Superfluid Transition of ^{4}He in Porous Gold

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(Received 14 November 1996)

We have measured the superfluid density and the heat capacity of ^{4}He confined in porous gold in order to study the effect of quenched disorder on the superfluid transition. The superfluid density exponent is found to be identical to that of bulk ^{4}He within experimental resolution. The heat capacity shows a sharp peak at the transition temperature. The size of the peak appears to be larger than that predicted by critical phenomena theory. [S0031-9007(97)03417-0]

PACS numbers: 67.40.Kh, 64.60.–i, 67.40.Hf, 67.40.Yv

Theoretical analysis by Harris and others [1] showed that if the introduction of impurities into a near critical pure system corresponds to imposing random unfrustrated coupling in the system, then the phase transition of the impurity diluted system will remain sharp. Depending on whether the specific heat exponent α of the pure system is positive or negative, the critical exponents of the diluted system are predicted either to change (for $\alpha > 0$), or remain the same (for $\alpha < 0$, and such systems are said to satisfy the Harris criterion). In other words, for $\alpha < 0$, the diluted system resides in the same universality class as the pure system. More generally, it has been shown that a wide class of disordered systems should satisfy the inequality $\nu \geq 2/d$ (ν and d are the correlation length exponent and dimensionality of the system) [2]. This inequality is identical to $\alpha \leq 0$ assuming that hyperscaling ($2 - \alpha = d\nu$) holds. Examples of such systems include percolation, disordered magnets, and Anderson localization [1,2]. Experimental confirmation of the first half of the Harris prediction, for $\alpha > 0$, was found in a site diluted three dimensional Ising antiferromagnet [3]. Upon dilution with nonmagnetic ions, the positive α of the pure system changes to a value that is less than zero [3]. Since α is less than zero for the pure superfluid transition, confining ^{4}He in a porous medium allows for the testing of the second half of the Harris prediction. The superfluid density (ρ_s) of ^{4}He in Vycor [4,5] and aerogel [4,6] shows a critical-point-like simple power law dependence on the reduced temperature, $t = (T_c - T)/T_c$ (T_c is the transition temperature). In Vycor, the exponent between $t = 10^{-2}$ and 10^{-3} is indistinguishable from the bulk value within the experimental uncertainty, consistent with the theoretical prediction. The expected heat capacity peak at T_c, however, has been elusive [7,8]. The superfluid density exponent of ^{4}He in aerogel (also a porous glass), in contrast to ^{4}He in Vycor, was found to be distinctly larger than that of pure ^{4}He [4,6,9]. This unexpected result could possibly be related to the fact that the silica network in aerogel is correlated to long distances, which complicates the situation [9]. Thus, the experimental confirmation for the case of $\alpha < 0$ is far from being satisfactory.

In this Letter, we present the results of detailed experiments on ^{4}He in porous gold (PG) that are consistent with the prediction of Harris for $\alpha < 0$. We found a superfluid density exponent, ζ (equal to ν for the superfluid transition), identical to that of pure ^{4}He extending over two decades in t, and we also observed a heat capacity peak at T_c. Our results on the size of the peak, in conjunction with those of previous experiments [8,10], reveal an interesting puzzle pertaining to the hyperuniversality hypothesis [11].

PG samples are made by selectively leaching out silver from a silver-gold alloy with 70 at. % silver [12]. The scanning electron microscope (SEM) picture of PG (inset of Fig. 1) shows an interconnected porous structure with a single characteristic length scale, similar to that of

