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Nuclear Antiferromagnetic Resonance in Solid ^{3}He

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Detailed measurements of the low-field antiferromagnetic resonance spectrum of spin-ordered bcc ^{3}He exhibit large shifts from the Larmor frequency, with a zero-field resonant frequency near zero temperature of $\Omega_0/2\pi \simeq 825$ kHz. Analysis of the spectrum leads to stringent constraints on possible sublattice structures. The temperature dependence of Ω_0 shows low-temperature behavior expected from spin-wave theory, and indicates a first-order transition at 1.03 mK.

The magnetic interaction between nuclear spins is extremely small, so that in most solids nuclear ordering occurs in the submicrodegree temperature range. In solid ^{3}He near melting pressures, however, the actual exchange of atoms between nearby sites leads to a much larger "exchange interaction" of order $J \simeq -0.75$ mK. The simplest description of this process, including only pair exchange, led to the expectation that the ^{3}He spins should order antiferromagnetically at $T_N \simeq 2.0$ mK. Measurements of the melting pressure showed, however, a substantial solid entropy to much lower temperatures,[1] and it is now clear that a more complicated exchange Hamiltonian must describe the spins, although its exact form remains unknown.[2] More recent thermodynamic measurements have determined $T_N \simeq 1.1$ mK, and show a nearly discontinuous drop in solid entropy at the transition,[3-5] along with a sharp decrease in the magnetic susceptibility.[6] Our work, however, provides the first information on the microscopic nature of the ordered state through detailed continuous-wave NMR studies.

Our compression cell was similar to that described by Osheroff, Richardson, and Lee,[1] but contained a heat exchanger with 100-m^2 surface area which attached to a copper nuclear demagnetization device. The cell also contained a capacitance pressure transducer, a displacement capacitor to monitor the cell volume, a pulsed platinum NMR thermometer, and an epoxy ^{3}He NMR insert. A ^{3}He NMR coil was cast into the epoxy insert near the bottom, and a tiny heater was contained in the 0.64-cm-diam open space probed by the NMR coil. The pressure transducer and thermometer were calibrated against proper-

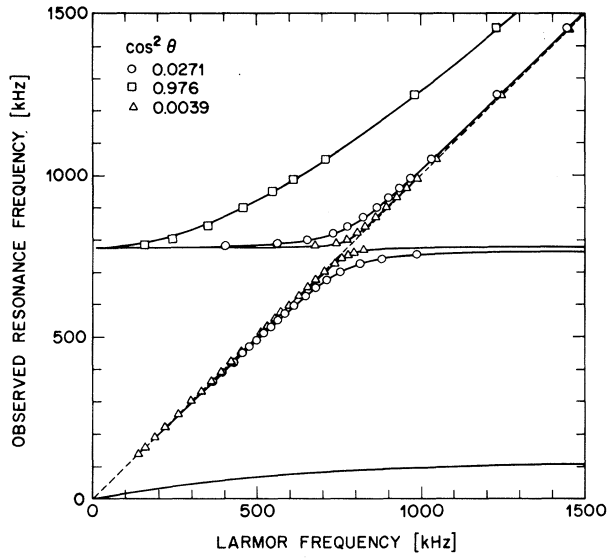

FIG. 1. NMR spectra for a single crystal of solid ^{3}He containing three domains. Lower and upper frequency modes of the same domain are denoted by the same symbol. The dashed line is the Larmor resonance line. The solid lines are theoretical curves generated from Eq. (1) with $\Omega_0/2\pi = 777.7$ kHz and values of $\cos^2\theta$ chosen to obtain a best fit to the data. For the upper mode, linewidths were typically 300 Hz in high fields, and 1.5 kHz at zero field. For the lower mode the linewidth broadened rapidly from about 300 Hz to about 2 kHz as the resonance frequency was decreased to slightly below one-half the value of the frequency of the upper mode in zero field. We believe this broadening results from a three-magnon process.

ties of the superfluid phases with use of $T_c = 2.752$ mK, and $P_{\rm melt}(B') - P_{\rm melt}(A) = 20.0$ mbar.[7]

Single crystals of solid ^{3}He were grown by first precooling the cell to ~0.5 mK and then raising the cell pressure 5–10 mbar *above* the melting pressure. Solid growth was initiated by applying a ~1-erg heat pulse to the heater wire, a process which nucleated ten to twenty small crystals. After about 15 min all but ~0.002 cm^3 (about 10%) of the initial solid was melted by decompression, leaving (in most cases) a single seed crystal. Finally, a large (0.1 cm^3) crystal was grown at constant temperature. Such crystals reached thermal equilibrium in about 10 min below $0.7T_N$, and in about 90 min above $0.9T_N$. All measurements discussed here were taken in thermal equilibrium.

