Superfluidity of ^{3}He in Aerogel Covered with a Thick ^{4}He Film

A. Golov, J. V. Porto,* and J. M. Parpia

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853
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We have examined the superfluidity of ^{3}He in 98.2% porous silica aerogel with up to 34% ^{4}He at 21.6 bars. The mixture is phase separated for ^{4}He fractions between ~11% and 34%, and the ^{4}He-rich phase preferentially occupies the regions of highest silica density in the aerogel, thus modifying the distribution of the correlated disorder seen by superfluid ^{3}He. The ^{3}He T_c increases slightly with ^{4}He content while the superfluid fraction decreases rapidly. This behavior is inconsistent with that of ^{3}He in a homogeneously scattering medium and is analogous to that of a granular superconductor. [S0031-9007(98)06127-4]

Liquid ^{3}He in highly porous aerogel provides a unique opportunity to study the influence of disorder on the triplet superfluid. In 98.2% porous aerogel, the ^{3}He superfluid transition is sharp and the transition temperature, T_c^{aero}, is reduced compared to that of the bulk, T_c^{bulk} [1–3], so that at 21.6 bars $T_c^{aero}/T_c^{bulk} \approx 0.77$ [4]. The superfluid fraction, ρ_s/ρ, is more sensitive to disorder and is reduced to $\rho_s^{aero}/\rho(T = 0) \approx 0.2$, compared to $\rho_s^{bulk}/\rho(T = 0) = 1$. Simultaneous measurements of ρ_s^{aero}/ρ and the NMR frequency shift have shown that the superfluid transition is coincident for both techniques, and therefore T_c^{aero} is homogeneous throughout the sample [5]. A further suppression of T_c^{aero} in a magnetic field is progressively eliminated by the substitution of surface ^{4}He for the localized ^{3}He atoms [6].

On a microscopic scale the base-catalyzed aerogel is a diffusively aggregated conglomerate of silica particles of ~50 Å size. The disorder, imposed by the aerogel on ^{3}He, can be characterized by a distribution of the correlations in silica [7]. For these aerogels of 98% porosity, the disorder spans the range up to ~1000 Å [8].

The broad distribution of length scales in aerogel introduces correlated disorder which is responsible for the qualitatively different behavior of the transition for superfluid ^{4}He [9]. However, in superfluid ^{3}He, the description is complicated because the internal length scale, the coherence length, ξ_0 (which varies with pressure from 800 to 150 Å), is encompassed by the distribution of silica correlations. It is not obvious which properties of ^{3}He are affected most strongly by pores smaller than ξ_0, and which ones are dominated by bigger pores [10]. Computer simulations of 98.2% porous base-catalyzed aerogels [7,8] reveal a mean free path ~2200 Å and that the largest distance from any point in the open volume to silica is 300 Å. Thus, all ^{3}He is within ~ξ_0 of silica.

In this experiment we set out to modify the distribution of the correlations of the disorder relative to ξ_0 in order to determine its role in suppression of the superfluid. This was accomplished by coating the aerogel with a thick ^{4}He film which preferentially fills the smallest pores and

thus raises the lower cutoff of the correlations of disorder sampled by the ^{3}He phase.

We monitored the period, $P \approx 1.14$ ms, of a torsional oscillator (TO) containing an aerogel sample. A concentric-plate capacitor in the head of the TO, inside which the aerogel was grown (inset of Fig. 1), allows us to determine the molar fraction of ^{4}He, x_4 [11]. The 98.2% porous base-catalyzed aerogel (0.4 cm^3 open volume and 9.3 m^2 area), sintered powder (0.4 cm^3 open volume and 4.2 m^2 area), and a 0.3 cm^3 bulk volume outside the cell constitute the experimental arrangement. The temperature was measured with a ^{3}He melting curve thermometer. Data were taken while ramping the temperature from 0.7 to 2.5 mK at ~8 μK/h.

