Eq. (3). The range of the fit in reduced temperature is given in the last column of the table.

Coverage (μmol)	T_c (mK)	ρ_{s_0} ($\mu\text{g}/\text{cm}^3$)	Exponent ζ	ξ_0 (nm)	t range (\log_{10})
Full pore ^a	2164.991	3.74×10^5	0.811 ± 0.004	0.32	-4.3 to -1.7
486	954.6 ± 0.1	1800 ± 80	0.61 ± 0.01	29 ± 1	-3.5 to -1.8
428	665.8 ± 0.1	1130 ± 60	0.62 ± 0.02	31 ± 2	-3.5 to -1.8
399	490.4 ± 0.1	620 ± 30	0.56 ± 0.02	44 ± 2	-3.3 to -1.5
369	284.9 ± 0.2	390 ± 90	0.66 ± 0.07	41 ± 9	-3.0 to -1.5
357.2	194.9 ± 0.1	223 ± 10	0.62 ± 0.02	49 ± 2	-2.4 to -1.3
353.2	169.0 ± 0.2	198 ± 20	0.65 ± 0.04	47 ± 5	-2.4 to -1.3
349.8	147.0 ± 0.1	190 ± 20	0.67 ± 0.03	43 ± 5	-2.4 to -1.3
344.5	115.5 ± 0.1	114 ± 10	0.62 ± 0.04	56 ± 5	-2.4 to -1.3
341.9	100.8 ± 0.2	113 ± 20	0.67 ± 0.06	50 ± 9	-2.4 to -1.3
337.9	79.7 ± 0.1	75 ± 10	0.64 ± 0.06	59 ± 8	-2.4 to -1.3
333.3	57.8 ± 0.1	44 ± 5	0.60 ± 0.05	73 ± 8	-2.0 to -1.0
332.0	51.2 ± 0.1	39 ± 4	0.61 ± 0.05	73 ± 8	-2.0 to -1.0

^aReference 11.

for several coverages. The data for the filled cell¹¹ are included for comparison. It is immediately apparent that the exponent ζ corresponding to the slope of each of the thin-film curves is significantly smaller than the full-pore value. This observation will be discussed below, where we will focus on its relation to the critical amplitude ρ_{s_0}

in the asymptotic power law $\rho_s(t) = \rho_{s_0} t^\zeta$ and the correlation length $\xi(t) = \xi_0 t^{-\nu}$. We determined ρ_{s_0} from the the best-fit values of ΔP_0 using Eq. (17), and ξ_0 was then calculated using the Josephson relation, Eq. (3). The values of ρ_{s_0} and ξ_0 for each coverage are included in Table II.

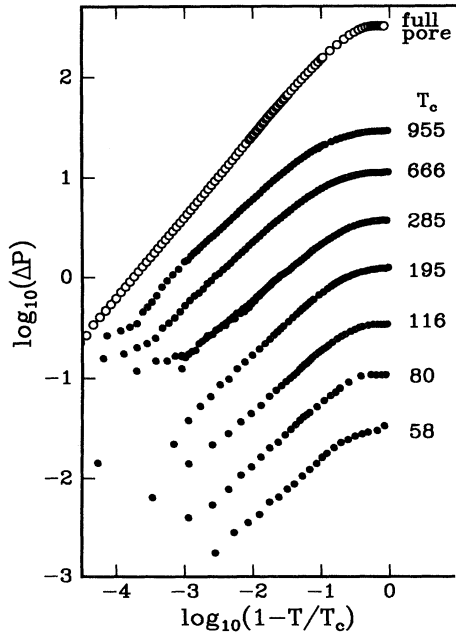


FIG. 7. The superfluid period shift ΔP for several coverages is shown as a function of the reduced temperature $t = 1 - T/T_c$ on a log-log scale. The curves have been offset for clarity. T_c for each coverage is indicated on the side of the plot.

