High-Resolution Measurements of the Larmor Frequency in Normal-Liquid ^{3}He

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Measurements have been made of the Larmor frequency of the normal ^{3}He Fermi liquid with resolution better than $1/10^7$. The Larmor frequency was observed to increase on increasing temperature from the superfluid transition to above 40 mK in a constant 0.12 T external magnetic field. The shift has a temperature dependence that is negative and approximately proportional to the inverse temperature. The magnitude is of order $1/10^6$ and depends on pressure in the investigated range 0.3 to 21.7 bars, decreasing substantially at the highest pressures.

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Helium-three is a classic example of a degenerate Fermi liquid at temperatures well below its Fermi temperature ($T_F \approx 1$ K) [1,2]. At sufficiently low temperatures, near 1 mK, a BCS-type superfluid condensate forms [3]. All low temperature properties of the ^{3}He Fermi liquid have the temperature dependence expected for an ideal noninteracting Fermi gas. For example, the specific heat is a linear function of the temperature and the magnetic susceptibility is temperature independent. The interactions between the Fermi quasiparticles are evident in the absolute magnitudes of these thermodynamic quantities and are parametrized in the phenomenological Fermi liquid theory of Landau [1]. In this Letter we report new, high-resolution, nuclear magnetic resonance measurements of the Larmor precession frequency that show significant deviation from the temperature dependence expected for a simple noninteracting Fermi gas. The nuclei of the ideal Fermi gas, characterized by a gyromagnetic ratio γ, should precess in a perfectly homogeneous field H_0 at the angular frequency given precisely by $\omega_0 \equiv \gamma H_0$. In normal-liquid ^{3}He the nuclear dipole interactions between atoms can, in principle, lead to frequency shifts [4,5], although we have not found a theoretical prediction consistent with our observations shown in Figs. 1 and 2.

In the following Letter [6] high-resolution NMR experiments in the ^{3}He-B superfluid phase are described. Observations are reported of small Larmor frequency shifts using the same highly homogeneous fields as in the present work. These are directly attributable to spontaneously broken spin-orbit symmetry [5], a characteristic of superfluid ^{3}He, but absent in the normal Fermi liquid with the possible exception of superfluid order parameter fluctuations. The B-phase shifts are of order $1/10^5$, an order of magnitude larger than those we have discovered in the normal Fermi liquid. Our results here are of immediate importance to that work, since a complete interpretation of the superfluid frequency shifts may require an understanding of the phenomenon responsible for the Larmor frequency shifts of the normal Fermi liquid.

Our apparatus was designed to allow high-resolution pulsed NMR Larmor frequency measurements in ^{3}He at temperatures well below 1 mK. The sample, a 0.5 cm long by 0.5 cm diameter cylinder of ^{3}He, was held in the

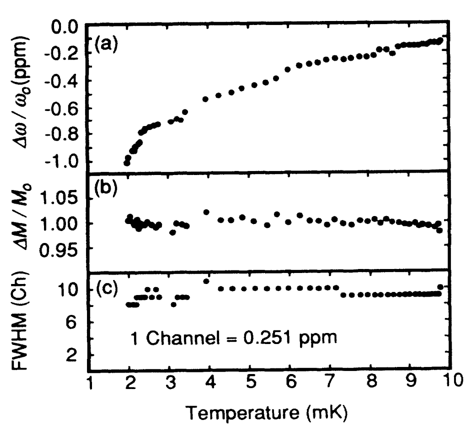

FIG. 1. Temperature dependence of NMR measurements for normal-fluid ^{3}He at 13.2 bars and 0.12 T. (a) Larmor frequency in parts per million (ppm) measured relative to its value at high temperatures. (b) Magnetization normalized to the value at T_c. (c) Linewidth measured in FFT channels.

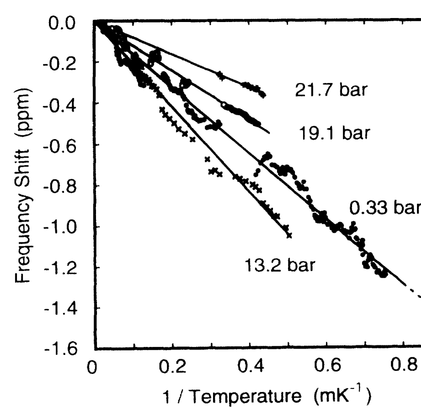

FIG. 2. Typical single parameter fits of the normal-fluid frequency shifts to inverse temperature at several pressures, as described in the text. The lines extend to $1/T_c$ except for the 0.33 bar data.

