Thermodynamic Magnetization Discontinuity at the *A*-*B* Transition in Superfluid ^{3}He

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The difference between total thermodynamic magnetizations in the *A* and *B* phases of superfluid ^{3}He was measured at the equilibrium *A*-*B* transition temperature over the entire range of applicable magnetic fields at pressures from 0 to 27 bars. We present evidence that dynamic measurements (NMR) only partially account for the thermodynamic magnetization changes in the superfluid. The present experiment also measures the nontrivial strong-coupling correction to the magnetic field coupling strength in the Landau-Ginzburg free energy. [S0031-9007(98)06626-5]

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Magnetism in superfluid ^{3}He must come from the nucleus [1]. This conventional wisdom derives from the fact that the only source of magnetization other than the nuclear moment lies in the electronic degrees of freedom which are apparently frozen out by the 8 orders of magnitude difference between available thermal energy, 1 mK, and the atom's lowest excited energy, 20 eV.

Nevertheless, the most persistent controversy in the field of superfluid ^{3}He involves the discrepancy between measurements of the total thermodynamic magnetization [2,3] and dynamic (NMR) measurements [4] which are nuclear selective. This discrepancy has consequences for microscopic models of *normal* liquid ^{3}He, for inferred values of the superfluid's characteristic Landau-Ginzburg phenomenological "β parameters," and for the identification of the symmetry of the ordered state itself [5].

Here we report precision measurements of the magnitude of the discontinuity in the total thermodynamic magnetization at the first-order *A*-*B* transition in superfluid ^{3}He over most of the superfluid phase diagram [6]. This is the first measurement of the thermodynamic magnetization of the superfluid over a sufficiently wide pressure and magnetic field range to enable a direct comparison between NMR and thermodynamic magnetization data.

A schematic view of the temperature and field dependences of nuclear magnetization in the *A* and *B* phases of superfluid ^{3}He, normalized to that of the normal liquid, is shown in Fig. 1. The normalized *A* phase magnetization is a constant and, to an accuracy of better than 1%, equal to the magnetization in the normal state. The normalized *B* phase magnetization, however, is dependent upon both field and temperature. The limiting zero field behavior is the lowest curve. Qualitatively, the effect of a finite field is to shift this curve upwards by an amount proportional to the square of the field. In addition, the temperature at which the *A*-*B* transition occurs is suppressed by a field approximately quadratically as measured in Ref. [7].

In order to avoid the difficulties of background magnetization drifts we have taken the approach of measuring the magnetization discontinuity at the *A*-*B* transition as a function of field as shown in Fig. 1. This measurement intrinsically incorporates the temperature dependence, so that the extrapolation to zero field and T_c is controlled and accurate.

Our magnetization measurements were performed in the simplest possible configuration, where we place a small superconducting coil around a tube containing liquid ^{3}He. This coil is connected to the input coil of a dc SQUID. The tube and coil are surrounded by a superconducting magnet capable of producing 0.5 T, suppressing the *B* phase to $T = 0$. The complications of this experiment come from the need to carefully shield the coil from external noise, and the need to accurately calibrate the sensitivity of the coil and SQUID. For this reason, the magnet is a specially designed "self-shielded" magnet with active compensation at each end and around the outside. This assembly is capable of producing full field at the center with less than 1% of this field appearing at a lead shield that surrounds the magnet, coil, and sample.

The calibration of our sensitivity to changes in magnetization of the ^{3}He depends on our knowledge of the dimensions of the coil, the diameter of the sample region, and the field produced by the magnet. Fortunately, most

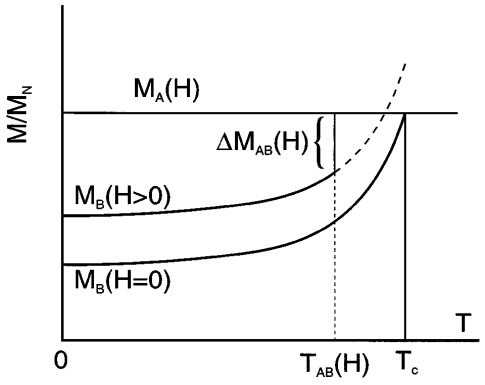

FIG. 1. Schematic field and temperature dependences of the magnetization in each superfluid phase, assuming a constant subtracted electronic contribution, normalized to the normal fluid's magnetization in the same field. This work measures ΔM_{AB} only at the transitions in finite fields as indicated by the brace.

