

Disorder and the Superfluid Transition in Liquid ^4He

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(Received 5 May 1988)

The influence of quenched disorder on the critical behavior of superfluid ^4He is studied by confinement of the helium to the pores of three different porous media: Vycor, xerogel, and aerogel glasses. In each case a well defined power-law behavior for the superfluid density is observed as the transition temperature is approached. In Vycor the superfluid critical exponent is bulklike, whereas strikingly different critical exponents are seen in the other two media.

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

The influence of quenched disorder on the critical behavior of a pure system is a topic of considerably experimental¹ and theoretical²⁻⁵ interest. When disorder is introduced into a uniform system a variety of changes in the critical behavior is possible. Perhaps the most common consequence of disorder is a smearing of the critical behavior so that the evidence of a sharp transition is destroyed. Such a broadening of the transition is often produced by concentration gradients, strains, or macroscopic inhomogeneities. In other cases the disorder may be irrelevant, in the sense discussed by Harris² and others,³⁻⁵ leaving the critical behavior of the disordered system identical to that of the uniform parent system. Finally, the introduction of disorder may leave the transition sharp but result in an entirely different critical behavior characterized by a new set of critical exponents.

In the work reported here, we have examined the influence of disorder on the critical behavior of a 3D system, liquid ^4He , at its superfluid transition. In our experiments the disorder is introduced into the pure system through displacement of the fluid by the solid structure of a porous medium. The helium must accommodate to the random potential that the porous medium provides. In earlier work with porous Vycor glass,⁶ the critical behavior, as indicated by the superfluid-density exponent, was found to be similar to that of the 3D pure system.⁷ In marked contrast, we have now discovered that for certain other classes of disordered porous structures there exist sharp phase transitions with well-defined critical exponents which are, nevertheless, different from that of the pure system. This new behavior should have strong implications for theories of critical phenomena in systems with quenched disorder. A preliminary report on a portion of this work has been presented.⁸

As the interest in and understanding of various porous materials has increased in recent years,⁹⁻¹¹ so has the opportunity for the investigation of the physics of systems constrained to lie within the pores. For our study, three different porous media were employed. These media have in common highly interconnected 3D microscopic porous structures with length scales lying in the 1-

to 100-nm range. The first of these materials is porous Vycor glass. The porous structure of Vycor is formed by leaching out one component of a spinodally decomposed glass.¹² In the process of spinodal decomposition a single Fourier component of the concentration fluctuations shows dominant growth. The pores of commercially available Vycor are less than 10 nm in diameter and form a highly interconnected microporous structure with an open volume fraction of approximately 30%.

The two other porous media employed, aerogel¹⁰ and xerogel¹³ glasses, were produced through a silica sol-gel process. The formation of gels proceeds by the diffusive aggregation of polymeric structures leading to a highly ramified structure.¹⁴ Porous xerogel glass is obtained by air drying and heat treating the gel.¹³ This process results in some collapse and reorganization of the gel structure. Our sample of the resulting glass has an open volume fraction of 60% and the pore size is restricted to a range near 10 nm. Aerogel is prepared through a hypercritical drying process.¹⁵ This technique allows the more delicate structures of the gel to be preserved and results in a low density material with an open volume fraction of 94% in the case of the samples used here. In contrast to Vycor and xerogel the pores of aerogel span a wide range in size.¹⁰

We have examined the temperature dependence of the superfluid fraction for helium contained in these three porous media. The method employed in our superfluid measurements is an adaptation of the torsional-pendulum technique originally introduced by Andronikashvili¹⁶ for the determination of the superfluid density of bulk liquid helium. In our case the porous material together with the liquid helium filling the pores form the moment of inertia of a torsional pendulum. In the present experiments, we have used lower oscillator frequencies and smaller experimental cells than in the past.⁷ These two changes substantially reduced the influence of spurious sound resonances, which tend to mar the quality of critical-exponent determinations.

