Anomalous NMR Frequency Shift in the Low-Field Phase of Solid ^{3}He

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We present results of the first orientation-dependent study of the spin dynamics of the low-field spin-ordered phase of solid ^{3}He, the U2D2 phase. The results of a hydrodynamical treatment of the spin dynamics describe the observed behavior exceedingly well; however, only if a field- and orientation-independent negative shift, not provided by the theory, is properly incorporated into the equations. This shift does not arise from a lattice distortion as had been expected, and its measured temperature dependence suggests an unknown nonhydrodynamical origin.

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The low-field spin-ordered phase of bcc solid ^{3}He is antiferromagnetic with a U2D2 structure, consisting of planes of uniformly aligned spins whose direction alternates in a sequence of two planes up and two planes down, etc. [1]. The domain direction, represented by a unit vector $\boldsymbol{l}$ normal to these ferromagnetic planes, lies along any of the three principal axes of the cubic lattice. In a single crystal, there usually exist three orthogonal domains. For each domain, there are two NMR resonant modes whose frequencies ν_i can be described by a hydrodynamical treatment of the spin dynamics:

$$(\nu_i^{\pm})^2 = \tfrac{1}{2}\{\nu_L^2 + \Omega_0^2 \pm [(\nu_L^2 - \Omega_0^2)^2 + 4\nu_L^2 \Omega_0^2 \cos^2\Gamma_i]^{1/2}\}, \quad (1)$$

where ν_L is the Larmor frequency, Ω_0 the temperature-dependent antiferromagnetic resonance frequency, $\cos\Gamma_i = \boldsymbol{l}_i \cdot \mathbf{h}$ with $\mathbf{h}$ the unit vector along the magnetic field $\mathbf{H}$, and $\sum_i \cos^2\Gamma_i = 1$ due to the orthogonality of the domains. We refer to Eq. (1), presented by Osheroff, Cross, and Fisher, as the OCF equations [2]. See the fine review article by Cross and Fisher for a full description of this approach [1]. Here Ω_0 varies from about 525 kHz at T_N to 825 kHz at $T=0$. The plus sign in Eq. (1) denotes the upper, or high-frequency, mode, which we have studied in this experiment. It is clear from Eq. (1) that ν_i^+ shifts above ν_L, and $\nu_i^+ = \nu_L$ only when $\cos^2\Gamma_i = 0$ for $\nu_L > \Omega_0$. We refer to this positive shift as the OCF shift, being 156 kHz at $\nu_L = 2.1$ MHz and $T=0$.

The OCF equations agree well with experiments; however, they have never been tested rigorously due to an inability to vary the field-sample orientation. Furthermore, OCF and other researchers [3] have occasionally observed that ν_i^+ drops below ν_L when $\mathbf{H}$ is applied nearly normal to $\boldsymbol{l}_i$ so that $\cos^2\Gamma_i \approx 0$. This anomalous negative shift Δ (of the order of 1 kHz) is inconsistent with the results [Eq. (1)] of hydrodynamical spin dynamics applied to the tetragonal U2D2 structure, owing to the isotropy of the dipole energy within the ferromagnetic planes.

We have speculated that Δ might result from a lattice distortion within the ferromagnetic planes associated with spin ordering, in which case one would expect to see a modulation of the minimum resonant frequency above and below ν_L as $\mathbf{H}$ is rotated about $\boldsymbol{l}$ in the plane [4]. OCF claimed, based on their measurements of the lower-frequency mode ν_i^-, that any such distortions had to be negligible. The inability of those authors to reorient their field, however, could have allowed such distortions to go undetected for the specific field orientations they studied. In this Letter we present the results of extensive studies of Δ involving continuous rotation of the static field, including both the temperature and field-orientation dependences.