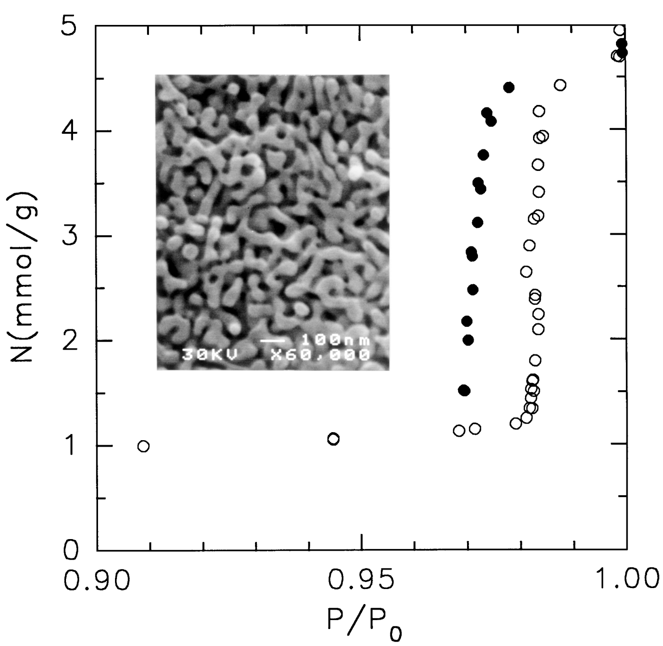

FIG. 1. Helium isotherm results on PGA at 2.18 K. The open symbols are for adsorption and filled symbols are for desorption. The inset is a SEM picture of PGA. The scale bar shown in the picture is 1000 Å.

porous Vycor glass [13]. A ^{4}He vapor pressure adsorption isotherm shows a sharp capillary condensation. Since the value of P/P_0 where a capillary condensation occurs is determined by the radius of curvature of the pores, a condensation at a well defined P/P_0 value, as shown in Fig. 1, indicates a porous structure with a very narrow distribution in pore diameter [14].

Two pieces of sample A (PGA), each 0.05 cm thick and 1 cm in diameter, one for ρ_s measurements and the other for heat capacity measurements, are cut from the same ingot. Silver in these two pieces is leached out in a 70% nitric acid solution. The porous structures of these two samples, as characterized by SEM pictures and ^{4}He adsorption isotherm (Fig. 1), are indistinguishable from each other. A methane vapor pressure isotherm at 77 K is made on the sample and the surface area and total pore volume are deduced. If a uniform cylindrical model is assumed for the pores, then the pore diameter is found to be 750 Å, consistent with that shown in the SEM picture. The porosity is found to be 69%. The porosity and pore diameter of sample B (PGB) are found to be 58% and 240 Å, respectively. Silver in PGB is leached out in a perchloric acid solution with an applied potential of 1.04 V. ρ_s and heat capacity measurements on PGB, which is 0.025 cm thick and 1 cm in diameter, are made sequentially on the same sample.

ρ_s is measured by the torsional oscillator technique [4]. The sample is encapsulated in a thin magnesium shell and then glued onto a hollow beryllium copper torsion rod 1.4 cm long and 0.89 mm in diameter, which also serves as a fill line. For PGA, the torsional oscillator has a resonant frequency of about 578 Hz with a mechanical Q of 7×10^5 at low temperature. We were able to resolve a mass loading or superfluid decoupling within 1×10^{-6} g/cm^3. The superfluid density of ^{4}He in PGA very close to T_c is shown in Fig. 2. A small quantity of bulk ^{4}He inside the cell provides a convenient fixed point in the temperature scale (bottom panel of Fig. 2 and Fig. 3).

The ρ_s data of PGA are analyzed in the form

$$\rho_s(t) = \rho_{s0} t^{\zeta}(1 + bt^{\Delta}). \qquad (1)$$

The correction to scaling exponent Δ is fixed at 0.5 [15]. The other parameters are determined by a nonlinear least squares fitting procedure. These parameters are found to be $\rho_{s0} = 0.14 \pm 0.01$ g/cm^3, $\zeta = 0.67 \pm 0.01$, $b = 1.6 \pm 0.5$, and $T_c = 2.1691 \pm 5 \times 10^{-5}$ K. The superfluid density, throughout this Letter, is expressed in grams per unit coarse grain averaged volume of the entire experimental system, i.e., the volume includes both the porous gold structure and liquid helium contained therein. As noted above, the temperature is determined against the bulk transition temperature (T_λ is found at 2.171 K because the measurements are made under pressure of 0.12 bar). The uncertainties quoted are standard deviations. In order to exclude data that may be affected by possible fourth sound resonances near T_c and/or rounding related to inhomogeneity in the porous structure, only the data in the range $2 \times 10^{-4} < t < 10^{-2}$ are included in the analysis. If only the data in the range $2 \times 10^{-4} < t < 2 \times 10^{-3}$ are included in a simple power law analysis (without the correction to scaling term), the parameters are found to be $\rho_{s0} = 0.15 \pm 0.01$ g/cm^3, $\zeta = 0.67 \pm 0.01$, and $T_c = 2.1691 \pm 5 \times 10^{-5}$ K.

