

## High-Resolution Nuclear Magnetic Resonance of Superfluid $^3\text{He-B}$

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High-resolution nuclear magnetic resonance measurements of bulk superfluid  $^3\text{He-B}$  have been performed at temperatures above 0.5 mK and at pressures from 0.3 to 21.7 bars. We have found that the resonance frequency is shifted from the Larmor frequency of the normal fluid. According to the theory of Greaves the shift at the superfluid transition determines a specific combination,  $\beta_{345}$ , of the 5 fourth-order coefficients of the order parameter invariants used in the Ginzburg-Landau description of superfluid  $^3\text{He}$ . We found that  $\beta_{345}$  approaches the weak coupling limit at low pressure, and decreases at higher pressures qualitatively consistent with the theory of Sauls and Serene but in disagreement with the results of Tang *et al.*

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One of the most intriguing aspects of superfluid helium-three is the property of spontaneously broken spin-orbit symmetry [1,2]. As a consequence, the nuclear dipole interaction gives rise to a static local field that shifts the Larmor frequency of the transverse nuclear magnetic resonance (NMR),  $\omega_{\perp}$ , away from its value in the normal liquid state,  $\omega_0 = \gamma H_0$ , where  $\gamma$  is the gyromagnetic ratio and  $H_0$  is the external field [3].

In the superfluid *A* phase of helium, generally accepted to be the axial state [1,2], the transverse frequency shift,  $\omega_{A\perp} - \omega_0$ , is given by a quadratic relation in terms of the longitudinal resonance frequency,  $\omega_{A\parallel}$ , and the external field [1],  $\omega_{A\perp}^2 = \omega_{A\parallel}^2 + \omega_0^2$ . Its measurement by Osheroff, Gully, Richardson, and Lee [4] was a significant step in the identification of superfluid  $^3\text{He}$  as a *p*-wave, pair-correlated state of the BCS type.

The *B* phase of superfluid helium is known to be the isotropic *p*-wave state [1,2]. For small excitations, tipping pulses much less than  $104^\circ$ , there is a prediction [5-7] of a shift in the transverse resonance frequency,  $\omega_{B\perp} = \gamma H_0(1+g)$ . The frequency shift, often referred to as the *g* shift, results from distortion of the energy gap by the magnetic field. It is small,  $\approx 1/10^5$ , which is approximately 3 orders of magnitude less than the transverse frequency shift that was first reported in  $^3\text{He-A}$  [4]. This effect in  $^3\text{He-B}$  was noted by Osheroff [8] at high pressure near 34 bars, but has not been studied in detail since then. A preliminary report of our results at a pressure of 1.03 bars was given by Hensley *et al.* [9]. The high resolution of our frequency measurements has permitted a study of the dependence of the *g* shift on temperature and pressure to an accuracy of 1%. We have extrapolated our data into the Ginzburg-Landau regime, close to the superfluid critical temperature. In this limit, the *g* shift can be related to thermodynamic parameters of the phenomenological Landau theory giving a measure of strong coupling interactions and their pressure dependence in

superfluid  $^3\text{He}$ . At low pressure, the weak coupling approximation in the BCS theory [2] accounts for most data rather well. At high pressure, strong coupling effects are clearly evident, but have been found by Gould and co-workers [10], from analysis of various experiments, to be in disagreement with existing theory. Our measurements of the *g* shift give an independent experimental determination of strong coupling effects, in better agreement with theory than previously believed.

The sample was a cylinder of  $^3\text{He}$ , 0.5 cm in diameter and 0.5 cm long, contained in an epoxy cell [11] and connected to a  $^3\text{He}$  reservoir through an opening 0.125 cm in diameter and 0.8 cm long. The NMR coil, 1.25 cm long and 1 cm diameter, was thermally isolated from the sample cell, and provided a homogeneous rf field over the sample to within 1%. A homogeneous static field of 0.12 T was produced by a superconducting NMR magnet set in persistent mode which could be adjusted for optimum homogeneity after demagnetization cooling of  $\text{PrNi}_5$ . The 7 T demagnetization magnet was well separated from the  $^3\text{He}$  sample.  $^3\text{He}$  temperatures below 0.5 mK were attained. Above 0.9 mK the temperature was measured with a  $^3\text{He}$  melting curve thermometer. The NMR frequency was nominally 3.89 MHz with a full width at half maximum linewidth of typically  $2/10^6$ . Approximately 30 h were allowed for the demagnetization field to settle before data were collected as the sample warmed at a rate of order  $10 \mu\text{K/h}$ .