As the ^{3}He-^{4}He sample can be prepared in different ways [11], the interface between the ^{3}He and ^{4}He phases can
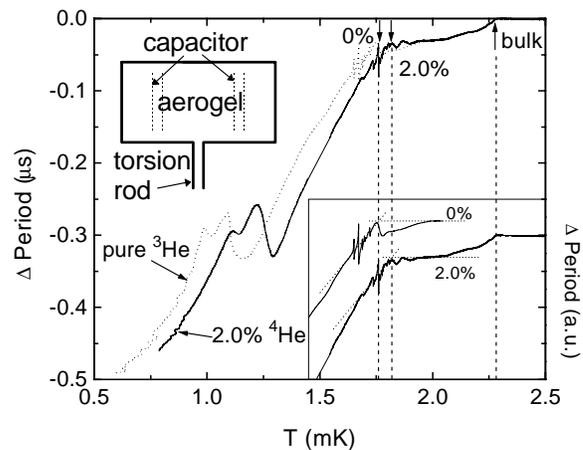
FIG. 1. TO period shifts for pure ^{3}He (dotted line) and for 2.0% ^{4}He (solid line) at 21.6 bar. Periods have been offset to be identical at T_c^{bulk}. Vertical dashed lines indicate (from left to right) T_c^{aero} for pure ^{3}He, T_c^{aero} for 2.0% ^{4}He, and T_c^{bulk}. The left inset shows a schematic of the TO, illustrating the capacitor. The lower inset highlights the region near T_c^{aero} showing how T_c^{aero} is determined from the intersection of dotted lines representing ΔP vs T in aerogel and the behavior in the bulk. The periods are offset for clarity; the temperature scale is identical to that of the main figure.

take on numerous metastable configurations. In this paper, with the exception of the 34% ^{4}He sample, we discuss only ^{3}He-^{4}He samples that are formed by slow cooling of a ^{3}He-^{4}He mixture through the phase separation transition. The 34% ^{4}He sample was prepared by sequential deposition of the ^{4}He and ^{3}He while the temperature of the aerogel was maintained below 100 mK.

We first surveyed the behavior of pure ^{3}He in aerogel as a function of pressure. The cell also contained a small ($\sim$150 μm thick) slab of bulk ^{3}He fluid between the aerogel and the mating cup of the TO. The T_c^{aero} and $\rho_s^{\text{aero}}/\rho$ as a function of pressure were similar to those of another sample [3]. In order to carry out the experiment to examine the effect of ^{4}He we restricted our experimental investigation to a pressure $p = 21.6$ bar. The period signal for pure ^{3}He is shown in Fig. 1.

The cell's construction (presence of the capacitor, large fill line together with an irregular bulk ^{3}He volume) results in numerous resonances that appear in Fig. 1. Other than making the selection of T_c difficult, they do not play a role in the discussion that follows. With the addition of more than 10% ^{4}He we find that these resonances are quenched and replaced with numerous small but reproducible resonances which can give the appearance of noise.

When 2.0% ^{4}He (enough to coat the entire aerogel surface with solid ^{4}He) was added [12], we observed that T_c^{aero} increased by $\sim$0.06 mK (Fig. 1). The shift in T_c^{aero} was determined from the onset of the decrease of the period (Fig. 1, inset). This increase is much smaller than the suppression of T_c^{aero} of pure ^{3}He to 1.76 mK from $T_c^{\text{bulk}} = 2.28$ mK. We also observed an increase of $\sim$20% in the magnitude of the period shift, ΔP, due to the decoupling of the superfluid ^{3}He. The elevation of T_c^{aero} by the addition of 2% ^{4}He is not directly comparable to that seen by Sprague $et\ al.$ [6] at $H = 1.5$ kOe. They found the suppression of T_c^{aero} due to magnetic scattering to be proportional to H^2 and would have predicted no elevation of T_c^{aero} for ^{4}He added at $H = 0$.

The ^{4}He molar fraction was not identical in the aerogel, sinter, and dead bulk volumes because of differences in their physical environments [11]. We found that samples with x_4 in aerogel between 2% and 11% were not stable, and that the ^{4}He was expelled in favor of ^{3}He (leaving only the $\sim$2% ^{4}He strongly attracted to the silica). Similarly, $x_4 \geq 40\%$ was unstable in the aerogel, and the ^{3}He phase was replaced by the ^{4}He-rich phase.