2. ^3He - ^4He mixture films

Four mixture films were studied in detail. The first three of these were prepared for a single ^4He coverage of $357.2 \mu\text{mol}$. The ^3He coverages were 3.0, 7.0, and $28.0 \mu\text{mol}$. The next dose of ^3He ($112.1 \mu\text{mol}$) was sufficient to push the superfluid transition temperature below

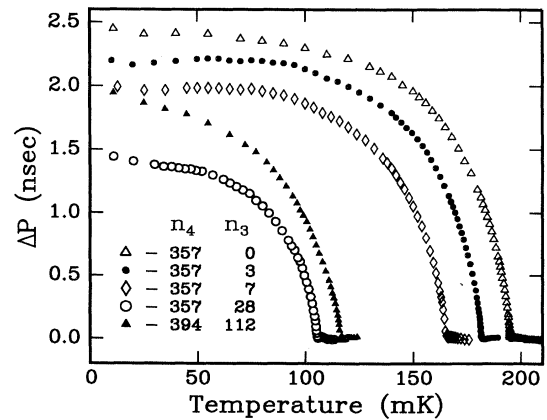


FIG. 8. Composite of the period shift data for the ^3He - ^4He mixture films. The legend indicates the ^4He and ^3He coverages for each film in μmol .

TABLE III. The results of the power-law fits for ^3He - ^4He mixture films adsorbed in 91% porosity aerogel glass. The respective coverages are given in the first two columns of the table. The other columns are the same as in Table II.

n_4 (μmol)	n_3 (μmol)	T_c (mK)	ρ_{s0} ($\mu\text{g}/\text{cm}^3$)	Exponent ζ	ξ_0 (nm)	t range (\log_{10})
357.2	0.0	194.9 ± 0.1	223 ± 10	0.62 ± 0.02	49 ± 2	-2.4 to -1.3
357.2	3.0	181.4 ± 0.1	186 ± 10	0.59 ± 0.02	54 ± 3	-2.4 to -1.3
357.2	7.0	165.3 ± 0.4	200 ± 40	0.67 ± 0.08	45 ± 9	-2.4 to -1.3
357.2	28.0	105.2 ± 0.2	103 ± 11	0.59 ± 0.05	57 ± 6	-2.2 to -1.2
394.1	112.1	116.2 ± 0.4	120 ± 40	0.64 ± 0.13	53 ± 20	-2.4 to -1.3

10 mK. The ^4He coverage was then increased for a series of measurements in which only the superfluid transition temperature was measured using a drift technique. A full set of data was obtained for a single film with ^4He and ^3He coverages of 394.1 and 112.1 μmol . A composite of the mixture film data is shown in Fig. 8 along with the data for the pure film of $n_4 = 357.2 \mu\text{mol}$. Power-law fits, the results of which are compiled in Table III, were conducted near the transition temperature using the algorithm described above.

B. Heat capacity

The heat capacity data were analyzed according to the procedure of Sullivan and Seidel.⁵⁷ The data for several coverages, corrected for the background contribution of the calorimeter, are shown in Fig. 9. We adopt a coarse-graining convention for the heat capacity as we have done for the superfluid density. We have therefore normalized the heat capacity by the total volume of the sample (0.044 cm^3).

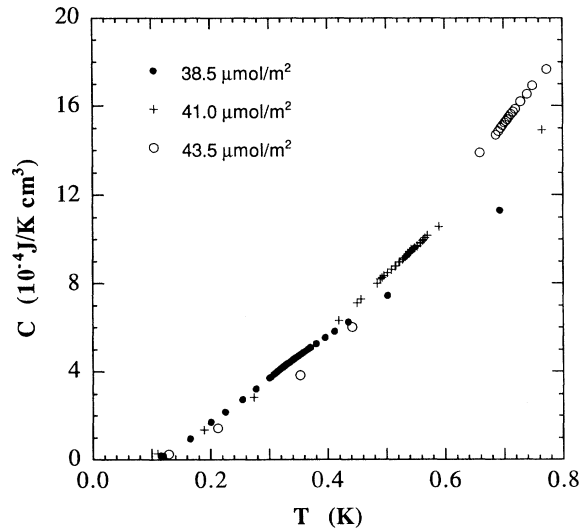


FIG. 9. The heat capacity C is shown as function of temperature for three ^4He films adsorbed in 95% porosity aerogel glass. The coverage for each film is indicated in the legend.