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constant 0.12 T magnetic field of a highly homogeneous superconducting magnet. This magnet was equipped with superconducting shim coils that permitted adjustment of the field homogeneity immediately after the sample was cooled by adiabatic nuclear demagnetization of $PrNi_5$. The 7 T demagnetization magnet was well separated from the sample. The residual demagnetization field was varied from 0.015 to 0.06 T in order to study effects of different warming rates. In all cases the homogeneity of the magnetic field over the sample volume could be adjusted with superconducting shim coils to be better than $2/10^6$ as defined by the NMR full width at half maximum (FWHM); see Fig. 1. The NMR coil was isolated from the sample cell and thermally anchored to the mixing chamber of the dilution refrigerator. The coil, 1.25 cm long and 1 cm in diameter, was wound with compensation and was measured to have inhomogeneity of the rf field over the sample limited to 1% [7]. The ^{3}He sample was connected through an opening 0.125 cm diameter and 0.8 cm long to a standpipe of larger dimension leading to the main ^{3}He reservoir and heat exchanger, 10 cm distant. The sample cell and standpipe were constructed with epoxy [8]. Temperature was determined above 1 mK with a ^{3}He melting curve thermometer (MCT) located on the heat exchanger.

Measurements were made at the nominal NMR frequency of 3.89 MHz using pulsed NMR with quadrature phase sensitive detection techniques. NMR excitation with 20° tipping pulses, 105 μs in duration, was used. Each data point was an average of 4 acquisitions with phase alternated excitation. The Larmor frequency was taken from a Gaussian fit to the upper 25% of the power spectrum obtained from a complex fast Fourier transform of the NMR free induction decay signal. With this technique the resonant frequency was resolved to better than $\pm 1/10^7$. Magnetization was determined from the amplitude of the free induction decay signal. After demagnetization cooling, data were accumulated continuously throughout the warm-up period using an automated NMR acquisition and control system.

Various environmental factors, including flux creep and drift in the NMR and demagnetization magnets, can contribute to systematic error in the frequency measurement. Such effects have been investigated and they do not contribute to the present results. ^{3}He magnetization measurements were referenced to the proton NMR signal of a glycerol sample held at room temperature. Both proton and ^{3}He NMR measurements were obtained throughout the data acquisition period using the same spectrometer with multiplexed coupling circuitry for the different nuclei [7]. Typical ^{3}He spectra are displayed in the accompanying Letter [6]. Frequency, magnetization, and linewidth data are shown in Fig. 1 at a pressure of 13.2 bars.

Experiments were conducted at pressures between 0.33 and 21.7 bars and are summarized in Fig. 2. These data indicate that there is a temperature dependent shift in the Larmor frequency of ^{3}He approximatley proportional to the inverse of the temperature. In these experiments it is not possible to identify an absolute frequency reference. For convenience, we have chosen the value extrapolated to high temperatures as an *ad hoc* reference as displayed in Fig. 2. With this convention the shifts we have observed are negative and can be fit to the form c/T, as shown in Fig. 2, with the pressure dependence of c given in Fig. 3(a). The effect is largest near 10 bars, after which it decreases with increasing pressure. In Fig. 3(b) we show the frequency shift determined at T_c as a function of pressure. In superfluid ^{3}He the observed frequency shifts are positive and depend on the pulse excitation tip angle. We performed measurements as a function of pulse excitation for tip angles between 10° and 180° at a pressure of 1 bar and at a temperature just above T_c. The frequency shift in the normal fluid was found to be independent of tip angle to an accuracy of $1/10^7$.

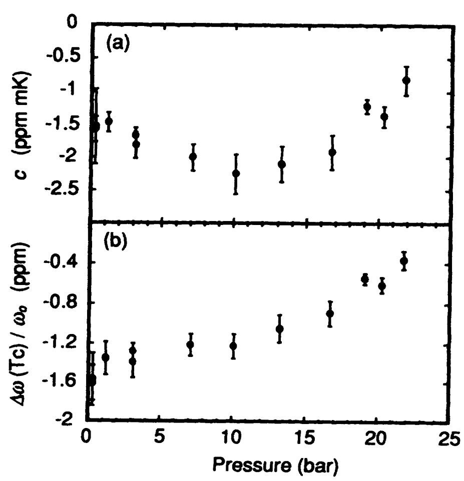

FIG. 3. (a) Pressure dependence of c, the slopes of the graphs presented in Fig. 2. (b) Frequency shift at T_c.

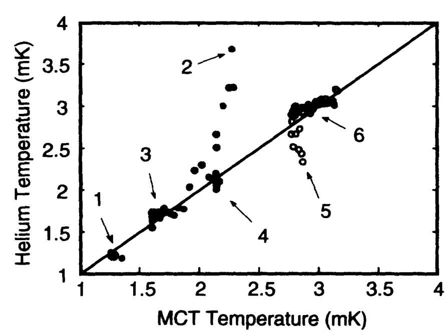

FIG. 4. Two heat pulse experiments. The temperature of the ^{3}He sample was inferred from the measured frequency shift. The MCT measures the temperature of the heat exchanger. The chronology follows the sequence 1 to 6 as described in the text, where the time at each point expressed in minutes from point 1 is 0, 8, 96, 956, 960, and 1016.

