# Superfluid Critical Behavior in $^4$He-Filled Porous Media

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We have observed sharp cusplike heat-capacity singularities, coincident with the superfluid transition, in two $^4$He-filled aerogel glasses. These features are interpreted as conclusive evidence of genuine critical behavior at the superfluid transition in these systems. The cusplike heat-capacity singularities confirm that $^4$He-filled aerogels are not in the same universality class as bulk helium.

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The nature of the superfluid transition in $^4$He-filled porous media has been the subject of a prolonged debate. Much of the original interest in this problem was based on the observation that these systems exhibit finite-size effects.[1] In contrast, others have argued that the critical behavior in these systems must ultimately be determined by the long-range 3D connectivity of the pore structure.[2,3] The latter point of view maintains that the existence of power-law behavior in the superfluid density[3] is evidence of genuine critical behavior at the depressed superfluid transition temperature $T_c$. The actual value of the superfluid density exponent is believed to reflect the long-range structure of the porous medium and is not necessarily equal to the bulk liquid value of $0.6717 \pm 0.0004$.[4] The experimental evidence in support of this interpretation has not been conclusive, however, for the singular behavior that is expected for other physical properties at the critical point has not been observed.

In particular, no one has observed a singularity in the heat capacity of any porous medium that has been *completely filled* with liquid $^4$He.[5,6] This absence is significant for two reasons. First, a singularity in the heat capacity is considered to be a principal feature of a continuous phase transition. Second, Finotello *et al.*[6] recently reported the observation of singular behavior in the heat capacity of *thin films* of $^4$He adsorbed in xerogel[7] and Vycor[8] glasses, although no singularity could be resolved when the pores were filled with helium. These results motivated us to investigate $^4$He-filled aerogels,[9] in which well-defined power-law behavior in the superfluid density has been observed with an exponent distinct from that of either $^4$He-filled xerogel or Vycor.[3,10] Aerogel glass is by far the most porous of these materials, allowing for a significantly larger volume of $^4$He than is the case with the other porous glasses. This Letter reports the observation of sharp heat-capacity singularities, coincident with the superfluid transition, in two $^4$He-filled aerogel glasses. We will also discuss how these observations are consistent with arguments based on hyperuniversality (or two-scale-factor universality),[11] which relates the singular component of the heat capacity to the superfluid density.

The aerogel samples used in the work reported here had porosities of 94% and 91% as opposed to typical porosities of 30% for Vycor and 60% for xerogel.[3] The structure and properties of aerogels are reviewed in the book edited by Fricke.[9] The glass consists of a tenuous mesh of $SiO_2$ chains. Neutron-scattering data[12] indicate that the structure is self-similar over a range of length scales extending from 10 to approximately 150 nm for aerogels of density comparable to that of our samples. The size of this self-similar regime as well as other structural details depends on the manufacturing process, however, and our samples have not been characterized. As discussed in a previous paper,[3] superfluid density measurements indicate that structural correlations at length scales up to several hundred nanometers may play a role in determining the superfluid critical behavior in $^4$He-filled aerogels.

The heat-capacity measurements reported here were done using an adiabatic calorimetry technique described elsewhere.[13] The critical components of the calorimeter were a magnetic thermometer with root-mean-square temperature resolution of at least 3 nK from 1.4 to 2.2 K,[14] and a low-temperature valve, which allowed us to thermally isolate the experimental cell from the remainder of the cryostat with a time constant of approximately $2.6 \times 10^5$ sec. A small reservoir containing a capacitive level sensor was included in the cell to ensure that the measurements were done at saturated vapor pressure.

Figure 1 shows the specific heat for the 94%-porosity aerogel on a variety of temperature scales. On a global scale, the singularity is comparable in size to the $\lambda$ anomaly found in pure helium. Closer examination, however, reveals the presence of two singularities. The logarithmic singularity, 4 mK above the main cusp singularity, is due to the presence of a small amount of bulk liquid in the cell. We used the existing heat-capacity data[15] for pure helium to subtract this contribution. This analysis led us to conclude that as much as (10–13)% of the pore volume inside the 94%-porosity aerogel may consist of large voids that contribute to the bulk singularity. A similar analysis for the 91%-porosity sample indicated that (6–7)% of the pore volume consists of large voids. The data for both samples, after the bulk subtraction, are shown in Fig. 2.