The data of Fig. 9 are most notable for the absence of any singularity like that observed for full-pore 95% porosity aerogel,^{11,12} even though each of the coverages is expected to have a superfluid transition. In interpreting these data, it is helpful to consider the case of ^4He films adsorbed in Vycor, for which a sharp signature of the transition appears in the heat capacity as well as the superfluid density.^{6,18} In Fig. 10, we show the heat capacity divided by the temperature for a coverage of 38.5 $\mu\text{mol}/\text{m}^2$ of ^4He on aerogel as well as a coverage of 30.9 $\mu\text{mol}/\text{m}^2$ adsorbed on Vycor. The latter set of data is taken from the work of Finotello *et al.*⁶ The onset of superfluidity at $T = 0$ for ^4He adsorbed in Vycor occurs at a coverage n_c of 27 $\mu\text{mol}/\text{m}^2$ as opposed to 34 $\mu\text{mol}/\text{m}^2$ for ^4He -aerogel. As a result, the superfluid coverages $n - n_c$ for the films in Fig. 10 are comparable. This figure shows

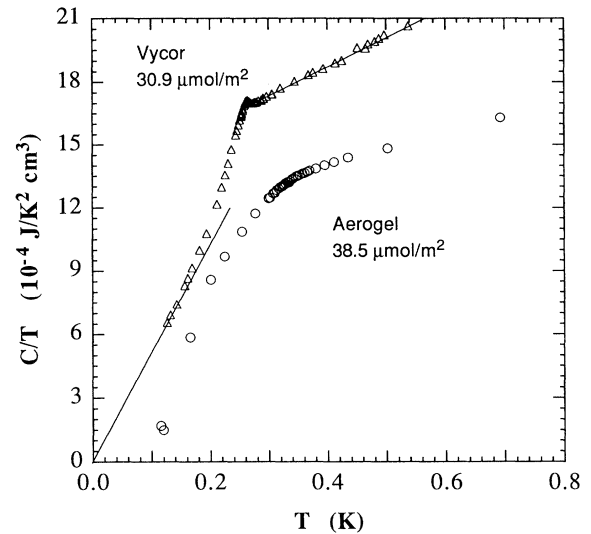


FIG. 10. The heat capacity divided by the temperature is shown as a function of temperature for a coverage of 38.5 $\mu\text{mol}/\text{m}^2$ adsorbed in 95% porosity aerogel and for a coverage of 30.9 $\mu\text{mol}/\text{m}^2$ adsorbed in Vycor. The data for Vycor have been divided by a factor of 3 to facilitate comparison. The two lines drawn through the Vycor data establish the background from which the singular part C_s of the heat capacity is determined according to the procedure discussed in the text.

two important consequences of the superfluid transition in the Vycor case. First, the overall temperature dependence of $C(T)$ is of the form $C(T) = AT + BT^2$ above T_c and is approximately quadratic below the superfluid transition. We have fit C/T to lines for ($T > T_c$) and for $T < 0.18$ K. The two fits are shown in Fig. 10, and they comprise a background heat capacity near the superfluid transition on which a peak is superimposed. This peak is the second important feature of these data. As discussed by Chan *et al.*,¹⁹ the two linear backgrounds reflect different excitation spectra in the superfluid and normal films. A kink in the C/T plot would thus occur even in the absence of fluctuations associated with the phase transition. The peak at the transition is a signature of fluctuations and is the strongest evidence for a diverging correlation length in the thin-film ^4He -Vycor system.

