Specific Heat of Disordered Superfluid $^3$He

H. Choi, K. Yawata, T.M. Haard, J.P. Davis, G. Gervais, N. Mulders, P. Sharma, J.A. Sauls, and W. P. Halperin

Department of Physics and Astronomy,
Northwestern University, Evanston, Illinois 60208

(Dated: Version November 7, 2018)

PACS numbers: 67.57.-z, 67.57.Bc, 67.57.Pq

The specific heat of superfluid $^3$He, disordered by a silica aerogel, is found to have a sharp discontinuity marking the thermodynamic transition to superfluidity at a temperature reduced from that of bulk $^3$He. The magnitude of the discontinuity is also suppressed. This disorder effect can be understood from the Ginzburg-Landau theory which takes into account elastic quasiparticle scattering suppressing both the transition temperature and the amplitude of the order parameter. We infer that the limiting temperature dependence of the specific heat is linear at low temperatures in the disordered superfluid state, consistent with predictions of gapless excitations everywhere on the Fermi surface.

Two essential characteristics of superconductors are manifest in the specific heat. First, there is a jump at the transition temperature of magnitude that depends on the strength of the electron pair interactions that lead to superconductivity. Secondly, the temperature dependence at low temperature gives a fingerprint of the energy gap structure. Consequently, the measurement of the specific heat is one of the most fundamental to understanding the nature of superconductivity. In fully gapped superconductors, such as aluminum, the specific heat decreases exponentially with decreasing temperature following an Arrhenius relation, thereby demonstrating unambiguously the existence of a full gap in the electron quasi-particle excitation spectrum and providing a direct measurement of its size. Both of these basic thermodynamic behaviors conform to predictions of the Bardeen, Cooper, and Schrieffer theory, and can be expected to hold in general for any condensate of fermion pairs into a state that is fully gapped. This is precisely what is expected and observed for the B-phase of superfluid $^3$He, a $p$-wave superfluid with an isotropic energy gap, similar to conventional s-wave superconductors.

Shortly after the success of BCS theory it was sought to understand how perturbations, such as impurities, modify superconducting behavior. It was found that magnetic scattering of quasiparticles suppresses both the transition temperature and the gap magnitude leading to ‘gapless superconductivity’. The same should be true for all superconductors and fermion superfluids even those that do not have s-wave states in which case all forms of elastic scattering will produce these suppression effects. In particular it should be true for superfluid $^3$He. In this letter we present systematic measurements of the heat capacity in the B-phase of superfluid $^3$He, disordered by impurities, demonstrating suppression of the transition temperature, suppression of the order parameter, and gapless superfluid behavior.

Superfluid $^3$He is a unique example of a Cooper pair condensation. It was the first unconventional pairing state discovered. By unconventional we mean that there are symmetries of the normal Fermi liquid that are spontaneously broken in the superfluid phases in addition to gauge symmetry, e.g. spin and spatial rotations. In this sense $^3$He is similar to cuprate superconductors and to the heavy fermion superconductor UPt$_3$ as well as a number of other recently discovered superconducting compounds. However, in contrast to the superconductors, superfluid $^3$He is a neutral system, easily prepared as the purest known material, whose general properties are very well established. It is not so straightforward, however, to introduce impurity scattering in a controlled way in $^3$He.

Porto et al. and Sprague et al. found that they could use very dilute silica aerogels to introduce impurity scattering in superfluid $^3$He and subsequently there has been a wealth of new information about the nature of this modified superfluid state. Impurity scattering models for $^3$He were developed by Thuneberg et al. with clear predictions for the suppression of the transition temperature and the heat capacity discontinuity. Sharma and Sauls have calculated transport behavior as well as the density of quasiparticle excitations inside the gap which define its gapless superfluid behavior. Our experimental results are quantitatively consistent with the calculations of these basic thermodynamic properties.

Earlier work of He et al. and preliminary results from our laboratory have provided convincing evidence for the existence of a heat capacity anomaly at the transition temperature. Additionally, Fisher et al. found that the thermal conductivity in the disordered A-superfluid state in a magnetic field has a linear temperature dependence when extrapolated to very low temperature. If the heat current were carried by gapless excitations at low energy there should be evidence for these excitations in the low temperature specific heat.