In Fig. 1, we show the temperature dependence of the superfluid density for helium filling the pores of the ma-

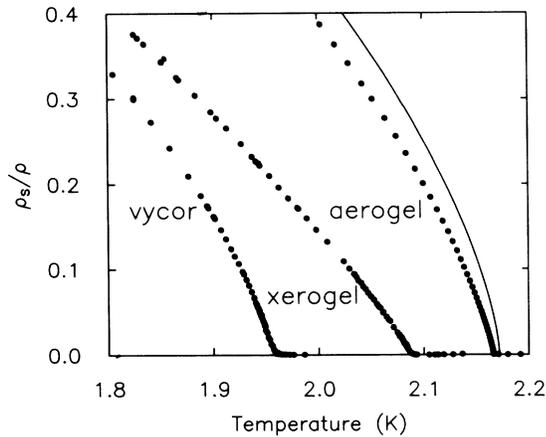


FIG. 1. The normalized superfluid-density data for helium contained in the three porous media are shown as a function of temperature. The solid curve gives the temperature dependence for the superfluid density of bulk helium.

materials. The superfluid-density data have been normalized to unity at zero temperature. Although all of these data sets show sharp transitions on the scale of this figure, they are in fact rounded near 10^{-2} (Vycor), 10^{-3} (xerogel), and 10^{-4} (aerogel) of T_c . For each sample, T_c is depressed below the value for bulk helium and the transition temperatures decrease with decreasing open volume fraction, perhaps suggestive of an impurity effect.

The data displayed in Fig. 1 can be fitted with a power law of the form $\rho_s = \rho_{s0} t^\zeta$, where $t = 1 - T/T_c$, and ζ is the superfluid-density exponent. In order to emphasize this power-law dependence, we display in Figs. 2 and 3 log-log plots of the normalized superfluid data as a function of reduced temperature. As has been found earlier,⁷ the Vycor data follow a well-defined power law for over a decade in reduced temperature. The superfluid-density exponent obtained from a fit to these data is $\zeta = 0.67 \pm 0.03$, a value identical within experimental uncertainty to the value, $\zeta = 0.674 \pm 0.001$, found by Greywall and Ahlers¹⁷ for the pure system. In striking contrast to this bulklike behavior of the ^4He -Vycor system, the superfluid-density exponent found for the xerogel case is 0.89 ± 0.02 , and 0.813 ± 0.009 for the aerogel. Both of these values are clearly different from the Vycor exponent as well as the bulk value. In Table I we summarize the pertinent numerical results.

The critical behavior of all but the thinnest films adsorbed on Vycor and xerogel substrates was found^{7,8} to be similar to that of their full-pore counterparts with nearly identical values for the superfluid-density exponents. The local confinement of the helium to thin films with thickness far smaller than a pore diameter seems to have no effect on the critical behavior other than a shift in the transition temperature. These earlier data were obtained with higher-frequency oscillators;

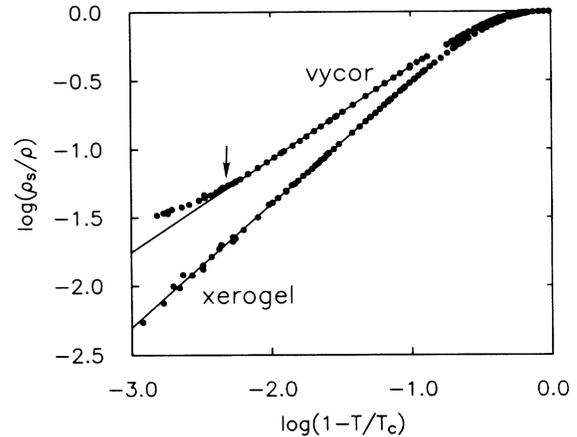


FIG. 2. The superfluid-density data for Vycor and xerogel are shown as a function of reduced temperature. The straight lines indicate power-law fits to the data. The arrow marks the beginning of the region of rounding in the Vycor data.

consequently, the data suffer to some extent from contamination by third-sound resonances.