Magnetization and resonance frequency measurements were made using pulsed NMR techniques with quadrature detection. The magnetization was determined from the initial amplitude of the free induction decay (FID) and was referenced to a proton NMR glycerol sample throughout the warm-up period. The experimental details are described elsewhere [9,12]. The resonance frequency was obtained from a Gaussian fit to the power spectrum of a complex fast Fourier transform of the FID

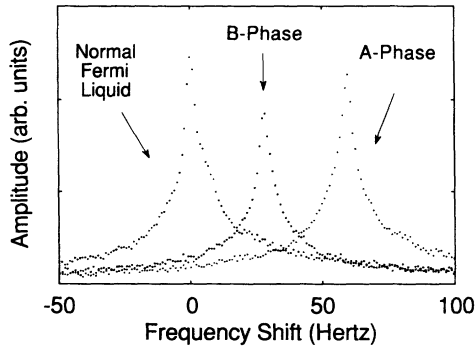


FIG. 1. Typical high-resolution power spectra from three phases of  $^3\text{He}$ . Frequency shifts are measured relative to 3.89 MHz.

allowing a precision of  $\pm 1/10^7$ . Both measurements were made using a phase alternating pulse sequence with  $20^\circ$  tipping pulses of  $105 \mu\text{s}$  duration. Typical power spectra for the different  $^3\text{He}$  phases are shown in Fig. 1. Since the linewidth is similar in all phases, we have inferred that inhomogeneity from superfluid textures was insignificant.

The Ginzburg-Landau expression for the free energy of the superfluid can be written [2] in terms of the superfluid order parameter,  $\Delta$ , and the 5 fourth-order invariants of the order parameter,  $I_i$ , in the form

$$F - F_N = \alpha\Delta^2 + \frac{1}{2}\Delta^4 \sum_{i=1}^5 \beta_i I_i, \quad (1)$$

where the  $\beta_i$  are phenomenological coefficients. Greaves [5] pointed out that the  $g$  shift in the Ginzburg-Landau regime could be used to determine the ratio of  $\beta_{345}$  to its weak coupling value,

$$\frac{\beta_{345}}{(\beta_{345})_{\text{wc}}} = \frac{18/7\zeta(3)}{1 + F\bar{\xi}} \frac{(C/\Delta C_B)^2}{(2\pi k_B T_c)^2} \frac{(\hbar\omega_{B\parallel})^2}{(1-t)^2} \frac{1-m}{g}, \quad (2)$$

TABLE I. Linear fit of  $g$  versus magnetization for various pressures,  $g = b_0 + b_1 m$ ; smoothed magnetization measurements,  $(1-m)/(1-t)$ , accurate to  $\pm 10\%$ ; longitudinal resonance frequency measurements at  $T_c$  in units of  $10^{10} \text{ Hz}^2$ ; and determination of  $\beta_{345}$  including magnetization strong coupling effects,  $(1-m)/(1-m)_{\text{wc}}$ , accurate to  $\pm 10\%$  for which smoothed values are shown in the last column.

$P$ (bar)	$b_0$ (ppm)	$b_1$ (ppm)	$\frac{1-m}{1-t}$	$\frac{(\nu_{B\parallel})^2}{1-t}$ $10^{10} \text{ Hz}^2$	$\frac{\beta_{345}}{(\beta_{345})_{\text{wc}}}$	$\frac{1-m}{(1-m)_{\text{wc}}}$
0.31	1.7	5.4	2.30	1.44	0.97	1.03
1.10	1.6	6.2	2.29	1.80	0.87	1.01
3.04	1.9	6.8	2.26	2.25	0.64	0.96
7.01	1.5	8.4	2.19	3.33	0.48	0.87
10.0	1.4	9.5	2.15	4.31	0.43	0.82
13.0	1.8	9.8	2.10	5.15	0.39	0.78
16.7	1.7	10.6	2.04	6.03	0.35	0.75
19.1	1.7	12.0	2.00	6.56	0.31	0.73
20.2	2.5	11.2	1.98	6.94	0.31	0.72
21.3	2.1	11.8	1.96	6.97	0.29	0.71
21.7	2.6	11.5	1.96	7.06	0.28	0.71