Our low temperature calorimeter was cooled with a PrNi$_5$ adiabatic nuclear demagnetization refrigerator to temperatures below 0.75 mK above which we made measurements of heat capacity using an adiabatic calorimeter.
ric method. Temperature was determined from measurement of the ac susceptibility of a lanthanum diluted cerium magnesium nitrate salt, calibrated with respect to a $^3$He melting curve thermometer according to the Greywall temperature scale[4]. The calorimeter volume was measured to be 1.91 ± 0.02 cm$^3$ with the aerogel sample in place, leaving 44 % of the space available for bulk $^3$He. The porous silica aerogel was in the form of a right circular disk 0.380 cm thick and 1.902 cm in diameter. From its dry weight, 42.4 mg, we found the porosity to be 98.2%. The sample was first cooled by nuclear demagnetization. By opening a cadmium superconducting heat switch we could isolate the sample cell from the nuclear stage, allowing the $^3$He to warm up slowly owing to the ambient heat leak, typically $\dot{Q} \sim 0.1$ nW, starting in the B-phase[19]. Heat pulses were applied with $\Delta Q$ in the range 0.1 $\sim$ 0.5 $\mu$J for 30 $\sim$ 60 s, and the temperature increments $\Delta T$ were measured to obtain the heat capacity $C_{tot} = \Delta Q/\Delta T$ shown in Fig. 2 after subtraction of the addendum. A warm-up trace without heat pulses, in the form $T^{-1} = C/\dot{Q}$, is shown in the inset to Fig.1 providing our highest resolution of both transition temperatures, in bulk liquid $^3$He and $^3$He in aerogel.

We assume that liquid $^3$He above its transition to superfluidity is a Fermi liquid with known interaction parameters even within the aerogel. This is reasonable since the aerogel structure is on a much larger scale than the Fermi wavelength[20]. Using the specific heat measurements by Greywall[4] on bulk $^3$He we can deduce from our experiments, a) the calorimeter background at temperatures above the superfluid transition in aerogel, b) the volumes of $^3$He in the aerogel and in the bulk, c) the heat capacity jump in the disordered superfluid phase, and d) the temperature dependence of the heat capacity in the disordered superfluid state.

The background heat capacity, $C_{add}$, can be entirely attributed to solid $^3$He on the surface of the aerogel. Two layers of $^3$He are paramagnetic[3, 15] and can be removed by substitution with two layers of non-magnetic $^4$He reducing the addendum to less than 0.25 mJ/K. The helium isotope mixture experiments were limited to $\geq 4$ mK owing to long thermal time constants in the calorimeter. The temperature and pressure dependence of the addendum is shown in Fig. 3, of a comparable magnitude[17] to that reported in vycor glass[18].

The measured heat capacity, $C_{tot}$, corrected for the
addendum, $C_{add}$, is

$$C(T, P) = C_{tot} - C_{add} = V_a c_a + V_{BCB}$$  \hspace{1cm} (1)$$

shown in Fig. 2. Above the bulk transition temperature, $T_c$, $C(T, P)$ is proportional to temperature, since the specific heats of the fluid inside, $c_a$, and outside, $c_B$, the aerogel are identical: $c_a = c_B = \gamma T$ where $\gamma = (\pi^2/3)k_B^2N_0$ and $N_0$ is the density of states at the Fermi energy. Knowing the total volume $V_a + V_B$ of aerogel and bulk fluids, we can determine the addendum in this temperature range. At $T_c$ the bulk heat capacity jump was compared with Greywall’s values to give a direct measure of $V_a$ from which we found the fluid volume in the aerogel to be $1.028 \pm 0.021 \text{cm}^3$, consistently at all pressures and close to the measured geometry of the aerogel giving a pore volume of $1.062 \pm 0.006 \text{cm}^3$. With $V_a$ and Eq. 1, we can determine the addendum in the range $T_c > T > T_{c,a}$, by comparing our measurements with the known specific heat of the bulk superfluid.