To continuously rotate the field, we attached a two-axis superconducting magnet [5] directly to our vacuum can using a worm gear assembly designed for telescope positioning. This was coupled to a further gear reduction assembly outside the cryostat, and, by rotating only in one direction to avoid backlash, we could rotate the field about the vertical with a resolution and reproducibility of a few hundredths of a degree. The ultralow-temperature apparatus used in this work has been described previously [6]. The NMR sample cell was a cylindrical chamber 2.5 mm in length and 2.3 mm in diameter, whose bottom was closed with a piece of filter paper to insure good thermal contact with the liquid. The single-crystal samples were grown by heat-pulse nucleation on a 25-μm platinum-tungsten wire stretched across the top of the chamber. After nucleation, the samples were partially melted until they fell from the wire to the bottom of the chamber, insuring a strain-free environment. The NMR resonance signal was measured using a conventional cw spectrometer.

To determine the magnetic field across our samples, we measured the Larmor frequency ν_L ($\sim$2.1 MHz at $H = 650$ G) by melting the solid first and then raising the liquid temperature to above the superfluid A transition by adding heat to the liquid [7]. The Earth's magnetic field caused a nearly sinusoidal variation of $\sim$600 Hz in ν_L when the field was rotated in the horizontal plane. Although our NMR magnet had a homogeneity of 10 ppm over a cubic centimeter, local magnetization at low temperatures degraded the field uniformity, producing a

FWHM of 200 Hz. More important was the uncertainty arising from the fact that our solid samples did not fill the NMR chamber, and hence the local field on the solid was not the same as the average field seen by the liquid. We could partially correct for this effect by studying the solid signal as a function of sample size. Because of the excellent stability of the magnet (field decay rate <0.1 Hz/h) and the short duration of each measurement, we were able to neglect the field decay. The uncertainty in the field caused by the magnetization [8] of the solid sample itself is estimated to be only about 20 Hz. In the end, we estimate an overall uncertainty in ν_L to be about 50 Hz at $\nu_L \approx 2$ MHz.

To measure Δ, it is necessary to work in the vicinity of $\cos^2\Gamma_i = 0$ to reduce the effect of the much larger OCF shift. For practical purposes [9], we have limited the field to the horizontal plane, i.e., its polar angle $\theta_H = 90°$, and varied only the azimuthal angle ϕ_H. We then simply rotated the field until the resonant frequency was a minimum. For an arbitrary crystal, there are two minimization angles ϕ_{i1} and ϕ_{i2} (180° apart) for each domain. Clearly $\phi_{i1,i2} = \phi_i \pm \pi/2$, where ϕ_i is the azimuthal angle of the ith domain. From Eq. (1), for Γ_i close to $\pi/2$, ν_i^+ can be expanded about ϕ_{i1} or ϕ_{i2} and shown to depend quadratically on rotation angle:

$$\nu_i^+ = \nu_L\{\nu_L \Omega_0^2(\sin^2\theta_i)/2(\nu_L^2 - \Omega_0^2)\}(\phi_H - \phi_{i1})^2, \quad (2)$$

where θ_i is the polar angle of the ith domain. All angles defined here are shown in the inset of Fig. 1(a) for clarity.

A typical parabolic minimization is shown in Fig. 1(a) with $\nu_L = 2.1$ MHz. We notice that within a finite range of ϕ_H, the resonance occurred below ν_L, revealing a negative shift inconsistent with Eq. (2). It is logical to assume that the minimum resonant frequency occurs precisely when $\Gamma_i = 90°$, and that the OCF shift from Eq. (2) completely vanishes at that point. Hence the *depth* of the parabola is a measure of the negative frequency shift Δ. [In fact, the shift above the minimum can be well accounted for by an OCF term (shown by the solid line), if Eq. (2) is properly modified.] The minimizations for the other two domains of the same crystal yielded comparable values of Δ within an uncertainty of 50 Hz. A number of different crystals were tested and no significant variation in its magnitude was observed, provided that Δ was measured at roughly the same temperature. We also varied the magnetic field from 308 to 1230 G ($\nu_L = 1$–4 MHz), and found that Δ was constant to within 50 Hz.