We therefore examined samples with 13%, 21%, and 34% ^{4}He content. Following the addition of ^{4}He, we observed a further increase in T_c^{aero} from 1.82 to 1.90 mK (Fig. 2) while in contrast to the 2% ^{4}He results, ΔP decreased substantially making the identification of T_c^{aero} progressively more difficult. Fortunately, with the addition of ^{4}He, the strong resonant structure (Fig. 1) is quenched, and we were able to use the sharp onset of dissipation (inset of Fig. 2) to fix T_c^{aero}.
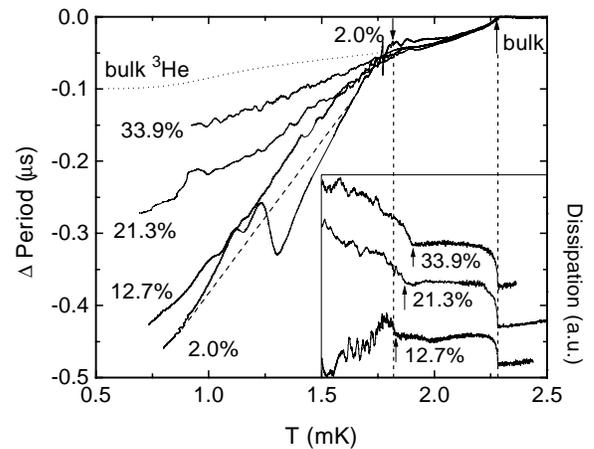
FIG. 2. TO period shifts for four different ^{4}He amounts at 21.6 bar. The dotted line shows the contribution from the slab of bulk ^{3}He. The dashed line for the 2% ^{4}He curve serves to guide the eye through the resonance. Inset: the TO dissipation signal for the 12.7%, 21.3%, and 33.9% ^{4}He samples showing the sharpness of the transition and how T_c^{aero} was chosen (arrows). Vertical dashed lines indicate T_c^{aero} for 2.0% ^{4}He (left) and T_c^{bulk} (right). The temperature scale is identical to that of the main figure.

In the following analysis we consider the ^{3}He-^{4}He mixture to be phase separated at $T < 3$ mK [11,14] and neglect the $\approx$14% molar volume difference between liquid ^{3}He and ^{4}He. The superfluid component, $\rho_s^{\text{aero}}(T)$, is related to the period of the TO, $P(T)$, by the relation

$$\frac{P(T_c) - P(T)}{P_0 - P_{\text{empty}}} = \frac{(1 - x_4)}{\alpha} \frac{\rho_s^{\text{aero}}(T)}{\rho}. \qquad (1)$$

Here $P_0 - P_{\text{empty}}$ is the total period shift due to filling the cell with pure ^{3}He at this pressure, the $(1 - x_4)$ term accounts for the replacement of ^{3}He by ^{4}He, and α is a measure of the tortuosity of the superflow through the porous medium. For an ideal cylindrical flow, $\alpha = 1$, and for a very tortuous one, $\alpha \rightarrow \infty$. For α to be appropriately used, it has to describe the tortuous streamlines of a homogeneous superfluid. Consequently, the scale of the structure of the porous medium has to be much greater than the characteristic length scale of superfluid ^{3}He, ξ_0. All smaller scale disorder would then be manifested as intrinsic to the properties of the superfluid in aerogel, and lead to a "coarse-grained" superfluid density [15]. At $p = 21.6$ bar $\xi_0 \approx 200$ Å, so one can only consider tortuosity as arising from structure ≥ 1000 Å in scale. Our aerogel appears to be homogeneous on these lengths, and the TO was cylindrically symmetrical, so $\alpha = 1$. The addition of ^{4}He progressively removes the densest regions of aerogel from contact with superfluid ^{3}He [16]. The ^{4}He-rich component (film) follows the structure of the aerogel, hence it cannot introduce new correlations with length scale larger than that of aerogel. Consequently, the sample remains homogeneous on length scales of ≥ 1000 Å, and we keep $\alpha = 1$.