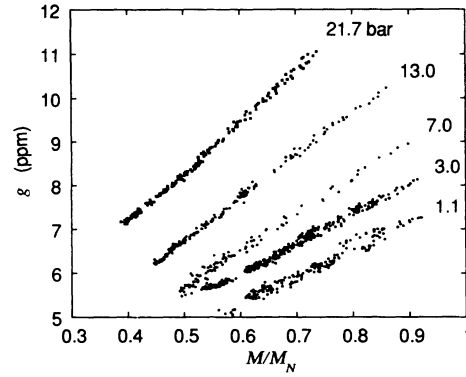


FIG. 2.  $g$  shift as a function of magnetization for 1.1, 3.0, 7.0, 13.0, and 21.7 bars.

where the notation  $\beta_{345} \equiv \beta_3 + \beta_4 + \beta_5$  is used. In our analysis, the critical temperature,  $T_c$ , and the heat capacity jump,  $\Delta C_B/C$ , are obtained from Greywall [13]. The Fermi liquid parameter  $F\bar{\xi}$  is taken from our previous work [12], and the dependence of the normalized magnetization,  $m = M/M_N$ , on temperature  $t = T/T_c$  is taken from our measurements.  $M_N$  is the normal fluid magnetization. In the Ginzburg-Landau regime, the  $B$ -phase longitudinal resonance frequency,  $\omega_{B\parallel}$ , can be determined from measurements of transverse frequency shifts in the  $A$  phase [14] as described below.

Our measurements of the  $g$  shift are plotted as a function of magnetization in Fig. 2 for pressures between 1.1 and 21.7 bars. It appears that the  $g$  shift is a linear function of  $m$  which we can represent as  $g = b_0 + b_1 m$ . We define frequency shifts relative to their value in the normal fluid at  $T_c$ , i.e.,  $g(T_c^+) = 0$ . The  $g$ -shift data in Fig. 2 and Table I are corrected for the shift in the local field owing to the  $^3\text{He}$  magnetization, increasing  $b_0$  and reducing  $b_1$  by  $4\pi M_N(1-D)/H_0$ , leaving  $g(T_c^-)$  unchanged.  $D$  is the demagnetization factor which for our geometry

we take to be  $1/2$ . These effects are small, less than  $6/10^7$ . Although there is no formal theoretical basis for the simple linear relation, microscopic theory gives predictions consistent with the experiments [9,15]. Calculations of the dependence of the  $g$  shift on magnetization [15] have been performed by one of us using weak coupling theory and the hydrodynamic equations given by Hasegawa [6] including terms in the free energy to all orders of magnetic field. These calculations [9,15] show that the  $g$  shift is approximately linear in magnetization, consistent with our data, with no explicit field dependence up to 0.3 T. The latter is confirmed by an experiment at 0.18 T and a pressure of 1.03 bars where we found results identical to those at 0.12 T [9]. The calculation uses three parameters,  $F_B^g$ ,  $F_A^g$ , and the dipole coefficient [2],  $g_D$ . Taking  $F_B^g$  from Ref. [12] we can extract  $F_A^g$ . We find  $F_A^g = -0.6 \pm 0.2$  at the pressure 1.1 bars. This value is in agreement with estimates from acoustic experiments,  $F_A^g = -0.9 \pm 0.3$  [16,17] and disagrees with those from field dependent susceptibility,  $F_A^g = +1.0 \pm 0.1$  [16].