Our ability to decouple from the OCF shift allowed us to measure the temperature dependence of Δ. As shown in Fig. 1(b), Δ increases below T_N, reaching a maximum at about $0.7T_N$, and then decreases and appears to approach zero as $T \to 0$. In measuring $\Delta(T)$, we first rotated the magnetic field until the frequency of one domain was minimized, and then varied the temperature. We found that throughout the measurement, the frequency

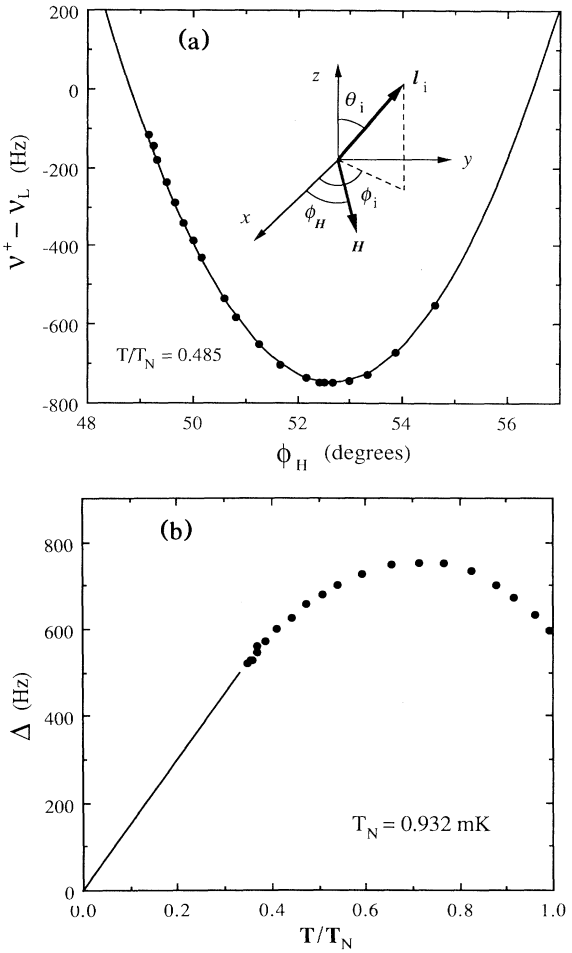

FIG. 1. (a) The variation of the NMR frequency minus the Larmor frequency as a function of angle as the horizontal static magnetic field is rotated about the vertical, showing the broad parabolic minimum as described by Eq. (2). (b) The temperature dependence of the negative frequency shift.

remained minimized. We recorded all three resonant frequencies to calculate T from Ω_0 using Eq. (1) with the negative shift neglected. We have obtained $\Delta(T)$ in several crystals, and found very consistent results. Offsets of the order of 50 Hz were observed among different sets of data, but the shapes of the curves were almost identical.

From our data on different domains of a single crystal as well as various domains in several different crystals, we believe that Δ is independent of the field orientation within the ferromagnetic plane. To investigate further, we attempted to grow a single crystal with one of its domains oriented exactly parallel to the vertical direction so that the domain would remain perpendicular to **H** when the magnetic field was rotated in the horizontal plane, thus allowing a continuous measurement of Δ from a single domain. In Fig. 2(a), we show the results for