On one run,⁸ the xerogel was preplated with two molecular layers of nitrogen and then filled with liquid helium. As with the films, there was no discernible change in the critical exponent. These two results make it unlikely that roughness of the surface or changes in pore diameter are playing any significant role in determining the critical behavior. Thus, our experiments suggest that it is the larger-scale morphology that is of paramount importance in controlling the critical phenomena.

The well-defined power-law behavior of the superfluid density near the transition temperature in each of these cases is clear evidence that the system undergoes a genuine phase transition at a suppressed critical temperature. Following Hohenberg *et al.*,¹⁸ the divergent superfluid correlation length near T_c is taken as

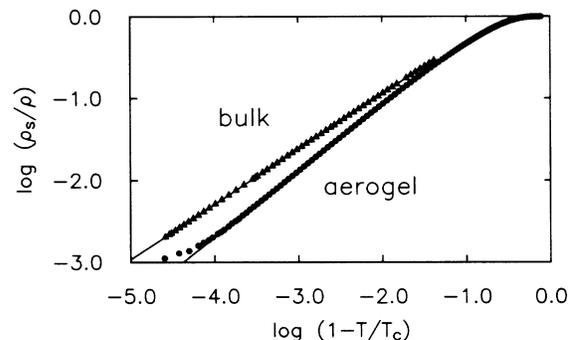


FIG. 3. The superfluid-density data for aerogel are shown as a function of reduced temperature. The straight line represents a power-law fit to the aerogel data. Bulk helium data (Ref. 17) are shown as triangles.

TABLE I. Experimental parameters for each of the systems discussed in the text. Values are listed for the approximate mean oscillator resonant frequency, the superfluid transition temperature, the superfluid-density exponent, the coefficient of the power-law fit, and the range spanned by the superfluid correlation length, $\xi(t)$, for the power-law region.

System	Frequency (Hz)	T_c (K)	ζ	ρ_{s0} (10^{12} g/cm ³)	Range of $\xi(t) = \xi_0 t^{-\zeta}$ (nm)
Vycor (full pore)	237	1.952	0.67 ± 0.03	1.03	$25 \leq \xi(t) \leq 170$
Vycor (film) ^a	1200	0.076	0.63 ± 0.03	0.02	$85 \leq \xi(t) \leq 360$
Xerogel (full pore)	203	2.088	0.89 ± 0.02	7.48	$12 \leq \xi(t) \leq 1100$
Xerogel (N ₂ plated) ^b	203	2.047	0.91 ± 0.05	4.79	$20 \leq \xi(t) \leq 5600$
Xerogel (film) ^b	1130	1.031	0.84 ± 0.04	1.22	$33 \leq \xi(t) \leq 590$
Aerogel (full pore)	204	2.167	0.813 ± 0.009	40.7	$12 \leq \xi(t) \leq 300$
Bulk ⁴ He ^c	n.a.	2.172	0.674 ± 0.001	35.0	$8 \leq \xi(t) \leq 480$

^aReference 7.

^bReference 8.

^cReference 17.

$\xi(t) = m^2 k_B T_c / \hbar^2 \rho_{s0} t^\zeta$, where the values of T_c , ζ , and ρ_{s0} are obtained from fits to the coarse-grain-averaged superfluid density¹⁹ measured in our experiments. The range of values listed in Table I show that in the critical region $\xi(t)$ can be much larger than the typical pore size. Thus, attempts to understand the critical phenomena of these systems will have to consider the nature of the porous media on length scales much larger than the typical pore diameter.