The aerogel data in Fig. 10 also show a crossover from roughly T^2 to $AT + BT^2$ dependence as T increases, with a rounded crossover region near the superfluid transition temperature expected for this coverage. A similar crossover is seen for another coverage as shown in Fig. 11. (The data for a third coverage do not extend to a high enough temperature to see clearly the crossover regime.) Another observation about the data in Fig. 11 is that the heat capacity *decreases* with increasing coverage at low temperatures, although it increases with coverage at high temperatures. This behavior is also seen for superfluid films on Vycor^{6,34,19} and is due to the fact that the excitations in the superfluid phase freeze out as $(T/T_c)^2$ while the total density of excited states at T_c increases very slowly with coverage. The rapid increase of T_c with coverage is thus the origin of the decrease in the heat capacity for $T < T_c$. The observation of this decrease for

films on aerogel is evidence that the films of Fig. 11 are superfluid at low temperatures. We conclude that, as for Vycor, the crossover of $C(T)$ to more linear behavior at higher temperatures is due to the superfluid transition.

VI. DISCUSSION

A. Superfluid density and the Harris criterion

One of the central results of this paper, shown in Fig. 7, is the dramatic contrast between the critical behavior of thin films of ^4He in aerogel and that seen for the full-pore case. The critical exponent ζ for full pores is 0.811 ± 0.004 , but all of the exponents measured for thin films fall between 0.56 and 0.67 (all but one are above 0.60). This is very different from the situation seen in the case of Vycor, where the observed critical behavior is essentially the same for full pores and all films with $T_c > 50$ mK. We now address two questions. First, why are the films so different from full-pore aerogel? Second, what is the actual significance of the measured exponent ζ ?

The difference between films and full-pore aerogel can be understood at two levels, the first of which involves consideration of the geometry of the substrate. The volume of the aerogel comprises two pieces, the open volume and the glass network. In the current experiment, the open volume is 91% of the total sample volume. The surface area of the substrate itself is 9.2 ± 0.9 m². From these numbers, we infer that the amount of helium in the cell drops by a factor of approximately 10 when the full-pore system is replaced by a film with a transition temperature of 1 K. The corresponding drop in the case of Vycor is only a factor of 2.⁵ The connectivity of the open volume is also much higher than that of the complementary substrate surface. This observation is consistent with the view of aerogel glass as a complex network of strands. ^4He can flow easily through the pore volume, but it must follow a very tortuous path along the surface. The experimental tortuosity factors χ reflect this picture. The full-pore χ factor is 0.16, and the χ factor measured for films with $T_c > 200$ mK is 0.90. This situation is again in sharp contrast with Vycor, where the full-pore and thin-film χ factors are essentially the same.⁴

The combined effects of these geometrical considerations reduce the superfluid period shift at $T = 0$, $\Delta P(0)$, from 3.0 μsec for full pores to 0.018 μsec for a film with a transition temperature of 955 mK. The difference in the critical amplitude ρ_{s0} is correspondingly large: It drops from 0.374 g/cm³ to 2.0×10^{-3} g/cm³. As the coverage is reduced further, however, ρ_{s0} decreases much more slowly, as can be seen in Table II. Note that although ρ_{s0} continues to decrease with decreasing coverage, T_c also decreases, so that the correlation length ξ_0 determined from Eq. (3) (see Table II) increases only by a factor of 3 as T_c drops from 955 to 50 mK. In contrast, ξ_0 increases by a factor of 100 as the system moves from the full-pore case to the thickest film.

Full-pore ^4He in aerogel is thus a dense well-connected system while its thin-film counterpart is dilute and poorly connected. In this sense, it is not surprising that the ob-

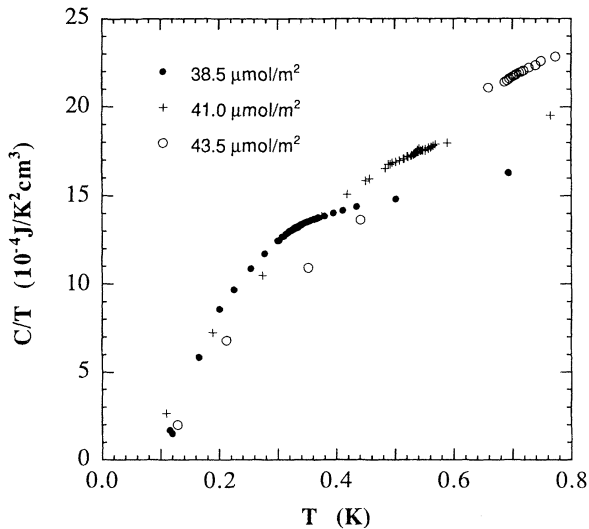


FIG. 11. The heat capacity divided by temperature is shown as a function of temperature for each of the coverages of Fig. 9.