Similar procedures are adopted to analyze ρ_s data of PGB. The scatter in the data is larger than that of PGA due to smaller signal and lower mechanical Q (1×10^5) of the oscillator. Deviation from simple power law behavior is noticeable for the data inside $t = 10^{-3}$ (Fig. 4). As a consequence, only data in the range $10^{-3} < t < 10^{-2}$ are included in the analysis. The parameters in Eq. (1) for PGB are found to be $\rho_{s0} = 0.07 \pm 0.01$ g/cm^3, $\zeta = 0.68 \pm 0.02$, $b = 0.7 \pm 0.5$, and $T_c = 2.156 \pm 5 \times 10^{-4}$ K. In Fig. 4, ρ_s of PGA and PGB are plotted against the reduced

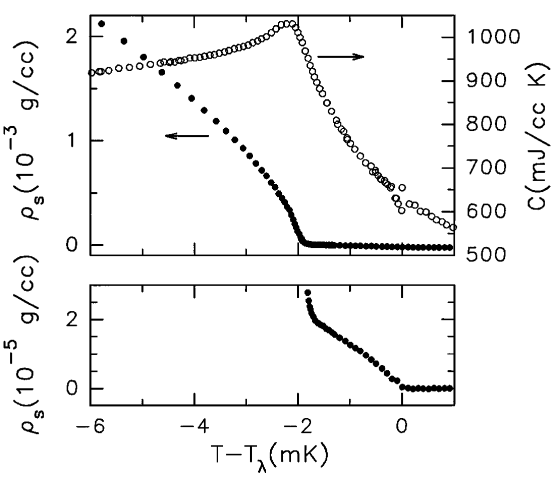

FIG. 2. The superfluid density (filled circles) and the heat capacity (open circles) of ^{4}He in PGA. The bottom panel shows, at a highly expanded scale, the superfluid signal of a small quantity of bulk ^{4}He inside the cell. This signal provides a convenient temperature fixed point.

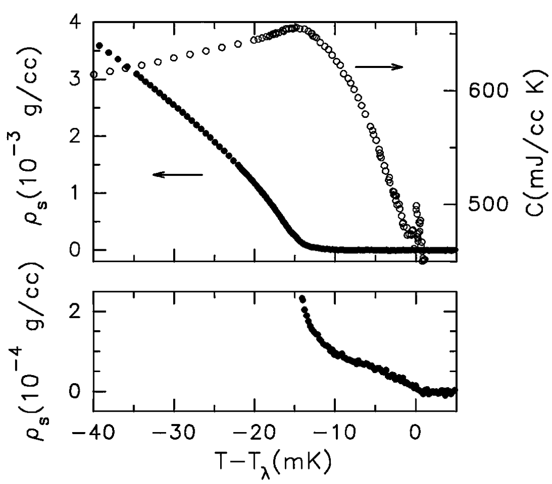

FIG. 3. The superfluid density (filled circles) and the heat capacity (open circles) of ^{4}He in PGB. The bottom panel shows at expanded scale the superfluid signal of a small quantity of bulk ^{4}He present inside the cell.

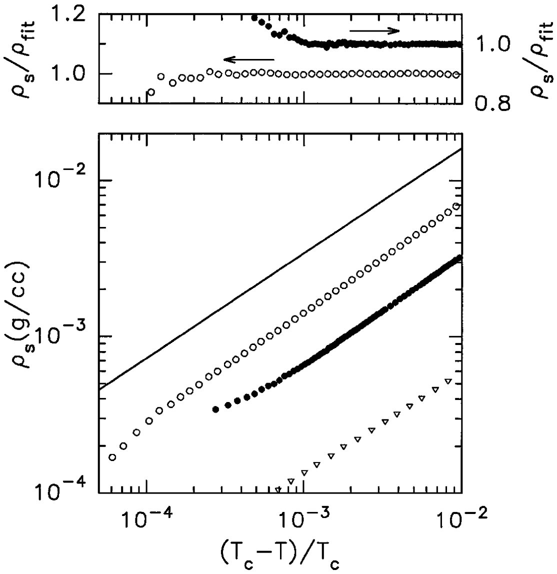

FIG. 4. Log-log plots for the superfluid density in PGA (open circles), PGB (filled circles), and Vycor (triangles, from Ref. [3]) as a function of reduced temperature. The solid line represents the fit of bulk helium data (Ref. [12]). The top panel shows fractional deviations of the superfluid density data of PGA (open circles) and PGB (filled circles) from the power law fit [Eq. (1)].