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of the geometrical quantities which affect sensitivity can be replaced by two measurable quantities. These are the mutual inductance between the sense coil and the magnet, $(\Delta\phi_{\text{sense}}/\Delta I_{\text{magnet}}) = 16.95\ \mu\text{H}$, and the output voltage change in the SQUID to a current step in the main magnet $(\Delta V_{\text{SQUID}}/\Delta I_{\text{magnet}}) = 2.28 \times 10^{-8}\ \Omega$. So the flux sensitivity is given by

$$\left(\frac{\Delta M_{\text{He}}}{\Delta V_{\text{SQUID}}}\right) = \left(\frac{\Delta M_{\text{He}}}{\Delta\phi_{\text{sense}}}\right)\left(\frac{\Delta\phi_{\text{sense}}}{\Delta I_{\text{magnet}}}\right) \Big/ \left(\frac{\Delta V_{\text{SQUID}}}{\Delta I_{\text{magnet}}}\right). \tag{1}$$

The ratio of the change in magnetization of ^{3}He to the amount of flux through the sense coil $(\Delta M_{\text{He}}/\Delta\phi_{\text{sense}})$ depends only on the area of the sample tube (3.08 mm^2) and the number of turns in the sense coil (26).

Our temperature was measured using the susceptibility of diluted cerium magnesium nitrate (CMN). This thermometer was calibrated against the normal to superfluid *A* phase transition as a function of pressure using the temperature scale of Greywall [8]. A second thermometer using the NMR susceptibility of Pt was also used to extend the calibration of the LCMN to below 1 mK. The Pt temperature was proportional to the LCMN scale over the calibration region.

We measured a series of magnetization discontinuities at increasing applied fields. Figure 2 shows the quantity $M_B/M_N \equiv [M_N - \Delta M_{AB}(H)]/M_N$, which would be the *B* phase magnetization under the conventional assumption that $M_A = M_N$. For comparison, we have indicated the data from Scholz [9] (the most extensive set of NMR data available). Errors in comparison between these two sets of data due to thermometry differences were eliminated by using the accurately known magnetic field of Scholz [9] and converting to the phase diagram as measured in our laboratory [10].

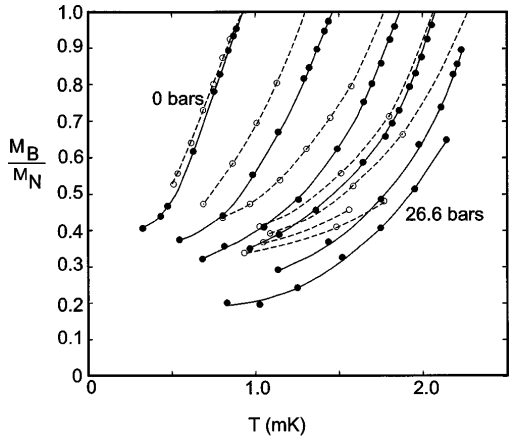

FIG. 2. Measured magnetization of the *B* phase at the field-dependent transition temperature. This graph assumes that the *A*-phase magnetization is equal to that in the normal state. Solid curves are drawn through our data points (•) at pressures of 0, 4.8, 10.6, 15.4, 22.1, and 26.6 bars, while the dashed curves are drawn through the NMR data points of Scholz (Ref. [8]) at pressures of 0, 3.1, 9.0, 15.0, 21.0, 27.0, and 32.7 bars.

From this direct comparison it is clear that the disagreement between SQUID and NMR values of magnetization is greatest at higher pressures and lower temperatures. For example, it is clear that NMR values of the low temperature limiting values of the magnetization at high pressures approach $0.3M_N$ while the SQUID values are considerably smaller. [This limiting magnetization should not be confused with the limiting NMR susceptibility at *low fields* which coincidentally approaches $(0.3-0.4)\chi_N$. The values plotted here are measured at the *highest field* for which the *B* phase exists at each temperature.] Even the apparent similarity at 0 bar is somewhat an artifact of the method of plotting, which in this case makes both NMR and SQUID data essentially vertical. A careful comparison of the slope, however, shows a nearly 10% disagreement.