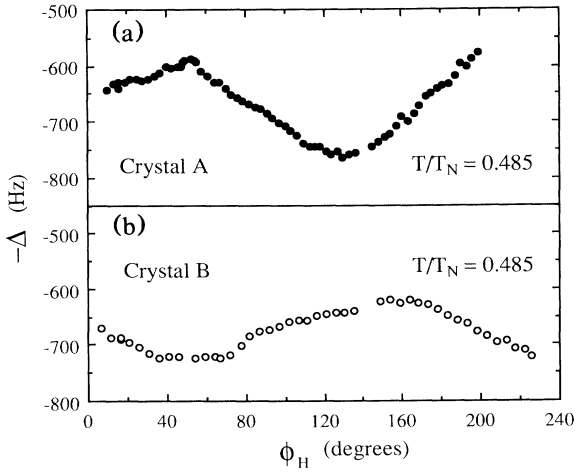

FIG. 2. (a) The total frequency shift, $\nu - \nu_L$ (●), as the static field is rotated in the horizontal plane for a domain oriented almost vertically. The variation as a function of angle sets model-independent limits on any possible distortion of the crystal in the ferromagnetic plane. (b) The variation of $\nu - \nu_L$ (○) with field orientation, corrected to eliminate the positive shift from the OCF equations which resulted from a slight deviation of the domain axis from the vertical. See text.

such a domain (of crystal A). We believe the variation in Δ was caused by a small angle (on the order of 1°) between the domain direction and the vertical and could be attributed to the OCF shift in Eq. (1). Although we were unable to determine the exact domain orientation before the crystal melted to correctly subtract the OCF shift, the fact that the resonant frequency remained below ν_L suggests that it was *not* caused by a lattice distortion, since such a distortion would produce a shift oscillating above and below the Larmor frequency [4]. Such an oscillation could not be masked by coupling to the OCF shift, which is always positive.

In Fig. 2(b), the negative shift of a near vertical domain in a second crystal (B) is shown for which we have corrected for the positive OCF shift by determining its precise domain direction. The precise orientation could only be calculated from the NMR frequencies using the OCF equations themselves. The presence of the negative shift, however, introduces large errors in the determination of the crystal orientation, therefore making analysis virtually impossible without first modifying the OCF equations. Hence we have incorporated Δ into the OCF equations by assuming that all three observed resonant frequencies must be shifted upward by some quantity $\Delta(\Gamma_i)$. Based on the above measurements on various domains and crystals, we first assumed that $\Delta(\Gamma_i) = \Delta'$ (as we shall discuss later, this gives a best fit). We then obtain

$$(\nu_i^+ + \Delta')^2 = \tfrac{1}{2}\{\nu_L^2 + \Omega^2 + [(\nu_L^2 - \Omega^2)^2 + 4\nu_L^2\Omega^2\cos^2\Gamma_i]^{1/2}\}, \quad (3)$$

where Ω is a parameter obtained from the condition that $\sum_i \cos^2\Gamma_i = 1$ for a given Δ'. If $\Delta' = 0$ then $\Omega = \Omega_0$ and the OCF equations are recovered. We had to allow Ω to vary, because of expected temperature changes due to eddy-current heating associated with the field rotation.

To determine the orientation $\boldsymbol{l}_c = (\theta_c, \phi_c)$ for the near vertical domain c of crystal B, we first calculated those of domains a and b, i.e., $\boldsymbol{l}_a = (\theta_a, \phi_a)$ and $\boldsymbol{l}_b = (\theta_b, \phi_b)$, and then made use of their orthogonality to obtain $\boldsymbol{l}_c = \boldsymbol{l}_a \times \boldsymbol{l}_b$. Using Eq. (3) and the relation

$$\cos\Gamma_i = \cos\theta_H \cos\theta_i + \sin\theta_H \sin\theta_i \cos(\phi_H - \phi_i)$$
$$= \sin\theta_i \cos(\phi_H - \phi_i),$$