From the knowledge of x_4 and α we can calculate the superfluid fraction with ^{4}He present in the cell, once the contribution of the bulk ^{3}He sample is subtracted off. We used the known hydrodynamic behavior of bulk ^{3}He in a slab [17] (dotted line in Fig. 2) to fit the period shift data in the region between T_c^{aero} and T_c^{bulk}. The curves for $\rho_s^{\mathrm{aero}}/\rho$ vs temperature are shown in Fig. 3. Our results are summarized in Fig. 4 where we show $\rho_s^{\mathrm{aero}}/\rho$ at $0.5T_c^{\mathrm{aero}}$ and T_c^{aero} for various x_4 in the cell.

Two different regimes characterize the ^{3}He-^{4}He interface as the ^{4}He amount is increased. In the first ["van der Waals regime" (VDW)], a thin VDW ^{4}He film coexists with ^{4}He filled pores as a consequence of the interplay between the VDW adsorption and capillary condensation [18]. Upon adding ^{4}He, successively larger pores are filled with the ^{4}He-rich phase, but the thin VDW film is still present. In the second ("capillary-condensed regime"), there is only a thick ^{4}He-rich film. Its capillary condenses around the regions rich in silica, leaving the ^{3}He phase in the center of the open regions devoid of silica (biggest pores), thus altering the structure of the medium sampled by the ^{3}He superfluid. Finally, at some $x_4 = x_c$, the regions of ^{3}He get completely encapsulated by the ^{4}He-rich phase and thus isolated into islands. The particular value of x_c depends on the structure of the big voids. For example, for closely packed identical spherical voids, $x_c = 1 - \pi/3\sqrt{2} = 26\%$.

The measured T_c^{aero} and $\rho_s^{\mathrm{aero}}/\rho$ for the sample with 13% ^{4}He do not differ much from that with 2% ^{4}He which is consistent with the system being in the van der Waals regime. By adding 2% and 13% of ^{4}He to ^{3}He in 98% porous aerogel, we decrease its effective porosity to about 96% and 85%, respectively. The ^{4}He occupies the roughest and densest regions of silica leaving the interconnected biggest pores virtually intact. This picture also fits in with the observation that superfluidity of pure ^{3}He is entirely suppressed in 95% open aerogel
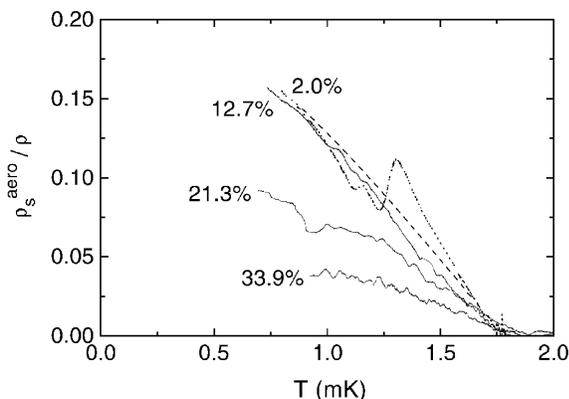
[19]. In 95% porous silica, the size of the biggest pores will decrease compared to those in 98% samples. Both results argue that T_c^{aero} and $\rho_s^{\mathrm{aero}}/\rho$ reflect the presence of interconnected voids in aerogel.

In the "capillary condensed region," the connectivity of different parts of ^{3}He starts to be affected by ^{4}He encapsulation; hence, the superflow can be suppressed even though the local Cooper pairing is still as strong as it was for pure ^{3}He. This picture is consistent with our observation that T_c is not suppressed further by the addition of ^{4}He, but that the ρ_s^{aero} is (21% and 34% samples). The behavior looks similar to the superconductivity of disordered metal films where, depending on preparation, the films could be either *uniformly disordered* or *granular* [20]. In the granular film, the phase coherence between the grains can be destroyed (while the local T_c and the order parameter in the grains are unaffected). The decrease in ρ_s^{aero} with the addition of ^{4}He could be attributed to weakened phase coherence between the more open regions. In this sense, superfluid ^{3}He in aerogel with 30% ^{4}He looks similar to a granular superconductor.

A somewhat similar behavior (relatively robust T_c, strong suppression of ρ_s) has also been seen in high-temperature superconductors with defects. The phenomenon is attributed to the inhomogeneity of scattering [21], and may also play a part in the ^{3}He-aerogel system.