served critical behavior is different in each case. In the case of Vycor, there is no drastic change in the morphology on passing from full-pore ^4He to a thin film, and so the critical behavior is unchanged. A more difficult question is the meaning of the power-law exponent measured in each experiment. In particular, does the *measured* power law correspond to the *asymptotic* critical behavior that would be observed at arbitrarily small reduced temperatures? This question can be answered only by application of the appropriate forms of the Harris criterion, Eq. (11). We consider two cases. The first of these includes systems in which the structural disorder is uncorrelated on the scale of the superfluid correlation length $\xi(t)$ over the entire power-law region observed for ρ_s . We find that thin-film aerogel falls in this class. In the second class of systems, which includes full-pore ^4He -aerogel, the disorder is correlated on the scale of $\xi(t)$ over a significant fraction of the power-law region.

The distinction between the two cases is apparent in Fig. 12, which shows the small-angle x-ray scattering data for 91% porosity aerogel. These data represent the Fourier transform of the structural correlation function for the porous medium. The flat regions for small values of q (large length scales) correspond to uncorrelated disorder. We have placed bars on the plot indicating the range of length scales over which $\xi(t)$ varies in the power-law regions found in the superfluid density measurements. For full-pore aerogel, $\xi(t)$ overlaps extensively with the region of correlated disorder at large q .

For the thin-film case, in which the disorder is essentially uncorrelated, we expect the traditional Harris criterion, Eq. (12), to apply, and the critical behavior should be that of the pure system. Unfortunately, corrections to the asymptotic critical behavior may still be large because of the small value of $|\alpha|$ in the pure system. It is therefore not possible to answer *a priori* our question as to whether the measured critical behavior is in fact the *asymptotic* critical behavior. Nonetheless, the average value of the exponent ζ for thin films on aerogel is 0.63 ± 0.03 , much closer to value found for pure ^4He than

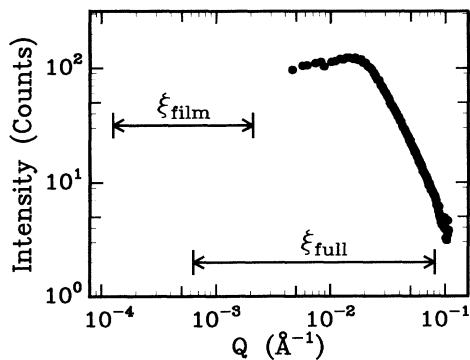


FIG. 12. The small-angle scattering spectrum for the 91% porosity aerogel is shown with bars indicating the range of the superfluid correlation length in the power-law regime for thin films and full pores.

for full-pore aerogel.

Full-pore aerogel remains an exceptional case. Figure 12 indicates that the range of ξ in the power law regime overlaps strongly with the region of correlated disorder. As for other base-catalyzed aerogels,⁵⁴ the structure factor of Fig. 12 does not conform to a simple power law in q , even for large wave numbers. We thus do not have simple power-law correlated disorder of the type considered in Sec. III C. If, however, we attempt to force a fit to a power law in q , we find $S(q) \sim u + vq^{-w}$, where w is between 2 and 3, giving a spatial correlation function $g(x) \sim x^{-a}$, where a is between 0 and 1. Because the decay of $g(x)$ is so slow, the ordinary Harris criterion, Eq. (12), does not apply. The extended Harris criterion, Eq. (13), applies instead, and it actually fails, implying that the critical behavior is not approaching that of the pure system. Although this statement is consistent with the experimental observations, it is not immediately clear how to account for the crossover back to uncorrelated disorder for $\xi > 1000 \text{ \AA}$, where the original Harris criterion should apply. Somehow, the large corrections due to the correlated disorder must eventually die out as the variance Δ^2 of Eq. (9) is evaluated on longer and longer length scales. The close proximity of α_{pure} to zero, however, ensures that these corrections disappear only logarithmically with reduced temperature.⁴⁴ The measured exponent $\zeta = 0.81$ for this aerogel probably reflects some of these corrections. An open question remains as to whether the same exponent would be measured if the correlated disorder extended out to infinite length scale. As discussed by Wong *et al.*,¹¹ the measured ζ does not agree with the prediction of Weinrib and Halperin⁴³ for infinite-range power-law correlated disorder.