temperature in a log-log scale. For comparison, ρ_s of bulk ^{4}He [16] and ^{4}He in Vycor [5] are also shown. The fact that the ρ_s data are parallel to each other is a visual confirmation of the results of the least squares fit that all the systems have the same critical exponent. In the top panel of Fig. 4, the fractional deviation of the data from the best fit [Eq. (1)] is shown. It shows deviation from the power law behavior inside $t = 10^{-4}$ for PGA and $t = 10^{-3}$ for PGB. A likely reason for the deviation, as noted above, is inhomogeneity in the porous structure of the PG samples. If we assume Josephson's relation [17],

$$\xi(t) = \xi_0 t^{-\zeta} = \frac{k_B T_c m^2}{\hbar^2 \rho_s(t)}, \qquad (2)$$

to be valid for ^{4}He in porous gold, then the amplitude of the correlation length is found to be $\xi_0 = 0.84$ nm for PGA and $\xi_0 = 1.7$ nm for PGB. In Eq. (2), k_B is Boltzmann's constant, m the mass of the ^{4}He atom, and $\hbar$ Planck's constant. Comparing with Fig. 4, these values of ξ_0 imply that macroscopic inhomogeneity at length scales larger than 400 nm (170 nm) is present in PGA (PGB).

Figures 2 and 3 also show heat capacity of ^{4}He in PGA and PGB measured by the ac technique. In order to eliminate draining of the applied heat through the superfluid film, a Vycor glass superleak (which closes for $T > 1.95$ K, T_c of ^{4}He in Vycor) is used to link the filling capillary to the sample cell. A careful configuration of thermal reservoirs provides a temperature stability of 100 nK for the calorimeter. A resolution in heat capacity of 1.5×10^{-3} is achieved. The presence of a small quantity of bulk liquid, as in the ρ_s measurements, provides a convenient temperature fixed point. The contribution due to bulk ^{4}He has been subtracted from the data shown in Figs. 2 and 3. The "scatter" near T_λ is due to an imperfect subtraction of the bulk signal.

The temperature at which the heat capacity peak is centered, as shown in Fig. 2, is found to be roughly 200 μK lower than the T_c of ρ_s results. The agreement in T_c may, in fact, be better. If the ideal heat capacity peak of PGA at T_c has a shape that resembles the λ peak, which has a much sharper drop off on the high temperature side, a shift of the apparent peak to lower temperature, in addition to a rounding of the peak, is expected when a finite ac heat is applied. Such a downward shift of the peak is also expected if there is a rounding due to inhomogeneity in the sample. The value of T_c, quoted above, on the other hand, is determined by the ρ_s data outside the rounded region assuming a power law behavior. The peak-to-peak temperature oscillation in the heat capacity measurements is tuned to be 150 μK near the peak so that it is less than the rounding observed in the ρ_s measurements, approximately 250 μK. The agreement of the heat capacity and ρ_s transition temperatures within 200 μK and the result on the ρ_s exponent indicate that the superfluid transition in porous gold is a genuine continuous phase transition residing in the same universality class as that of bulk ^{4}He.