In Fig. 1 we show a typical NMR spectrum, as a function of applied field, H, for a single, randomly oriented crystal at 0.487 mK. The spectrum was measured with a Rollin spectrometer by sweeping the field at a rate of 0.1 Oe/sec to avoid eddy-current heating and to limit uncertainties in H (typically about 0.1 Oe even at fields below 5 Oe). Three resonances were observed in high fields. These, we will show, originate from domains within the sublattice structure. As the field was lowered the frequencies of the resonances approached a single value, Ω_0, equal to 778 kHz at this temperature. This mode was also observed in a field of less than 1 Oe by sweeping the frequency. Associated with each high-frequency mode, there is a corresponding low-frequency mode, with a frequency that saturates at high fields, and a frequency that approaches zero as the field decreases to zero. The correspondence between high- and low-frequency peaks was generally clear from a comparison of the peak heights (indicating different amounts of the three domain orientations). For this crystal only two of the three expected low-frequency branches were accessible experimentally, the third saturating at too low a frequency to be observed.

The solid lines in Fig. 1 are a theoretical fit given by

$$\omega^2 = \tfrac{1}{2}\{\omega_L^2 + \Omega_0^2 \pm [(\omega_L^2 - \Omega_0^2)^2 + 4\omega_L^2\Omega_0^2\cos^2\theta]^{1/2}\} \quad (1)$$

with use of the measured Ω_0 and with three angles θ chosen to give the best fit. Here ω_L is the Larmor frequency, γH. Equation (1) follows from a dynamical description of the NMR described below, where θ is the angle between the field and a fixed direction in each domain. The ± in Eq. (1) gives the upper/lower mode, so that for each pair of branches there is a single parameter. Except for a deviation of the highest-frequency curve near $\gamma H = 0$, which is probably a result of slight heating during the field sweep, the fit is excellent.

In addition to data of the sort shown in Fig. 1, we have followed the high-frequency mode for domains oriented so that $\cos^2\theta \approx 1$ to higher fields. At about 0.5 mK the "Pythagorean relationship," $\omega^2 = (\gamma H)^2 + \Omega_0^2$, is obeyed for such domains to better than 0.5% up to fields of 770 Oe. At T_N there is a progressive deviation from the Pythagorean relation. In fields from 0.1 to 2 kOe the empirical relationship $\Omega_0^2(H)/\Omega_0^2(H=0) = 1 + 0.052(H/1\ \text{kOe})^2$ is obeyed to within 2%. Also in high fields, we occasionally observed a small (~1 kHz) negative frequency shift in the high-frequency mode. The origin of this shift is not understood, but it was nearly independent of temperature and field, and therefore is not believed to result directly

from any anisotropy in the dipolar energy.

In Fig. 2 we show the temperature dependence of the characteristic frequency Ω_0. The data were obtained at 284 Oe by measuring the resonant frequency of a carefully selected crystal which exhibited a yearly maximum shift. We then inferred Ω_0^2 using a simplified form of Eq. (1) valid for small θ. The angle θ was determined by measuring Ω_0 directly in zero field at ~0.5 mK. Values of Ω_0^2 should be accurate to about 2% near T_N, and to better than 1% below 0.7 mK. At low temperatures Ω_0^2 varies linearly with $(T/T_N)^2$, as we expect from spin-wave theory. At T_N, Ω_0^2 has a nonzero value, indicating a first-order transition. In related experiments[8] we have measured the ^{3}He melting pressure from 0.35 mK to slightly above T_N. The low-temperature melting pressure ($T<0.7$ mK) varies as T^4, again as is expected from spin-wave theory. At $T_N = 1.03 \pm 0.01$ mK we find a discontinuous change in solid entropy of $0.443R \ln 2$.