We have presented here a mixed interpretation, appealing to the Harris criterion for systems which are neither strictly correlated nor homogeneously disordered. The results for the ^4He -aerogel system are consistent with the existence of a correlation length $\xi(t)$ which diverges at the superfluid transition. In both the full-pore and thin-film cases, $\xi(t)$ is inferred from the superfluid density $\rho_s(t)$ using the Josephson relation Eq. (3). The critical amplitude ρ_{s0} for thin films is at least two orders of magnitude below the full-pore value, thus leading to a large increase in $\xi(t)$ for thin films. The difference in the correlation lengths implies that corrections to the asymptotic critical behavior should be much weaker for thin films than for full-pore ^4He -aerogel. The measured values of the superfluid density exponent ζ are consistent with this argument.

Although the argument presented here works at a qualitative level, we are far from a global understanding of the effects of correlated disorder. For example, ξ for full-pore Vycor overlaps with the region of correlated disorder (albeit less dramatically than full-pore aerogel), but the measured exponent $\zeta = 0.67 \pm 0.03$ (Ref. 2) agrees with that found for pure ^4He . We have no explanation as to why helium in Vycor should be less "sensitive" to disorder than helium in aerogel. Xerogel, studied by Chan *et al.*,² might be regarded as an intermediate case. Like aerogel, the full-pore exponent is anomalous (0.89 ± 0.02), but it only drops slightly for a film with a T_c of 1 K. We

can say little about this result, but we note that the superfluid correlation lengths are almost the same in both the thin-film and full-pore xerogel systems. The reasoning of this paper would thus lead one to expect similar critical behavior in the two systems.

B. Superfluid density of mixture films

The most important observation about the critical behavior of the mixture films is that it is not significantly different from that of the pure films, even though a relative ^3He concentration of less than 1% decreases T_c by 7%. This observation is consistent with the observations for bulk mixtures and the theoretical arguments presented above.^{46,47} For relative ^3He concentrations below 2%, we find almost no effect on the temperature dependence of the period shift over the entire temperature range of the experiment. This is shown in Fig. 13, in which the period shift $\Delta P(T)$, normalized by the transition temperature T_c , is shown as a function of T/T_c for all of the mixture films. The pure film ($n_4 = 357.2 \mu\text{mol}$) and the first two mixture films all follow a nearly identical curve. The third mixture film shows some deviations at low temperatures, and the fourth film ($n_4 = 394 \mu\text{mol}$, $n_3 = 112 \mu\text{mol}$) is significantly different. Figure 13 indicates that for a given value of $\Delta P(0)$, the films with higher ^3He concentrations have a lower T_c , due to new excitations which deplete ρ_s as T increases from zero. For the case of the most concentrated film in Fig. 13, the excitations are so prominent that there is no appreciable region of zero slope in ΔP as $T \rightarrow 0$. As $T \rightarrow T_c$, however, only the critical fluctuations are important, and the data for all of the films collapse onto a universal curve.