In Fig. 5, the heat capacity of PGA [trace (*a*)] is compared with the heat capacity of ^{4}He confined in Nuclepore filter papers of pore diameter 800 Å [trace (f)] [18]. The pores in these filter papers are essentially one dimensional. Although the pore diameters of these two systems are comparable, the heat capacity of PGA shows a rounded "diverging" feature near the maximum, which is not present in the Nuclepore data. The maximum of the heat capacity of PGA is found at 2.1 mK below T_λ; in Nuclepore it is 1 mK below T_λ. The shape and the position of the heat capacity peak in Nuclepore are found to be consistent

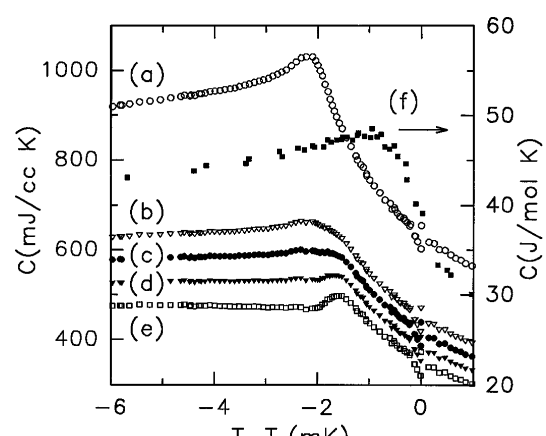

FIG. 5. Measured heat capacity of ^{4}He in PGA [trace (*a*)]. Traces (*b*), (*c*), (*d*), and (*e*) are the residuals when a Gaussian rounded λ peak positioned at T_c with a multiplying factor of 0.17, 0.2, 0.23, and 0.26 is subtracted from trace (*a*). Trace (*f*) is the heat capacity of ^{4}He confined in Nuclepore filter papers of pore diameter 800 Å (Ref. [14]).

with those expected from finite size rounding of an "ideal" superfluid transition at T_λ [19]. These differences again confirm that a genuine continuous transition at T_c, rather than a finite size rounding, is the appropriate framework to understand the results in PGA.

For different systems residing in the same universality class, the hyperuniversality hypothesis predicts a universal parameter R_ξ of the form [11]

$$R_\xi = \left(\frac{A}{k_B}\right)^{1/3} \xi_0 \,, \tag{3}$$

where A is the amplitude of the singular part of the heat capacity, and ξ_0 the amplitude of the correlation length. A constant value of $R_\xi = 0.84 \pm 0.01$ is obtained for the superfluid transition of ^{4}He along the λ line, as pressure is changed from 0.05 bar to 30 bars [10].

Because of the rounding of the peak, a direct determination of A in Eq. (3) for PGA is not possible. We make the assumption that the measured heat capacity consists of a singular part related to the criticality of the transition at T_c and a nonsingular part. We further assume that the singular part can be mimicked by rounding the bulk lambda peak [20] by a Gaussian function with a width of 250 μK and multiplying by a factor (smaller than 1). The width is consistent with the rounding observed in the ρ_s measurements. Traces (*b*), (*c*), (*d*), and (*e*) in Fig. 5 are the residuals when such a peak with a multiplying factor of 0.17, 0.2, 0.23, and 0.26 is subtracted from trace (*a*). If we make the assumption that the residual or the nonsingular part should be smooth resembling trace (*f*), then traces (*b*), (*c*), and (*d*) would suggest that the amplitude A in Eq. (3) for PGA is 20 $\pm$ 3% that of bulk. Using the value for ξ_0 determined from the ρ_s measurements, R_ξ is calculated to be 1.2 $\pm$ 0.1 for PGA. The same analysis of the heat capacity peak found at T_c of PGB gives $R_\xi = 1.1 \pm 0.1$. If we take 0.84 to be the correct value of R_ξ, then the size of the singular peak is about 3 times larger than that predicted by Eq. (3). In the experiment of bulk ^{4}He along the λ line, the value of ξ_0 changes from 0.345 to 0.310 nm or by 10% [10]. In contrast, the values of ξ_0 of ^{4}He in PGA and PGB are larger than that of bulk ^{4}He by factors of 2.5 and 5, respectively. Further experimental and theoretical work seems to be necessary to clarify this issue.

We thank A. Wong and D. Fisher for bringing porous gold to our attention, and K. Sieradzki and R. Li for introducing us to the fine art of porous gold making. We thank J. Reppy, M. Fisher, F. Gasparini, G. Zassenhaus, and J. Banavar for fruitful discussions, and F. Gasparini for providing us the Nuclepore data in a tabulated form. This work was supported by the national Science Foundation under Grants No. DMR-93311918 and No. DMR-9630736.

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