We now return to the issue of the validity of SQUID measurements of magnetization, compared to NMR measurements. Early criticisms of SQUID measurements, regarding calibration and regarding limited field and pressure ranges, are no longer valid. Webb's measurement [3] was insensitive to this problem because he used the same SQUID (and calibration) to measure *both* static and dynamic differences between the *A* and *B* phases (at high pressure and in only one field) and found they disagreed. While the calibration of the SQUID apparatus is critical in our measurement, our measurements agree with Webb's where he worked, and extend the measurements over the phase diagram. Where comparable, all SQUID measurements agree with each other [11], and all NMR measurements agree with themselves. The conclusion must be that SQUID-based experiments consistently measure something different from what NMR does.

An objection is occasionally raised that the Kramers-Kronig relations demand that NMR and SQUID measurements should agree. This is incorrect. The Kramers-Kronig relations provide an equivalence between the true (static) thermodynamic susceptibility (measured in our present experiment) and an integral involving the imaginary component of the dynamic susceptibility (measured as absorption in NMR) *integrated over all frequencies*. Restricting the integral to only those frequencies near the nuclear Larmor frequency yields the *nuclear contribution* to the thermodynamic susceptibility, but ignores the electronic contribution, and in the case of ^{3}He thereby gets even the sign of the total susceptibility wrong. The relevant frequency range in which the "missing weight" is located is nowhere near typical Larmor frequencies, but is rather set by atomic level energies.

Given that the thermodynamic value of the magnetization discontinuity at the *A*-*B* transition differs from that used previously in inferring values for Landau-Ginzburg phenomenological β parameters, we finally turn to the issue of their now improved measurement. Since these

parameters apply only asymptotically as $T \to T_c$, we focus attention on the low-field limiting behavior of our data. Figure 3 shows our measured magnetization discontinuities at four pressures normalized to the magnetic field (normal state magnetization) as a function of temperature close to T_c.

The magnetization discontinuity at the A-B transition has a field dependence, which varies as $\Delta M \propto B^3$ asymptotically in the low field $T_{AB} \to T_c$ limit. Two powers of the field derive from the quadratic field dependence of the temperature of the A-B transition ($1 - T_{AB}/T_c \propto B^2$). The third power in B comes from the relationship between magnetization and magnetic field in both A and B phases, which to lowest order is linear ($M_A, M_B, M_N \propto B$). The first correction to each of these effects is two powers higher in B, leading to a B^5 correction to ΔM. The limiting low-field value of the discontinuity (after accounting for these higher order correction terms) is

$$\frac{\Delta M}{M_N} = \hat{g} M(\beta)(1 + F_0{}^a)^{-1}[1 - T_{AB}(B)/T_c], \quad (2)$$

where $F_0{}^a$ is a Fermi liquid parameter with a known dependence on pressure [12], $\hat{g}$ is the strong-coupling correction to the magnetic field coupling strength, and $M(\beta)$ depends solely upon the Landau-Ginzburg parameters

$$M(\beta) = \frac{10}{3}\left[\frac{2\beta_{13} - \beta_{345}}{\beta_{245}\beta_{345}(3\beta_{12} + \beta_{345})}\right]^{1/2}. \quad (3)$$

Here we adopt the Mermin-Stare convention wherein repeated indices denote summation, e.g., $\beta_{13} = \beta_1 + \beta_3$. In this expression we have also taken the conventional identification of the order parameter of the A phase to be the axial state [13].

The strong-coupling correction $\hat{g}$ has ordinarily been taken to be equal to its weak coupling value of unity [7]. Theoretical expectations are that it should not deviate from this value significantly, a prejudice which is supported by experimental evidence below.

To better exhibit the corrections to the asymptotic formula in Eq. (1) we divide the normalized magnetization discontinuity by $(1 - T_{AB}/T_c)$. This has the added benefit of providing a cross-check that we accurately know the magnetic field and transition temperature. If we did not know them accurately, small errors would cause the denominator to approach zero at a different point than the numerator, leading to spurious divergences in the plotted points either up or down depending upon the sign of the errors. The lack of such spurious effects gives us confidence in our final results.