In order to demonstrate unambiguously that the frequency shifts we have observed are intrinsic to the helium sample we have performed several heat pulse experiments shown in Fig. 4. The helium temperature was calculated from the measured frequency shift using the results of Fig. 2. The abscissa is the temperature of the heat exchanger determined with the melting curve thermometer. The chronology following two different types of heating pulses is shown in sequence from 1 to 6 in this figure. First, at point 1, heat was applied directly to the sample using a small positive pressure pulse that injected warm helium into the cell without significantly changing its pressure. The temperature of the helium increased suddenly to the point 2 and then recovered at 3. For comparison, at point 4, a pulse of heat was applied electrically to a heater attached to the heat exchanger. The temperature of the heat exchanger increased suddenly, out of equilibrium with the helium NMR sample, reaching point 5, later recovering to equilibrium at point 6. In these two experiments, disequilibrium is a result of the Kapitza thermal resistance between sample and heat exchanger, allowing us to demonstrate that the frequency shifts are intrinsic to the helium sample and are not associated with temperature change of the sample cell walls, the heat exchanger, or other parts of the demagnetization apparatus.

We have made careful studies of the A- and B-phase superfluid frequency shifts but do not have data at low enough temperature and sufficient sensitivity to observe a $1/T$ divergence. Consequently, it is not clear whether this temperature dependence persists below T_c.

On rather general grounds Leggett [5] has shown that there may be frequency shifts, $\omega_\perp - \gamma H_0$, of transverse NMR attributable to the nuclear dipole-dipole interaction, H_D,

$$\omega_\perp^2 = (\gamma H_0)^2 + Q_\perp/\chi_\perp , \tag{1}$$

where $\chi_\perp$ is the transverse susceptibility and $Q_\perp$ is

$$Q_\perp = -\frac{1}{\hbar^2}\langle [M_\perp,[M_\perp,H_D]]\rangle . \tag{2}$$

Leggett has pointed out that the sum of the components of Q is inherently positive since $\langle H_D\rangle$ is negative, and that it can also be shown that $Q_\perp \geq 0$, implying that $\omega_\perp$ is positive. Furthermore, there should be a contribution to the linewidth of the same size as the shift [4,5], which we do not observe, such as shown in the data in Fig. 1. Homer and Richards [9] found a small positive shift in solid helium at high temperatures, $T \approx 2$ K, consistent with theory [4] involving the nonsecular terms of the dipolar interaction. From Eqs. (1) and (2) it can be estimated [4,5] that the dipolar shifts in a liquid or gas have magnitude given approximately by $gD^2/\omega_\perp$. This is of second order in the strength of the dipole interaction, g_D, where $g_D \equiv \mu_n/a^3$, μ_n is the nuclear magnetic moment, and a is the average atomic spacing. On this basis the dipolar shift in ^{3}He would be approximately $2/10^6$ at low pressure [10]. The theory does not predict a temperature dependence to the shift although calculations have not been performed in the low temperature limit of the degenerate Fermi liquid.

Polarization of the ^{3}He can in principle shift and broaden the NMR line. Such a contribution to the local field is $4\pi\chi(1-D)$ where χ is the ^{3}H susceptibility and D the demagnetization coefficient, $\approx \frac{1}{2}$ for our geometry, giving a shift of $2/10^7$ at low pressure increasing to $6/10^7$ at 21 bars [10]. The magnetization, and consequently the frequency shift, is temperature independent in this range as can be seen in our measurements in Fig. 1. This effect cannot account for our observations. The NMR sample contains a small amount of solid ^{3}He adsorbed to the wall which is expected to exhibit a Curie susceptibility. A magnetic shielding effect due to this solid layer will produce a frequency shift that is negative, but that is also second order in the susceptibility, resulting in a frequency shift proportional to the square of the inverse temperature, inconsistent with our results. Moreover, we estimate the shift at 1 mK to be less than 1 part in 10^{14}. Similarly, if we consider all of the protons in the epoxy cell they will produce a shift less than 1 part in 10^9. It seems that these effects cannot account for our observations either.

Another possibility might be interaction of the ^{3}He liquid with the solid at the walls inducing a polarization in the liquid. However, this is not evident in our susceptibility measurements. Furthermore, the size of a static polarized layer of fluid can be estimated to be less than the diffusion distance of a quasiparticle during its relaxation time $\approx 10^{-7}$ s. This distance is less than 15 μm —which we estimate to be much too small to affect our measurements. It should be possible to check how significant such effects might be by introducing ^{4}He into the cell to coat the walls, thereby changing the nature of the surface interactions between the liquid ^{3}He and the walls.

One might expect a frequency shift to arise from superfluid order parameter fluctuations. In general such fluctuations can be expected to have a temperature dependence [11] proportional to $(T/T_c - 1)^{-1/2}$ which we believe is inconsistent with our results. Furthermore, the superfluid phases have positive frequency shifts so one might expect fluctuation contributions to have the same sign, but this is opposite to what we observe. Finally, while fluctuations into the isotropic state will decrease the static susceptibility producing a negative shift, it is likely too small to be seen and is not evident in our direct measurements of the susceptibility. Consequently, it is unlikely that superfluid fluctuations contribute to the frequency shifts in the normal liquid.

In conclusion, we have discovered a Larmor frequency shift in the normal phase of ^{3}He which has a characteristic $1/T$ dependence and is negative in sign, becoming less so at higher pressures. After examining a number of possibilities we have not been able to identify a theoretical basis for these observations.

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