We must also allow for contributions to the heat capacity from a normal state region at the surface of the heat exchanger, approximately a coherence length, $\xi(T)$, in depth. This small contribution is found by fitting the slow warm-up curves as shown in the inset to Fig.1 to obtain an effective area of the heat exchanger $A = 1.71 \text{m}^2$, valid at all pressures. The temperature dependence of the addendum at 1.02 bar is smooth to below 1 mK. Consequently, it is reasonable to assume that this is the case at higher pressures and to extend linearly the temperature dependence of the addendum from $T_{c,a}$ to lower temperatures in order to extract the heat capacity, $C_a$, of the disordered superfluid.

We show $C_a/T$ on the right side of Fig.2 after subtracting the bulk contribution, the surface contribution, and the addendum. The heat capacity jumps appear quite sharp in the figure but substantially reduced even compared to the weak coupling BCS value of $\Delta C/C = 1.43$. They are shown as a function of pressure in Fig. 4, normalized to their bulk values. Consequently, the amplitude of the order parameter is also reduced since the jump is related to the temperature dependence of the gap function, $\Delta_a(T)$, near $T_{c,a}$, in the Ginzburg-Landau limit,

$$\Delta_a(T) = \pi k_B T_{c,a} \left( \frac{2 \Delta C_a}{C_a} \frac{T_{c,a}}{T} \frac{T}{T_{c,a}} \right)^{1/2} \ \ \ \ \ T \lesssim T_c$$  \hspace{1cm} (2)$$

At lower temperatures, below $T_{c,a}$, we note that $C_a/T$ appears to be quite linear. We take advantage of this fact and we invoke the third law of thermodynamics,

$$0 = \int_{T_{c,a}}^{T} (\gamma - C_a/T) \,dT,$$  \hspace{1cm} (3)$$

to determine the limiting behavior of $C_a$ as $T \to 0$ shown by dotted curves in Fig. 2. Graphically, this means that for each pressure the two shaded areas must be equal. If the heat capacity is a monotonic function of temperature, as is expected theoretically, then to satisfy Eq. (3), $C_a/T$ must have a non-zero intercept at $T = 0$ requiring a non-zero density of states, $N_a(0)$, at the Fermi energy in the superfluid state. The curves describing this behavior in the disordered superfluid were constructed phenomenologically to satisfy the constraints and our interpretation does not depend on its functional form nor is it particularly sensitive to the data at the lowest temperatures. For example in Fig.2 at $P = 29.02$ bar the data fall below the curve likely owing to our overestimate of the addendum. The low energy excitations affecting the low temperature specific heat are a broad band of Andreev bound states, centered at the Fermi energy, that form near the aerogel strands.

As a result superfluid $^3$He-B in aerogel is a ‘gapless superfluid’.

The simplest theoretical description considers isotropic quasiparticle scattering spread homogeneously throughout the aerogel volume, characterized by a single parameter, the transport-mean-free path, $\lambda_T$. This model (HISM) provides a qualitatively consistent account of the superfluid transition temperature suppression, the suppression of the A-phase and the polycritical point, measurements of spin diffusion, and the thermal conductivity. Here we see that the model gives a quantitatively consistent picture for the heat capacity jump over a wide range of pressure. A calculation with the HISM gives a best fit value of $\lambda_T = 180 \text{ nm}$. In this fit we rescale strong coupling contributions to the free energy by the ratio $T_{c,a}/T_c$, where $T_{c,a}$ is calculated self-consistently within the HISM. We also find reasonable agreement with the prediction from the HISM for the density of states of gapless excitations. The calculations as a function of

FIG. 3: Heat capacity addendum. The temperature and pressure dependence of the heat capacity addendum is shown for bulk $^3$He in aerogel with an estimated accuracy of 0.25 mJ/K.
pressure for $N_a(0)$ are shown in Fig.4 for several values of $\lambda_T$ near 150 nm and are qualitatively consistent with our analysis of the heat capacity jump, $\lambda_T = 180$ nm.