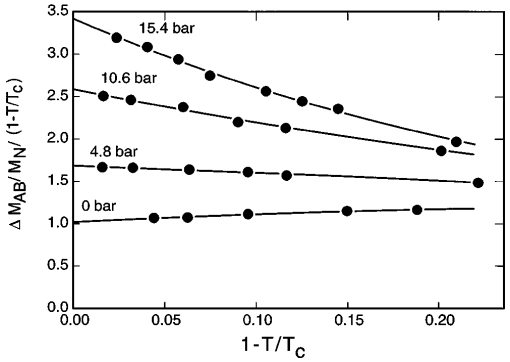

FIG. 3. Magnetization discontinuity at the A-B transition with the leading temperature dependence divided out. The extrapolated intercept at T_c determines the β parameter combination of Eq. (3).

One goal of this measurement is a set of values of $M(\beta)$ defined in Eq. (2). In Fig. 3, the feature which determines $M(\beta)$ is the extrapolation of our data to the intercept on the vertical axis as $T_{AB} \to T_c$. This intercept is the B^3 coefficient of ΔM_{AB}. The finite slope of the data corresponds to the leading B^5 correction, which if ignored could have affected our derived values of $M(\beta)$ by as much as 30%. The curvature of the data (the next order B^7 correction) turns out to have an insignificant effect upon our derived values of $M(\beta)$.

In order to extract values of $M(\beta)$ from the data in Fig. 3 we have to know the strength of the magnetic field coupling parameter $\hat{g}$. In previous reports from our laboratory (cf. Ref. [7]) this parameter was taken to be identically 1, i.e., equal to its weak-coupling value. Although it is always close to 1, the small corrections have a significant impact on the analysis of this and previous work in terms of the β parameters. The present measurement gives the first reliable quantitative value for its strength including real strong-coupling corrections at the polycritical point (PCP) as we now describe.

At the pressure of the polycritical point the magnetization parameter $M(\beta)$ is determined solely by a known specific heat measurement through the identity [14],

$$M(\beta)_{\mathrm{PCP}} = \frac{2}{3}\frac{(\Delta C_B/C_N)_{\mathrm{PCP}}}{(\Delta C_B/C_N)_{\mathrm{w.c.}}}, \quad (4)$$

where $\Delta C_B/C_N$ is the normalized specific heat jump at the normal to superfluid-B transition, whose weak-coupling value ("w.c.") is $12/7\zeta(3) \approx 1.426$. According to Greywall's measurement of ΔC_B [8], Eq. (4) requires $M(\beta)$ at the PCP to be 0.87. Our experiment determines the product of $\hat{g}M(\beta)$ at the PCP to be 0.96 (to within a few percent, the limit of accuracy of our extrapolation to the PCP). Combining these two results, we find that $\hat{g}$ at the PCP is 1.10. This supports the theoretical bias that strong-coupling corrections to the coupling strength $\hat{g}$ should be small.

While the identity (4) applies at only one pressure (PCP) the value of $\hat{g}$ is expected theoretically to always remain close to the weak-coupling value. Since all strong-coupling corrections are expected to scale as a low order power series in T_c/T_f, and the changes in $\hat{g}$ are small, we have applied a linear scaling in T_c/T_f in deriving the β parameters reported elsewhere [15]. As evidence for the reasonableness of the linear scaling in T_c/T_f we plot in

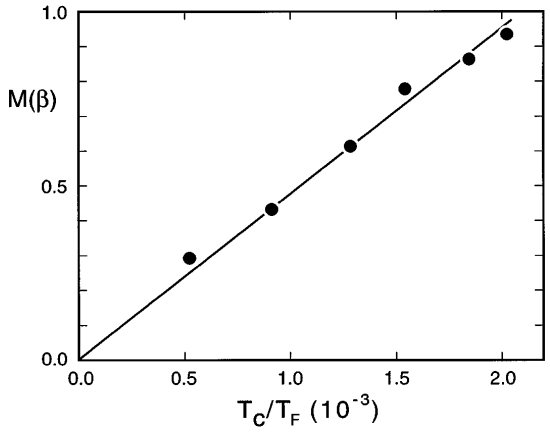

FIG. 4. Derived values of $M(\beta)$ at all measured pressures showing the accuracy of its linear scaling in T_c/T_f. The point (0,0) is the weak-coupling limit.

Fig. 4 our deduced values for $M(\beta)$ at each pressure, in which it is clear that a low order power series is adequate.