Immediately after the discovery of superfluidity in aerogel the homogeneous scattering model [2,22] that treats aerogel as a collection of small uncorrelated impurities was proposed and showed qualitative success in describing the $T_c^{\mathrm{aero}}(\xi_0)$ [1,2,6]. In this theory, the suppression of both ρ_s^{aero} and T_c^{aero} is determined by the ratio of the coherence length to the quasiparticle mean free path. In our experiment, with the addition of ^{4}He, the mean free path should decrease because of the increased cross section of the ^{4}He-covered silica, further suppressing both ρ_s^{aero} and T_c^{aero}. Thus, the observed enhancement of T_c^{aero} accompanied by the reduction of ρ_s^{aero} is inconsistent with this model. By design, this theory characterizes all the disorder to be on

FIG. 3. $\rho_s^{\mathrm{aero}}/\rho$ obtained from the data in Fig. 2 using Eq. (1) after the bulk ^{3}He contribution (dotted line in Fig. 2) is subtracted off. The dashed line guides the eye through the resonance in the 2.0% ^{4}He data (dotted line).
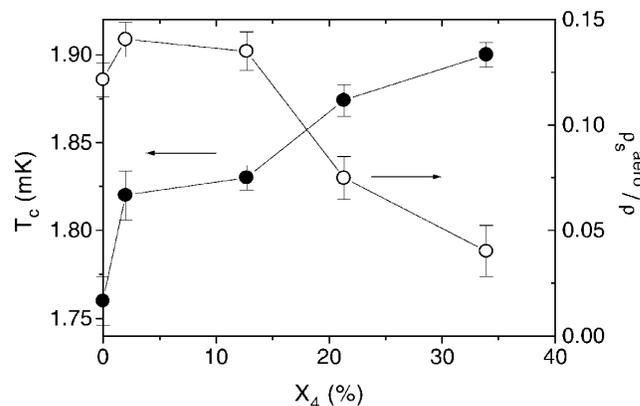
FIG. 4. T_c^{aero} ($\bullet$, left axis) and $\rho_s^{\mathrm{aero}}/\rho$ at $T = 0.5T_c^{\mathrm{aero}}$ ($\circ$, right axis) vs x_4. The lines connect the data points.

a scale much smaller than ξ_0, resulting in a superfluidity that is uniformly suppressed everywhere. A more realistic modification has appeared recently [22], which models the long range inhomogeneity of aerogel as a collection of periodically distributed spherical voids. Because of the proximity effect, it produces a single T_c^{aero} for the sample as a whole. However, the weight of the strongly scattering regions in the determination of T_c^{aero} is exponentially small compared to that of weak scattering regions. This result is very much in agreement with our finding that T_c^{aero} did not decrease (and even slightly increased) after the strongly scattering regions are filled with ^{4}He.

Confinement of ^{3}He by surfaces suppresses the T_c [23–25]. The addition of even a thin layer of superfluid ^{4}He increases the T_c [24], presumably due to change of the scattering from diffusive to specular which restores the component of the order parameter parallel to the surface. It is reasonable to assume that for a ^{4}He film to restore the T_c, its surface has to be flat on the scale of ξ_0. In aerogel, only at the highest ^{4}He amounts (21% and 34% samples) is the ^{3}He-^{4}He interface expected to be smooth on the scale of $\xi_0 = 200$ Å. Thus, the small increase in T_c^{aero}, observed at highest x_4, could also be a consequence of the changed boundary conditions around the strongly scattering regions.

In summary, we have examined the superfluid density, transition temperature, and ^{4}He content of ^{3}He-^{4}He mixtures in 98.2% open aerogel using a novel torsion-oscillator and capacitance sensor cell. The results show that T_c is relatively insensitive to the ^{4}He content, while ρ_s is suppressed very strongly by the addition of ^{4}He. Combined with the earlier observation of a homogeneous onset of superfluidity of ^{3}He in 98% aerogel, and no superfluidity of ^{3}He in 95% aerogel, it implies that the most open regions in the aerogel with long range correlations are responsible for the T_c. In addition, modification of the short range correlated regions by the presence of ^{4}He affects the long range order in a manner similar to that of granular superconductors.

*Present address: Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139.