Discrepancy between the experiment and the model for the density of states can be attributed in part to estimated systematic errors including the extrapolation of the addendum to $T \leq T_{c,a}$ and possible anisotropic or inhomogeneous scattering. Nonetheless, the consistency that is apparent within the heat capacity experiment provides strong evidence that elastic scattering suppresses superfluidity and is approximately isotropic and homogeneous. It is interesting to compare our results with unconventional superconductors in several cases. Precision measurements of the transition temperature of UPt$_3$ indicate [24] suppression of the transition temperature with increased scattering but, in this case, with a significant anisotropy. In cuprates doped with Zn impurity, such as YBa$_2$(Cu$_{1-y}$Zn$_y$)$_3$O$_7$, suppression effects have been observed in the heat capacity [24], strikingly similar to those we report here.

We acknowledge support from the National Science Foundation DMR-0244099 and we thank Ryuji Nomura, and John Halpine for assistance in the early stages of this project and helpful conversations with Yoonseok Lee, John Reppy, and Jeevak Parpia.

[1] N.E. Phillips. Phys. Rev. 114, 676 (1959).
[2] M. Tinkham, Introduction to Superconductivity McGraw-Hill, 2nd edition, 1959.
[3] A.J. Leggett, Rev. Mod. Phys. 47, 331 (1975).
[4] D.S. Greywall, Phys. Rev. B 33, 7520 (1986).
[5] A.A. Abrikosov, L.P. Gor’kov, Sov. Phys. JETP 8, 1090 (1959).
[6] A.A. Abrikosov, L.P. Gor’kov, Sov. Phys. JETP 12, 1243 (1961); T. Tsuneto, On Dirty Superconductors Technical Report of the Institute of Solid State Physics, University of Tokyo, series A, No. 47 (1962); A.I. Larkin, JETP Lett. 2, 130 (1965).
[7] J.V. Porto and J.M. Parpia, Phys. Rev. Lett. 74, 4667 (1995).
[8] D.T. Sprague et al., Phys. Rev. Lett. 75, 661 (1995).
[9] D.T. Sprague et al., Phys. Rev. Lett. 77, 4568 (1996).
[10] E.V. Thuneberg, S.K. Yip, M. Fogelstrom, and J.A. Sauls, Phys. Rev. Lett. 80, 2861 (1998).
[11] P. Sharma and J.A. Sauls, J. Low Temp. Phys. 125, 115 (2001).
[12] J. He, A.D. Corwin, J.M. Parpia, and J.D. Reppy, Phys. Rev. Lett. 89, 115301 (2002).
[13] K. Yawata, T.M. Haard, G. Gervais, N. Mulders, and W.P. Halperin, Physica B, 329, 327 (2003).
[14] S.N. Fisher, et al. Phys. Rev. Lett. 91, 105303 (2003).
[15] B.I. Barker, Y. Lee, L. Polukhina, D.D. Osheroff, L.W. Hrubesh and J.F. Poco, Phys. Rev. Lett. 85, 2148 (2000).
[16] A. Golov and F. Pobell, Phys. Rev. B 53, 12647 (1996).
[17] The aerogel surface area is 45 m$^2$ from helium BET measurements on a similar sample giving a surface solid specific heat of 11 J/Km near 4 mK similar to that found by Golov and Pobell [16] for $^3$He on vycor glass, 13 J/Km$^2$.
[18] There have been various reports of bulk $^3$He regions hidden within the aerogel volume, sometimes as large as 50%, Yu. M. Bunkov et al., Phys. Rev. Lett. 85, 3456 (2000). In the present case this is 3%.
[19] G. Gervais, K. Yawata, N. Mulders and W.P. Halperin, Phys. Rev. B 66, 054528 (2002).
[20] J.A. Sauls and P. Sharma, Phys. Rev. B 68, 224502 (2003).
[21] E.Collin, Ph.D. thesis, Université Joseph Fournier Grenoble 2002.
[22] P. Sharma and J.A. Sauls, Physica B 329-333, 313 (2003).
[23] J.A. Sauls, to be published.
[24] J.B. Kycia, J.-I Hong, M.J. Graf, J.A. Sauls, D.N. Seidman, and W.P. Halperin, Phys. Rev. B 58, R603 (1998).
[25] J.W. Loram et al. Physica C, 235 134 (1994).