To understand the NMR spectra it is important to realize that the NMR probes the energy dependence of spatially uniform spin rotations. Thus the exchange Hamiltonian cannot produce shifts of the resonance away from the Larmor frequency. This leaves, as the energy leading to shifts, the anisotropic part of the much smaller nuclear magnetic dipole interaction E_D. To account for

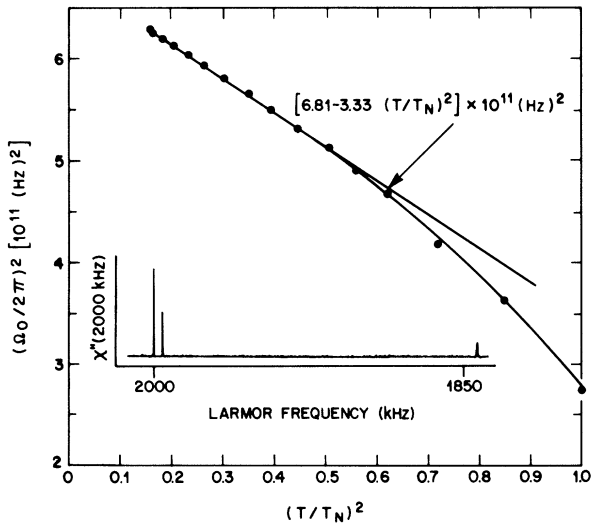

FIG. 2. NMR frequency $(\Omega_0/2\pi)^2$ plotted against $(T/T_N)^2$. The straight line is a best fit to the low-temperature data. In the inset we show a typical NMR spectrum in which χ'' (2000 kHz) is plotted vs applied field (Larmor frequency). The three peaks originate from different domains in a single crystal of solid ^{3}He.

the large shifts measured it is sufficient to calculate E_D neglecting small perturbations due to E_D itself or due to lattice strains. Then symmetry arguments alone show that the classic antiferromagnetic states—normal antiferromagnetic (NAF) and two states each consisting of two interpenetrating, simple-cubic, antiferromagnetic sublattices (SCAF$\parallel$ and SCAF$\perp$) in the notation of Roger, Delrieu, and Landesman[9]—give an isotropic E_D, and hence no NMR frequency shifts of the size measured. We immediately specialize to states in which E_D and the susceptibility $\chi_{\alpha\beta}$ remain invariant under spin rotations about an axis $\hat{d}$.[10] This includes states with all spins "up" or "down" ($\langle \vec{S}_i \rangle$ parallel or antiparallel to $\hat{d}$) and "helicoidal" states in which $\langle \vec{S}_i \rangle$ spirals in the plane normal to $\hat{d}$. The formula Eq. (1) fitted to the data is the solution for small oscillations about equilibrium of the quasihydrodynamic equations of motion (valid for $\omega \ll J$):

$$\dot{\hat{d}} = \hat{d} \times (\gamma \vec{H} - \gamma^2 \chi_0^{-1} \vec{S}), \tag{2}$$

$$\dot{\vec{S}} = \gamma \vec{S} \times \vec{H} - \lambda (\hat{d} \cdot \hat{l})(\hat{d} \times \hat{l}) \tag{3}$$

with $\hat{H} \cdot \hat{l} = \cos\theta$. The first equation arises from the role of $\hat{d}$ as a vector in spin space specifying the broken spin-rotational symmetry.[11] We have written $\chi_{\alpha\beta} = \chi_0(\delta_{\alpha\beta} + \delta d_\alpha d_\beta)$ and the high-field asymptotic behavior requires $\delta < 0$, i.e., $\hat{d}$ lies perpendicular to an applied field. The second equation gives the precession of the total spin density about the field $\vec{H}$, with an additional torque from the dipole energy density $E_D = \frac{1}{2}\lambda(\hat{d} \cdot \hat{l})^2$, $\lambda > 0$, with $\hat{l}$ a direction fixed with respect to the sublattice structure. The general form of the dipole energy is $E_D = -\frac{1}{2}\hat{d} \cdot \overleftrightarrow{A} \cdot \hat{d}$, with $\overleftrightarrow{A}$ a second-rank tensor, with eigenvalues $\lambda_1 \geq \lambda_2 \geq \lambda_3$, reflecting the symmetry of the spin arrangement under lattice rotations. The good fit given by the simpler form of Eqs. (2) and (3), and in particular the zero interecept for the lower mode found on extrapolation to zero field, requires $\lambda_1 = \lambda_2$ (and then we write $\lambda_1 - \lambda_3 = \lambda$). A careful analysis of deviations of the lower modes from Eq. (1) in small fields sets an upper bound of $(\lambda_1 - \lambda_2) < 10^{-3}\lambda$.