Unlike the accumulated data for pure films, the temperature dependence of the period shift of mixture films on aerogel is very different from that found for mixture

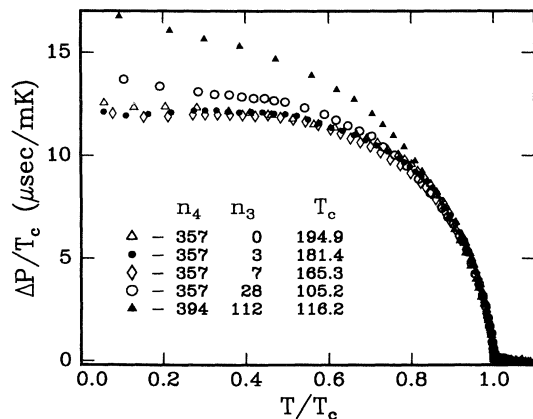


FIG. 13. The period shift data for the mixture films with ΔP and T scaled by T_c . The ^4He and ^3He coverages in μmol and the transition temperature in mK are indicated for each data set.

films on Vycor by Smith *et al.*⁵⁸ These authors found that ^3He concentrations of less than 1% broadened the superfluid transition and radically changed the overall temperature dependence of the period shift. We suspect that the small open volume of Vycor inhibited the proper annealing of these films, so that the ^3He was not uniformly distributed. The films in the current experiment were carefully annealed. Furthermore, the overall dependence of T_c and $\Delta P(T)$ on ^3He coverage for aerogel is consistent with that found by McQueeney⁵⁹ for mixture films on Mylar, even though the latter system is two dimensional. The current data, however, are far from complete. In particular, they do not address the interesting issue of phase separation in mixture films,⁶⁰ which can be explored only at higher ^3He concentrations.

C. Heat capacity and hyperuniversality

As discussed above, the heat capacity of ^4He films on aerogel exhibits a crossover from a roughly T^2 dependence at low temperature to an $AT + BT^2$ dependence at high temperatures. We have argued that this crossover is due to superfluidity. Unlike for Vycor, however, there is no peak in the heat capacity at the superfluid transition. We find that this observation is consistent with the hyperuniversality hypothesis discussed above. For the discussion here, the most convenient formulation of the hypothesis is Eq. (16), which states that the singular part of the heat capacity per unit volume should scale with the inverse correlation volume for systems within the same universality class. We assume here that thin-film ^4He -Vycor and thin-film ^4He -aerogel are in the same universality class as bulk ^4He , which is consistent with the superfluid density measurements. Our argument, however, should hold even if this assumption is relaxed, provided that the constant X of Eq. (15) varies by less than order of magnitude among the three systems.

In Table IV we list the correlation length amplitude ξ_0 in the power law $\xi(t) \sim \xi_0 t^{-\nu}$ for bulk ^4He and for the two thin-film systems considered in Fig. 10. The correlation length for the 95% aerogel is assumed to be the same as for the 91% aerogel used in the ρ_s measurements. This assumption probably underestimates the correlation length, which should increase with porosity, but a larger

TABLE IV. Correlation length ξ_0 and the factor $\xi_{0\text{bulk}}^3/\xi_0^3$ determining the size of the singular part of the heat capacity, relative to bulk, according to the hyperuniversality argument discussed in the text.

System	T_c (mK)	ξ_0 (nm)	$\xi_{0\text{bulk}}^3/\xi_0^3$
Bulk ^4He	2172	0.36 ^a	1
^4He -Vycor	260	10 ^b	5×10^{-5}
^4He -aerogel	300	40	7×10^{-7}

^aReference 29.

^bReference 32.

value would strengthen our conclusions. The correlation length for the film on Vycor is estimated from the data of Crooker,³² and the bulk value is taken from the review of Ahlers.³⁷ The third column of Table IV gives the ratio $\xi_{0\text{bulk}}^3/\xi_0^3$, which is the factor by which the singular part of the specific heat should be reduced from the bulk value according to Eq. (16). Finotello *et al.*,⁶ Murphy and Reppy,¹⁸ and Chan *et al.*¹⁹ have verified that the scale factor for films on Vycor is correct within an order of magnitude for several different transition temperatures. Since ξ_0 for a thin film on aerogel with a T_c of 300 mK is approximately 4 times larger than for a Vycor film with $T_c = 260$ mK, the corresponding singularity in C should be smaller by a factor of about 64 and is therefore too small to be observed at our resolution.