We believe that this measurement of $\hat{g}$ is more reliable than previous reports based upon NMR data as evidenced by the following thermodynamic requirement. In a fixed field near T_c the B-phase magnetization as a function of temperature should show a linear dependence with a coefficient at an arbitrary pressure given by

$$\frac{dM_g/dT}{(dM_B/dT)_{\text{w.c.}}} = \hat{g}\,\frac{\Delta C_B/C_N}{(\Delta C_B/C_N)_{\text{w.c.}}}. \qquad (5)$$

If the analysis of NMR measurements, such as those reported in Ref. [16] were accurate, then in order to satisfy this thermodynamic requirement, the strong-coupling term $\hat{g}$ would have to change by a factor of 2 as pressure increases from 0 to the PCP. This violates theoretical expectations and would certainly be unprecedented in superfluid ^{3}He. In fact, a reanalysis of the data in Ref. [16] by its authors [17] shows that the extrapolation to T_c required in the original analysis was difficult to control well. We conclude that our present measurement is a reliable measure of $\hat{g}$.

We have demonstrated that the *difference* in thermodynamic magnetizations between the A and B phases differs from the conventionally accepted NMR value. We cannot, however, identify whether the source of this difference lies in the thermodynamic magnetization being larger than expected in the A phase, or smaller than expected in the B phase. To date there has been no measurement of the absolute thermodynamic magnetization in either superfluid phase, only of their difference.

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[1] Here we discount the temperature- and field-independent electronic core diamagnetism.
[2] D. N. Paulson, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. **31**, 746 (1973); R. E. Sager, R. L. Kleinberg, P. A. Warkentin, and J. C. Wheatley, J. Low Temp. Phys. **31**, 409 (1978).
[3] R. A. Webb, Phys. Rev. Lett. **38**, 1151 (1977).
[4] D. D. Osheroff, Phys. Rev. Lett. **33**, 1009 (1974); A. I. Ahonen, T. A. Alvesalo, M. T. Haikala, M. Krusius, and M. A. Paalanen, Phys. Lett. **51A**, 279 (1975).
[5] C. M. Gould, Physica (Amsterdam) **178B**, 266 (1992).
[6] Early work on this experiment has been reported by I. Hahn, S. T. P. Boyd, H. M. Bozler, and C. M. Gould, J. Low Temp. Phys. **101**, 781 (1995).
[7] Y. H. Tang, I. Hahn, H. M. Bozler, and C. M. Gould, Phys. Rev. Lett. **67**, 1775 (1991).
[8] D. S. Greywall, Phys. Rev. B **33**, 7520 (1986).
[9] H. N. Scholz, Ph.D. thesis, Ohio State University, 1981 (unpublished).
[10] I. Hahn, Ph.D. thesis, University of Southern California, 1993 (unpublished).
[11] A short review of these experiments appears in H. M. Bozler, P. D. Saundry, I. Hahn, S. T. P. Boyd, and C. M. Gould, J. Low Temp. Phys. **89**, 5 (1992). An exception to our general conclusion appears in S. T. P. Boyd and G. W. Swift, J. Low Temp. Phys. **87**, 35 (1992) who claim agreement between their SQUID and other NMR results, but their data were too limited in pressure and temperature to be useful in resolving the ΔM_{AB} discrepancy.
[12] H. H. Hensley, Y. Lee, P. J. Hamot, T. Mizusaki, and W. P. Halperin, J. Low Temp. Phys. **90**, 149 (1993).
[13] If the order parameter is actually the axiplanar state, then this expression and the numbers derived from it change, but the qualitative conclusions of this paper are not altered.
[14] This identity is easily derived from the definition of $M(\beta)$ by inserting the condition for the PCP, that the A and B phase-free energies in zero field are equal, $3\beta_{13} = 2\beta_{345}$.
[15] H. M. Bozler and C. M. Gould, Czech. J. Phys. **46**, Suppl. S1, 165 (1996).
[16] J. B. Kycia, T. M. Haard, M. R. Rand, H. H. Hensley, G. F. Moore, Y. Lee, P. J. Hamot, D. T. Sprague, W. P. Halperin, and E. V. Thuneberg, Phys. Rev. Lett. **72**, 864 (1994).
[17] W. P. Halperin (private communication).