We have explicitly calculated $\overleftrightarrow{A}$ in mean-field theory for states described by a single plane wave $\vec{k}$: $\langle \vec{S}_i \rangle \propto \mathrm{Re}[\vec{\sigma} \exp(i\vec{k} \cdot \vec{r})]$, with the further restriction that all $|\langle \vec{S}_i \rangle|$ are equal. With suitable choices of $\vec{\sigma}$ this includes (a) all two sublattice states ($\vec{k} = \frac{1}{2}\vec{G}$, $\vec{G}$ a reciprocal lattice vector), (b) states with ferromagnetic planes arranges in up-up-down-down sequence ($\vec{k} = \frac{1}{4}\vec{G}$), and (c) the helicoi-

dal states. Then, $\overleftrightarrow{A}$ is given by[12]

$$A_{\alpha\beta}(\vec{k}) = 3(\gamma\hbar/2)^2\rho^2\psi^2 f \times [\rho^{-1}\sum_i \exp(i\vec{k}\cdot\vec{r}_i) r_i{}^{\alpha} r_i{}^{\beta}/|r_i|^5], \quad (4)$$

where $\psi = 2|\langle\vec{S}_i\rangle|/\hbar$ gives the amount of ordering, ρ is the number density and $f = +1$ for classes (a) and (b) and $-\frac{1}{2}$ for class (c). We find only two sets of states consistent with the requirement $\lambda_1 = \lambda_2 > \lambda_3$.[13] These are as follows: (i) a state in class (b) with $\vec{k} = (\frac{1}{4},0,0)$ with respect to the conventional cubic unit cell and $\hat{l} \| \vec{k}$; (ii) a set of helicoidal states based on the F points of the bcc Brillouin zone, and $\hat{l}$ parallel to a [111] direction. We show below that only (i) is in fact consistent with all the data. For this state we estimate the zero-field frequency $\Omega_0{}^2 = \gamma^2\chi_0{}^{-1}(\lambda_1 - \lambda_3)$. We use the static susceptibility measurement[6] near T_N for χ_0, and we correct ψ for zero-point fluctuations using calculations for other structures[14] ($\psi \simeq 0.85$). This yields $\Omega_0/2\pi = 880$ kHz, as compared with the zero-temperature extrapolation of the experimental values, $\Omega_0/2\pi = 825$ kHz.

In perhaps twenty separate attempts to grow single crystals, never less than three NMR peaks were observed in high fields. The next lowest number was six. Such a result is expected if the sublattice anisotropy axis is oriented along a different crystal axis in different parts of the crystal. Such a domain structure might result from crystal strains associated with a lattice distortion linked to the sublattice orientation. If indeed domains are responsible, then of all possible sublattice structures considered, only those based on a wave vector in a [100] direction can possibly explain our results, since only such structures have exactly three possible domain orientations. The helicoidal states, (ii) for example, have four, and planar states with a [110] wave vector have six. To test the domain hypothesis, it is only necessary to note that for the [100] structures the angles θ_i for the three possible domains are related: $\sum_i \cos^2\theta_i = 1$. For the data shown in Fig. 1 this sum is 1.007. For all other spectra where enough information existed to determine θ_i, the sum was always unity to within 2%. We consider this convincing support for the domain hypothesis.

In summary, the large zero-field NMR frequency we measure is clearly inconsistent with the "classic" bcc states (NAF, SCAF$\|$, and SCAF$\perp$). The observation of multiples of three peaks in the upper mode with correlated NMR frequencies directly points to a wave vector along the [100] direction. All the data, including the $T = 0$ value of Ω_0, are consistent with the $\vec{k} = (\frac{1}{4},0,0)$ antiferromagnetic state consisting of [100] planes of parallel spins arranged in the sequence up-up-down-down etc., although we cannot conclusively rule out more exotic states based on a [100] wave vector. Our identification of the ordered state, together with the measured temperature dependence of Ω_0 will provide a severe test of any exchange Hamiltonian proposed to describe the system.

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