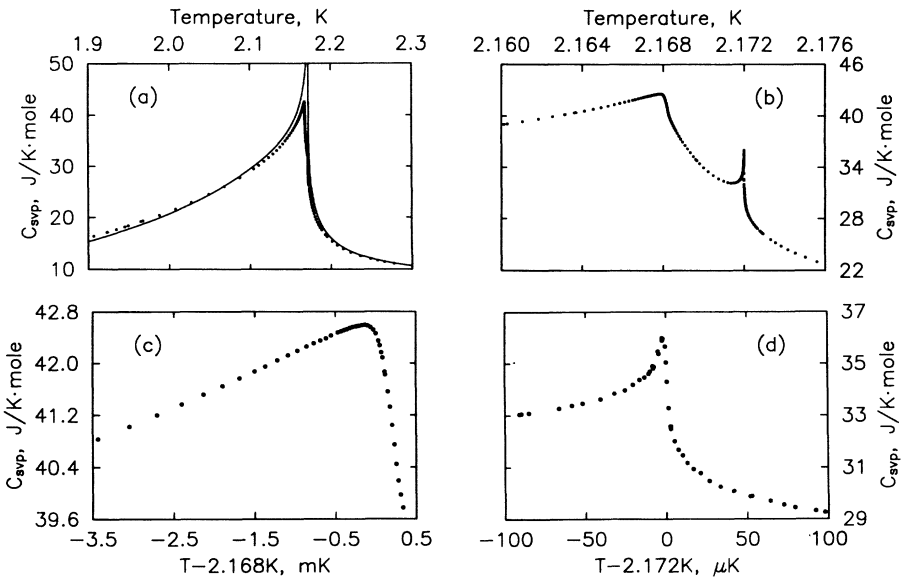


FIG. 1. The specific heat at saturated vapor pressure $C_{svp}$ of $^4$He-filled 94%-porosity aerogel, shown on four different temperature scales. In (a), the solid line represents the heat capacity of pure helium (Ref. 15). Two very distinct singularities are visible on the scale of (b). The gel-system transition is emphasized in (c), and the bulk transition is shown in (d).

The measured $T_c$'s for samples cut from the same block of aerogel have varied by as much as 1 mK for reasons which are not known. The existence of a distribution of $T_c$'s required that the heat capacity and superfluid density be measured simultaneously in order to verify that the observed heat-capacity singularity is coincident with the superfluid transition. An experimental cell was fabricated for this purpose by winding a heater coil around the head of a torsional oscillator which contained an aerogel sample of 94% porosity. A carbon-glass resistance thermometer was thermally anchored to the cell, which was isolated from the remainder of the cryostat by a packed-powder superleak. The results of the simultaneous measurement are shown in Fig. 3. The smallest temperature step size possible with this cell was only 0.44 mK because of the constraints imposed by the use of resistance thermometry and the superleak isolator. Figure 3 shows that the superfluid density and heat-capacity signatures are coincident within the resolution allowed by this step size.

The absence of an experimentally observable heat-capacity singularity in $^4$He-filled Vycor, as well as the existence of large heat-capacity singularities in the $^4$He-filled aerogels, is in general agreement with arguments based on hyperuniversality[11] and the relation[16] $\xi(T) = k_B T m^2/\hbar^2 \rho_s(T)$ between the correlation length $\xi$ and superfluid density $\rho_s$. The general hypothesis of hy-

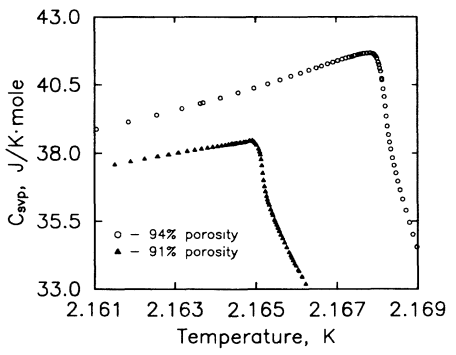


FIG. 2. The specific heats at saturated vapor pressure of two $^4$He-filled aerogels after the contributions due to bulk liquid have been subtracted from the data. The bulk transition temperature is $T_\lambda = 2.172$ K.