we determined $\boldsymbol{l}_a$ and $\boldsymbol{l}_b$ by fitting the actual measurements of ν_a^+ and ν_b^+ for $6° < \phi_H < 225°$. In Fig. 3, we show the best fit with $\Delta' = 700$ Hz, $(\theta_a, \phi_a) = (89.69°, 142.66°)$, and $(\theta_b, \phi_b) = (86.25°, 232.68°)$. The average differences between the measured and predicted frequencies, also shown, are equivalent to errors in angle of about 0.1°. We note that the three maxima of the deviation do not coincide with the angles where Eq. (3) has the maximum angular sensitivity. Our value of $\Delta' = 700$ Hz is consistent with the operating temperature $T/T_N = 0.49$ and the value of $\Delta(T)$ in Fig. 1(b). From the above parameters we find that $(\theta_c, \phi_c) = (3.76°, 47.86°)$. Using $\boldsymbol{l}_c$, we eliminated the OCF shift for domain c and then obtained the negative shift as shown in Fig. 2(b). Note that the average value of Δ agrees with the assumed value for

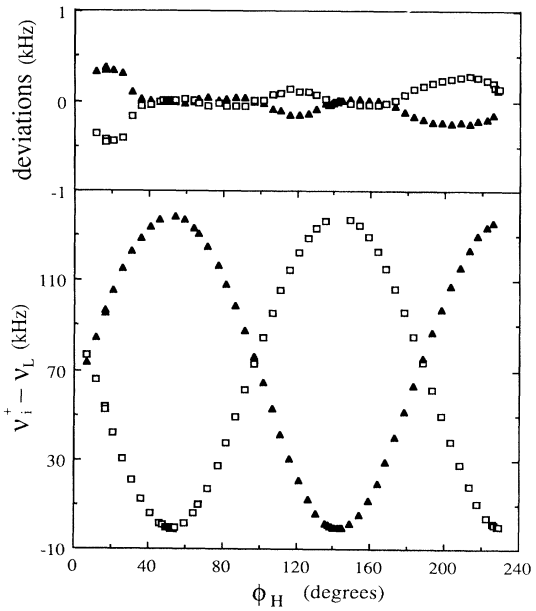

FIG. 3. The NMR frequencies of domains a (□) and b (▲) in crystal B [Fig. 2(b)] which were almost in the horizontal plane. By fitting the OCF equations to these data the orientation of crystal B was determined. Deviations of the fit to the data are shown above the main figure.

Δ' and its residual variation is well within experimental uncertainty of 50 Hz. We repeated the fit assuming that $\Delta(\Gamma_i) = 700 \sin^2(\Gamma_i)$ Hz. The deviations from this fit were nearly twice as large as with a constant Δ', and after subtracting off the OCF shift, a 600-Hz variation as a function of rotation remained for domain c, totally inconsistent with all our other measurements. Since θ_c was very small, Δ was measured effectively in the ferromagnetic planes. We thus conclude that, within the experimental accuracy, Δ is independent of the field orientation in the ferromagnetic planes.

The constancy of Δ is inconsistent with a simple lattice distortion, which would produce a $\sin(2\phi_H)$ variation. In fact, from the uncertainty of Δ in Fig. 2(b), we can now place smaller but far more conservative limits on any anisotropy of the dipole energy in the ferromagnetic planes: $(\lambda_1 - \lambda_2)/(\lambda_1 - \lambda_3) < 5 \times 10^{-4}$, where λ_i is the ith component of the dipole energy tensor [2].

The nonmonotonic temperature dependence of Δ seen in Fig. 1(b) is somewhat surprising. We speculate that at low temperatures Δ drops because the number of thermal magnons decreases, as one might expect for a nonhydrodynamic effect. We especially note that the maximum Δ occurs when $\omega_L \tau \approx 1$, where τ is the mean time for magnon umklapp scatterings [10]. However, the presence of Δ in the absence of any other frequency shift and its overall independence on field orientation are uncharacteristic of nonhydrodynamic effects. In any event, the lack of variation of Δ from sample to sample shows that it is an intrinsic property of the U2D2 solid. At the same time, after incorporating an independent $\Delta(T)$ into the OCF formalism, we find impressively good agreement between our measured frequencies and those predicted by the modified OCF equations. We hope the highly characterized behavior of this magnetically ordered system will pose a challenge to theorists in the field.

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