To conclude, we recall that the Josephson relation, Eq. (3), states that ξ_0 is inversely proportional to the critical amplitude ρ_{s_0} . The absence of a singular contribution to the heat capacity for ^4He adsorbed in aerogel is thus integrally related to the small superfluid density found in the torsional oscillator measurements. The dramatic reduction in ρ_s that occurs upon going from full-pore aerogel to a thin film is mirrored in the heat capacity. The singularity in the heat capacity observed for full-pore aerogels of 91% and 95% porosity¹⁰⁻¹² disappears in the case of thin films. The observation for Vycor is exactly the opposite: The singularity observed for thin films disappears when the pores are filled. The key point here is that the reduction in C_s in going from full-pore Vycor to thin films is small (less than a factor of 10 for $T_c > 100$ mK) because ρ_{s_0} decreases only gradually as helium is removed from the system. The nonsingular part of the heat capacity decreases much more rapidly,⁶ allowing C_s to be resolved in the thin-film limit.

VII. CONCLUSIONS

The measurements discussed in this paper confirm that thin films of ^4He on aerogel are similar in many respects to those adsorbed on Vycor. For films with $T_c > 50$ mK, power-law behavior in ρ_s is observed over at least one order of magnitude in reduced temperature t . The observed exponent $\zeta = 0.63 \pm 0.03$ is much closer to that found for bulk helium and ^4He -Vycor than the exponent measured for full-pore aerogel. This observation is consistent with the small values of the critical amplitude ρ_{s_0} for thin films, which imply much larger correlation lengths than for full-pore aerogel. Because of this large length scale, the correlated disorder of the aerogel is much less important for thin films of ^4He than in the full-pore case. The large correlation lengths are also consistent with the observed heat capacity, which differs qualitatively from that found for films on Vycor only in the absence of a peak singularity at the superfluid transition. Hyperuniversality predicts that the singularity should shrink as $1/\xi^3$, and it therefore could not be resolved given our experimental resolution. Finally the power-law behavior in ρ_s seen for pure films was also observed for several mixture films, in general agreement with universality arguments. Our data for mixtures are, however, qualitatively different from those found in the Vycor case.

We have only constructed an argument that is self-consistent in the sense that we cannot demonstrate explicitly the existence of a correlation length ξ that diverges as $T \rightarrow T_c$. Since the regions of power-law behavior seen for films are much smaller than for bulk helium or even full-pore ^4He -aerogel and the heat capacity singularity is apparently immeasurably small, the indirect evidence for a diverging correlation length in films is correspondingly weaker. We cannot, however, explain all of the results discussed in this paper without invoking a diverging correlation length. In spite of this, we emphasize that the theoretical arguments, which invoke abstractions such as a T_c distribution, do not constitute an *understanding* of superfluidity in ^4He films adsorbed in porous media. In this sense, we remain handicapped by the relatively poor understanding of the superfluid transition in pure ^4He , a situation which persists in spite of the fact that the various critical exponents can be calculated with remarkable precision.

How can the experimental situation be improved? One question that can in principle be addressed with the aerogel system is whether a crossover back to full-pore behavior is seen for films with transition temperatures above 1 K. Unfortunately, capillary condensation will complicate the interpretation of the data in this regime, and so careful chemical potential measurements will also be required to determine when liquid begins to condense in the aerogel. Another recourse is to continue with thin-film studies using other systems, but given the time-consuming nature of these measurements, it would be wise to address some specific questions. First, can the power-law region be extended to smaller reduced temperatures? To this end, new materials must be exceptionally uniform in order to minimize rounding due to T_c fluctuations at long length scales. Furthermore, experimental effort will have to be devoted to conquering the sound-resonance barrier that limited the range of the measurements described here. A second question addresses the hyperuniversality argument, which has been applied in this paper to systems in which the heat capacity singularity is expected to be small. Can we find a system in which the heat capacity singularity is only one-tenth or one-hundredth the size of that for bulk helium as opposed to 1 part in 10^5 ? Meeting this criterion for thin films will be particularly difficult, because it requires increasing the surface area per unit volume while preserving a connected structure with a low tortuosity.

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