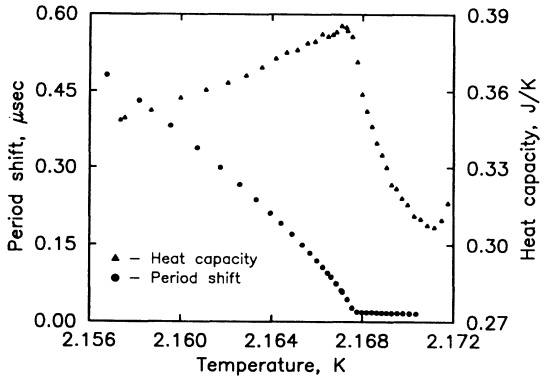


FIG. 3. The simultaneous measurement for the $^4$He-filled 94%-porosity aerogel demonstrating that the superfluid density and heat-capacity singularities are coincident. Note that the superfluid density is proportional to the period shift and that, for this figure only, each heat-capacity point is averaged over a 0.44-mK interval.

peruniversality is that the free energy in the correlation volume $\xi^3$ is a constant, independent of temperature, within a universality class. Specifically, we can predict the singular part of the specific heat per unit volume, $C(t) \sim (A/\alpha)|t|^{-\alpha}$ as $t \to 0$, where $t = 1 - T/T_c$, from our knowledge of the superfluid density, $\rho_s(t) \sim \rho_{s0} t^{\zeta}$, and the universality of the dimensionless constant $X$:

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

For two systems (denoted primed and unprimed) within the same universality class, the relation (1) can be rewritten as

$$A'/A = (\rho'_{s0} T_c / \rho_{s0} T'_c)^3 , \quad (2)$$

where we have used the power laws stated above for $C(t)$ and $\rho_s(t)$. This provides a means of predicting the relative magnitudes of the critical amplitudes $A$ and $A'$ from superfluid density measurements. Recent analyses for the case of $^4$He films adsorbed in Vycor found that the relative sizes of the singular contributions to the heat capacity agree with estimates based on relation (2) within an order of magnitude.[17]

Renormalization-group theories such as hyperuniversality apply to macroscopically homogeneous systems; therefore, we treat the porous structure as an effective medium and average $\rho_s$ over a volume much larger than the correlation volume. In pure helium, it does not matter how big a volume we average over in order to define $\rho_s$. In a $^4$He-filled porous medium, however, the divergent correlation volume must eventually encompass both the liquid and the porous structure. A second consideration is that the superfluid mass in a porous medium is reduced by tortuosity corrections[18] to a value smaller than that which would be measured on length scales smaller than the pore size. In general, the effect of this coarse-grain interpretation for both the volume and the superfluid mass is that $\rho_s$ decreases as the porosity of the medium decreases.

The critical amplitude $\rho_{s0}$ for the superfluid density power law is 0.35 g/cm$^3$ for pure helium, 0.41 g/cm$^3$ for $^4$He-filled 94%-porosity aerogel, 0.37 g/cm$^3$ for $^4$He-filled 91%-porosity aerogel, and 0.010 g/cm$^3$ for $^4$He-filled Vycor. Typical depressions of $T_c$ below $T_\lambda$ are 5 and 7 mK for the 94%- and 91%-porosity aerogels, respectively, and 220 mK for Vycor.[3,10] The superfluid density exponent $\zeta$ is the same, within error limits, for pure helium and $^4$He-filled Vycor, suggesting that these systems are in the same universality class. The hyperuniversality relation (2) then predicts that the singular part of the heat capacity is about a factor of $10^4$ smaller in $^4$He-filled Vycor than in bulk helium and hence has been too small to resolve in previous experiments because of the large nonsingular background.[6] The aerogel systems are apparently in a different universality class than bulk helium, so that the relation (2) is not applicable. If we assume, however, that the constant $X$, which is 0.63 for bulk $^4$He, is always of order unity, then the relation (1) indicates that the large heat-capacity singularity that is seen in the $^4$He-filled aerogels is reasonable.

We now turn to a brief discussion of scaling in $^4$He-filled aerogels. Both of the systems studied in the work reported here are characterized by a superfluid density power law for reduced temperatures $t$ in the range $10^{-4} < t < 10^{-2}$. The exponent $\zeta = 0.81 \pm 0.01$ was measured for both aerogels.[3,10] The hyperscaling argument of Josephson[19] predicts that the heat capacity should follow a power law $C(t) = (A/\alpha)|t|^{-\alpha} + B$ with an exponent $\alpha = -0.43 \pm 0.03$ over the same reduced temperature range. The heat-capacity data shown in Fig. 2 do not conform to the indicated power law over this range of reduced temperature, suggesting that hyperscaling may be violated in $^4$He-filled aerogels. Successive magnifications of the critical region indicate a possible crossover to a linear dependence on reduced temperature near the transition.

The fact that both the superfluid density and heat capacity of $^4$He-filled aerogels display distinctly different critical behavior from pure helium is strong evidence that the presence of long-range correlated disorder leads to a new universality class. In the case of $^4$He-filled aerogels, a precise characterization of the new universality class will require a theoretical understanding of the heat-capacity data presented here.

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