In the accompanying Letter [18] we have also found that there are small but significant temperature dependent Larmor frequency shifts in the normal liquid [3] approximately 1 order of magnitude smaller than the  $g$  shift in the  $B$  phase. We believe that it is reasonable to assume that the phenomenon responsible does not change discontinuously at the superfluid transition. In fact, our frequency measurements in the  $A$  phase just below  $T_c$  are consistent with this assumption to an accuracy of  $1/10^7$ . Consequently we ascribe the shift in frequency in the  $B$  phase, extrapolated to  $T_c$ , to be the  $g$  shift. This leaves open the issue as to whether the temperature dependence, i.e., the magnetization dependence, of the  $g$  shift, as represented in Fig. 2 and Table I, have a contribution from the same source as that producing frequency shifts in the normal state. If we were to presume that the temperature dependent shifts in the normal state [3] extend into the superfluid, the data sets in Fig. 2 would remain approximately linear having the same value at  $T_c$  but with a larger intercept at zero magnetization. At 1.1 bars this would increase  $b_0$  from 1.6 to 2.9 ppm and our fit value of  $F_A^g$  to 0.6. Resolution of this question will require further work.

The linear dependence of the shift on magnetization, independent of magnetic field, allows us to obtain an accurate extrapolation of the zero field shift to  $T_c$ . It is in this Ginzburg-Landau limit that one can correctly apply Eq. (2). The analysis also requires measurements of the  $B$ -phase longitudinal resonance frequency in the Ginzburg-Landau limit which we describe next.

Based on the symmetries of the axial and isotropic states, the longitudinal resonance frequencies of the  $A$  and  $B$  phases are related in the Ginzburg-Landau regime by [1,2]

$$\left( \frac{\omega_{B\parallel}}{\omega_{A\parallel}} \right)^2 = \frac{5}{2} \left( \frac{\Delta_B}{\Delta_A} \right)^2 \frac{\chi_A}{\chi_B}, \quad (3)$$

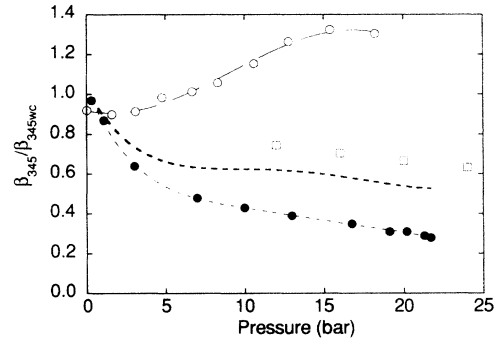


FIG. 3.  $\beta_{345}/(\beta_{345})_{wc}$  as a function of pressure. The open circles are taken from Tang *et al.* [10]; the solid circles from this work using Greaves's expression Eq. (2); and the squares are from the theory of Sauls and Serene [19]. The dashed line would be the result obtained if there were no strong coupling contribution to the magnetization, i.e.,  $(1-m)/(1-m)_{wc}$  were set to 1.0.

where  $\Delta_A$  and  $\Delta_B$  are the energy gaps for the  $A$  and  $B$  superfluid phases, respectively. This result is independent of strong coupling and quasiparticle interaction effects. As  $T$  approaches  $T_c$  the ratio of the gaps is given by the heat capacity jumps [10,13], and the susceptibility ratio,  $\chi_A/\chi_B$ , approaches 1.0. The axial state longitudinal resonance frequency is related to the transverse shift by [1,2]  $\omega_{A\perp}^2 = \omega_{A\parallel}^2 + \omega_0^2$ . Our measurements of  $(\omega_{A\perp})^2/(1-t)$ , consistent with those of others [14], combined with Eq. (3), give  $\omega_{B\parallel}$  near  $T_c$ .

With Greaves' expression, Eq. (2), we have determined the strong coupling corrections to  $\beta_{345}$  shown in Fig. 3. Our results are consistent with weak coupling values at low pressure decreasing at higher pressures qualitatively similar to the predictions of Sauls and Serene [19]. Tang *et al.* [10] have reported an analysis of magnetization and phase diagram measurements with results for  $\beta_{345}$  in disagreement with our work and the theory. In making use of Eq. (2) we explicitly include nontrivial strong coupling corrections to the magnetization neglected by Tang *et al.* [10]. The magnetization,  $1-m$ , is proportional to  $1-t$ , in the Ginzburg-Landau regime. We find that without our accuracy of  $\pm 10\%$  the coefficient  $(1-m)/(1-t)$  can be represented as a weak linear function of pressure for which smoothed values are presented in the table and are compared with the weak coupling expression,

$$\frac{(1-m)_{wc}}{1-t} = \frac{7\zeta(3)}{18} \frac{(\Delta C/C)_{wc}}{1+F_B^g} = \frac{2}{3} \frac{1}{1+F_B^g}. \quad (4)$$

Strong coupling is expected to reduce  $1-m$  as noted in previous theoretical estimates [20,21]. We have also analyzed magnetization data of Scholz [22] and find his results in agreement with ours to within our experimental error.