[1] J. V. Porto and J. M. Parpia, Phys. Rev. Lett. **74**, 4667 (1995); J. V. Porto and J. M. Parpia, Czech J. Phys. **46**, 2981 (1996), Suppl. S6.

[2] D. T. Sprague *et al.,* Phys. Rev. Lett. **75**, 661 (1995).

[3] K. Matsumoto *et al.,* Phys. Rev. Lett. **79**, 253 (1997); **79**, 2922(E) (1997).

[4] Depending on the growth conditions, different aerogel samples of the same porosity can give slightly different values of T_c^{aero} and ρ_s^{aero} [3].

[5] H. Alles *et al.,* Physica (to be published).

[6] D. T. Sprague *et al.,* Phys. Rev. Lett. **77**, 4568 (1996).

[7] A. Hasmy *et al.,* Phys. Rev. B **50**, 6006 (1994).

[8] J. V. Porto, Ph.D. thesis, Cornell University, 1994 (unpublished); J. V. Porto *et al.* (to be published).

[9] M. H. W. Chan *et al.,* Phys. Rev. Lett. **61**, 1950 (1988).

[10] We use the term *pore* even though the conventional concept of pores inadequately describes this dilute material in which the voids are bounded by a sparse collection of strands instead of well defined walls.

[11] A. Golov, J. V. Porto, and J. M. Parpia, J. Low Temp. Phys. **110**, 591 (1998).

[12] The surface solid has a density of ≈ 35 μmol/m^2 on aerogel [13]. For ^{3}He at 21.6 bars in our aerogel sample this corresponds to about 2.2% of the helium atoms.

[13] P. A. Crowell *et al.,* Phys. Rev. B **51**, 12 721 (1995).

[14] S. B. Kim, J. Ma, and M. H. W. Chan, Phys. Rev. Lett. **71**, 2268 (1993); N. Mulders and M. H. W. Chan, Phys. Rev. Lett. **75**, 3705 (1995).

[15] For a related discussion, see Ref. [13], p. 12 729.

[16] Our measurements of the tortuosity of the ^{4}He component at $p = 21.6$ bars [11] reveal that between $x_4 = 2\%$ and 20%–30% it drops from $\alpha_4 \geq 5$ to $\alpha_4 \approx 2$. At $x_4 = 100\%$, $\alpha_4 = 1.11$. The results for thin ^{4}He films and ^{4}He in full pores in 91% porous aerogel, $\alpha_{\text{film}} = 10$ and $\alpha_{\text{full}} = 1.19$, are similar [13]. Thus, for small x_4, the ^{4}He-rich phase forms a filmlike state which follows the tortuous interconnected silica strands. As x_4 increases to 20%–30%, the connectivity of the ^{4}He-rich phase improves as it capillary condenses around neighboring strands.

[17] J. M. Parpia *et al.,* J. Low Temp. Phys. **61**, 337 (1985).

[18] P. G. de Gennes, in *Physics of Disordered Materials,* edited by D. Adler, H. Fritzsche, and S. R. Ovshinsky (Plenum, New York, 1985).

[19] T. Hall and J. M. Parpia, Bull. Am. Phys. Soc. **34**, 1197 (1989).

[20] R. C. Dynes, R. P. Barber, Jr., and F. Sharifi, in *Ordering Disorder: Prospect and Retrospect in Condensed Matter Physics,* edited by V. Srivastava, A. K. Bhatnagar, and D. G. Naugle (AIP, New York, 1993).

[21] M. Franz *et al.,* Phys. Rev. B **56**, 7882 (1997), and references therein.

[22] E. V. Thuneberg *et al.,* Phys. Rev. Lett. **80**, 2861 (1998).

[23] K. Ichikawa *et al.,* Phys. Rev. Lett. **58**, 1949 (1987).

[24] M. R. Freeman and R. C. Richardson, Phys. Rev. B **41**, 11 011 (1990); S. C. Steel *et al.,* Physica (Amsterdam) **165-166B**, 599 (1990); S. M. Tholen and J. M. Parpia, Phys. Rev. Lett. **68**, 2810 (1992).

[25] L. H. Kjaldman, J. Kurkijarvi, and D. Rainer, J. Low Temp. Phys. **33**, 577 (1978).