Vycor and the sol-gel glasses are manufactured by fundamentally different processes and these differences are preserved in the final structures. Independent evidence that these differences persist up to long lengths can be found in x-ray and neutron-scattering experiments which measure correlations in density fluctuations in the materials.^{20,21} For Vycor a prominent peak in the scattering intensity, believed to reflect the dominant wavelength involved in the spinodal decomposition, is seen centered near 25 nm.²⁰ At longer lengths the scattered intensity decreases by an order of magnitude. In contrast, the scattered intensity from gels²¹ generally increases monotonically as the probed length scale increases. These scattering curves often exhibit power-law regimes indicating that the structures are self-similar for lengths up to tens of nanometers. Unfortunately, not all of the scattering results extend up to the longest correlation lengths in our power-law regimes. Further experiments would be highly desirable.

Existing theories on the effects of quenched disorder on critical behavior seem inadequate to successfully treat the present experiments. Theories of short-range correlated disorder predict that the disorder is irrelevant if the specific-heat exponent is negative for the pure system.^{2,3} For bulk superfluid helium the observed exponent is slightly negative,²² $\alpha = -0.0127 \pm 0.0026$; therefore, these theories would predict no change in the critical exponent. Even if this is the proper explanation of the Vycor data, it fails to explain the new critical exponents found for the sol-gel glasses.

The scattering data indicate that gels are characterized by particularly long-range density correlations in contrast to Vycor, where the correlations fall off rapidly above the scattering peak. Weinrib and Halperin⁴ have developed a theory for long-range correlated disorder in which the disorder is characterized by variations in a local transition temperature. The critical exponents will be altered if the correlations in local T_c fluctuations decay sufficiently slow. However, this theory is expected to be valid only when the reduced temperature is smaller than the reduced T_c shift—a condition which is not generally true within our power-law regimes. Moreover, we are not certain that the local T_c shifts are only a function of the local impurity densities and therefore we do not know if we can relate the measured density correlations to the theoretical T_c correlations. Since the bulk-helium heat-capacity exponent is so close to zero, one might also wish to consider the possibility that a large part of the critical region is dominated by some disorder fixed point. None of our gel results, however, show evidence of an eventual return to bulklike behavior.

The experiments reported in this Letter demonstrate the profound influence of quenched disorder on the critical behavior of superfluid helium. In the case of the two silica gel glasses we find well-defined critical exponents for the superfluid density that are clearly different from that of the pure system. This strongly suggests the possibility of new universality classes for disordered helium. These results present a challenge to both theory and experiment to develop an adequate understanding of the role of disorder in modifying the critical behavior of simple systems such as superfluid helium.

We would like to thank Dr. David Awschalom and Dr. Merrill Shafer of IBM Research for providing the xerogel samples, Dr. G. Poelz of DESY for donating the aerogel samples, and Dr. David Bishop of AT&T Bell Laboratories for supplying the Vycor samples. One of us (M.H.W.C.) would like to thank the John Simon Guggenheim Memorial Foundation for financial support and

the Cornell Physics Department and Laboratory of Atomic and Solid State Physics for their hospitality. J.D.R. acknowledges the hospitality of the The Aspen Center for Physics. G.K.S.W. would like to acknowledge the support of Natural Sciences and Engineering Research Council. This work has been supported by the National Science Foundation under Grant No. NSF-DMR-8418605 and by the Cornell Materials Science Center through Grant No. NSF-DMR-82-17227A, MSC Report 6465.

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¹See, for example, R. A. Dunlap and A. M. Gottlieb, *Phys. Rev. B* **23**, 6106 (1981); R. J. Birgeneau, R. A. Cowley, G. Shirane, H. Yashizawa, D. P. Belander, A. R. King, and V. Jaccarino, *Phys. Rev. B* **27**, 6747 (1983); J. M. Hastings, L. M. Corliss, and W. Kunmann, *Phys. Rev. B* **31**, 2901 (1985).

²A. B. Harris, *J. Phys. C* **7**, 1671 (1974).