In summary, we have performed high-resolution NMR frequency measurements in the superfluid  $B$  phase, deter-

mining the pressure dependence of the  $g$  shift. In our analysis of  $g$ -shift measurements we have taken magnetization strong coupling effects into account and found qualitative agreement with theory for the pressure dependence of  $\beta_{345}$  in contrast with earlier work [10].

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[1] A. J. Leggett, *Rev. Mod. Phys.* **47**, 331 (1975).

[2] D. Vollhardt and P. Wölfle, *The Superfluid Phases of Helium* (Taylor and Francis, London, 1990).

[3] It has been generally accepted that there is no significant shift of the Larmor frequency in normal liquid  $^3\text{He}$ . The report in the preceding Letter indicates that this is not the case.

[4] D. D. Osheroff, W. J. Gully, R. C. Richardson, and D. M. Lee, *Phys. Rev. Lett.* **29**, 920 (1972).

[5] N. A. Greaves, *J. Phys. C* **9**, L181 (1976).

[6] Y. Hasegawa, *Prog. Theor. Phys.* **70**, 1141 (1983).

[7] H. Smith, W. F. Brinkman, and S. Englesberg, *Phys. Rev. B* **15**, 199 (1977).

[8] D. D. Osheroff, *Phys. Rev. Lett.* **33**, 1009 (1974).

[9] H. H. Hensley, G. F. Moores, M. R. Rand, T. M. Haard, J. B. Kycia, P. J. Hamot, Y. Lee, W. P. Halperin, and E.

V. Thuneberg, *J. Low Temp. Phys.* **89**, 505 (1992); H. H. Hensley, Ph.D. thesis, Northwestern University, 1992 (unpublished).

[10] Y. H. Tang, I. Hahn, H. M. Bozler, and C. M. Gould, *Phys. Rev. Lett.* **67**, 1775 (1991); C. M. Gould, *Physica (Amsterdam)* **178B**, 266 (1992).

[11] Stycast 1266, Emerson and Cuming, Inc.

[12] H. H. Hensley, Y. Lee, P. J. Hamot, T. Mizusaki, and W. P. Halperin, *J. Low Temp. Phys.* **90**, 149 (1993).

[13] D. S. Greywall, *Phys. Rev. B* **33**, 7520 (1986).

[14] M. R. Rand, H. H. Hensley, J. B. Kycia, T. M. Haard, Y. Lee, P. J. Hamot, and W. P. Halperin, *Physica B&C* (to be published); P. J. Hakonen *et al.*, *J. Low Temp. Phys.* **75**, 225 (1989).

[15] G. F. Moores, Ph.D. thesis, Northwestern University, 1993 (unpublished).

[16] R. S. Fishman and J. A. Sauls, *Phys. Rev. B* **33**, 2526 (1986).

[17] W. P. Halperin and E. Varoquaux, in *Helium Three*, edited by W. P. Halperin and L. Pitaevskii (North-Holland, Amsterdam, 1990), p. 353.

[18] T. M. Haard *et al.*, preceding Letter, *Phys. Rev. Lett.* **72**, 860 (1994).

[19] J. A. Sauls and J. W. Serene, *Phys. Rev. B* **24**, 183 (1981).

[20] J. W. Serene and D. Rainer, *Phys. Rev. B* **17**, 2901 (1978).

[21] Magnetization strong coupling [20] is often written in terms of a free energy coefficient  $g_z$  as  $g_z/(g_z)_{wc} = (\Delta C_{wc}/\Delta C_B)(1-m)/(1-m)_{wc}$ .

[22] H. N. Scholz, Ph.D. thesis, Ohio State University, 1981 (unpublished).