³T. C. Lubensky, *Phys. Rev. B* **11**, 3573 (1975); G. Grinstein and A. Luther, *Phys. Rev. B* **13**, 1329 (1976).

⁴A. Weinrib and B. I. Halperin, *Phys. Rev. B* **27**, 413 (1983).

⁵P. B. Weichman and M. E. Fisher, *Phys. Rev. B* **34**, 7652 (1986).

⁶Trademark of Corning.

⁷C. W. Kiewiet, H. E. Hall, and J. D. Reppy, *Phys. Rev. Lett.* **35**, 1286 (1975); D. J. Bishop, J. E. Berthold, J. M. Parpia, and J. D. Reppy, *Phys. Rev. B* **24**, 5047 (1981); B. C. Crooker, B. Hebral, E. N. Smith, Y. Takano, and J. D. Reppy, *Phys. Rev. Lett.* **51**, 666 (1983).

⁸K. I. Blum, S. Murphy, M. H. W. Chan, D. D. Awschalom, and J. D. Reppy, in *Proceedings of the Eighteenth International*

Conference on Low Temperature Physics, Kyoto, Japan, 1987 [*Jpn. J. Appl. Phys.* **26**, Suppl. 26-3, 275 (1987)].

⁹*Physics of Finely Divided Matter*, edited by N. Boccarda and M. Daoud, Springer Proceedings in Physics Vol. 5 (Springer-Verlag, Berlin, 1985).

¹⁰J. Fricke, *Sci. Am.* **258**, No. 5, 92 (1988); *Aerogels*, edited by J. Fricke (Springer-Verlag, Berlin, 1986).

¹¹*Physics and Chemistry of Porous Media*, edited by D. L. Johnson and P. N. Sen, AIP Conference Proceedings No. 107 (American Institute of Physics, New York, 1984).

¹²See, for example, J. W. Cahn, *J. Chem. Phys.* **42**, 93 (1965), and references therein; N. S. Andreev, G. G. Boiko, and N. A. Bokov, *J. Non-Cryst. Solids* **5**, 41 (1970).

¹³M. W. Shafer, D. D. Awschalom, J. Warnock, and G. Ruben, *J. Appl. Phys.* **61**, 5438 (1987).

¹⁴See, for example, A. Chhabra, D. Matthews-Morgan, D. P. Landau, and H. J. Hermann, *Phys. Rev. B* **34**, 4796 (1986); P. Meakin, *Phys. Rev. Lett.* **51**, 1119 (1983); M. Kolb, R. Botet, and R. Jullien, *Phys. Rev. Lett.* **51**, 1123 (1983).

¹⁵R. A. Laudise and D. W. Johnson, Jr., *J. Non-Cryst. Solids* **79**, 155 (1986).

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

¹⁷D. S. Greywall and G. Ahlers, *Phys. Rev. A* **7**, 2145 (1973).

¹⁸P. C. Hohenberg, A. Aharony, B. I. Halperin, and E. D. Siggia, *Phys. Rev. B* **13**, 2986 (1976).

¹⁹The superfluid density must be averaged over a length scale which is large, i.e., coarse grained, compared with the length scale of the disorder. In particular, we have taken an average over the total volume of the sample cell.

²⁰D. W. Schaefer, B. C. Bunker, and J. P. Wilcoxon, *Phys. Rev. Lett.* **58**, 284 (1987); P. Wiltzius, F. S. Bates, S. B. Dierker, and G. D. Wignall, *Phys. Rev. A* **36**, 2991 (1987).

²¹D. W. Schaefer and K. D. Keefer, *Phys. Rev. Lett.* **56**, 2199 (1986); R. Vacher, T. Woignier, J. Pelous, and E. Courtens, *Phys. Rev. B* **37**, 6500 (1988); P. Schmidt, private communication.

²²J. A. Lipa and T. C. P. Chui, *Phys. Rev. Lett.* **51**, 2291 (1983).