NMR in Magnetically Ordered Solid ^{3}He

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Magnetically ordered solid ^{3}He, produced by compressional cooling to $T \approx 1$ mK, has been studied by high-resolution NMR at fields up to 2.98 T. In the ordered solid, at low fields $B_0 \leq 410$ mT, peaks in the spectrum above and below $\gamma B_0/2\pi$, characteristic of antiferromagnetic ordering, are observed. For $B_0 \geq 430$ mT, there is a temperature-dependent shift of the resonance to higher frequencies and a large rapid growth in magnetization, beginning at the ordering temperature.

Previously, magnetic ordering of the nuclear spins of solid ^{3}He has been observed at $T_c \approx 1$ mK.[1-3] At low fields, $B \leq 420$ mT, the transition appears to be first order to some type of antiferromagnetic phase.[3,4] For $B \geq 420$ mT, the ordering region is broader, probably indicating a second-order transition.[2,4] Analysis of the melting pressure as a function of field shows that the high-field phase has a high susceptibility, leading to the suggestion of a weakly ferromagnetic state.[4]

We have made NMR measurements with use of a spectrometer[5] with excellent frequency stability and signal-to-noise ratio, and a highly stable magnet of homogeneity ~1 in 10^5. The ^{3}He was cooled by compression along the melting curve. Above T_c there was good thermal equilibrium with only 2–3% solid in the NMR coil. The spin-lattice relaxation time T_1 in the solid was found to be only 60 ± 20 sec at $B = 1.67$ T and 110 ± 30 sec at 2.98 T for $2 \lesssim T \lesssim 10$ mK. When it was desired to produce a large quantity of ordered solid after reaching T_c, solid was formed at rates in excess of 3 mm^3/sec. Some features of the NMR spectrum characteristic of the ordered solid, to be described below, could be seen for 1 to 2 hours after the end of the compression as the cell warmed slowly at approximately constant volume.

Upon reaching T_c, marked changes in the spectrum occurred, which were entirely different for fields above and below $B \approx 400$ mT. Shown in Fig. 1 is a time sequence of spectra for a field of 390 mT. Peak A, a continuation of the peak followed from above T_c, underwent no frequency shift nor decrease in magnitude (area) at T_c. The latter might appear to contradict the sharp decrease in susceptibility observed by Prewitt and Goodkind.[3] However, the low-field transition is probably first order, in which case some of the solid formed at T_c would be in each phase, with very little of that formed above T_c being cooled into the ordered phase. Thus at T_c, a decrease in magnetization would not occur because of the warmer solid remaining above T_c.

On the low-frequency side of the main peak in Fig. 1 is a smaller peak, labeled B, which develops quickly after reaching T_c and continues to grow during the compression and for some time during the warmup. It then begins to decrease in magnitude, but remains at a fixed frequency as it slowly disappears during warmup.

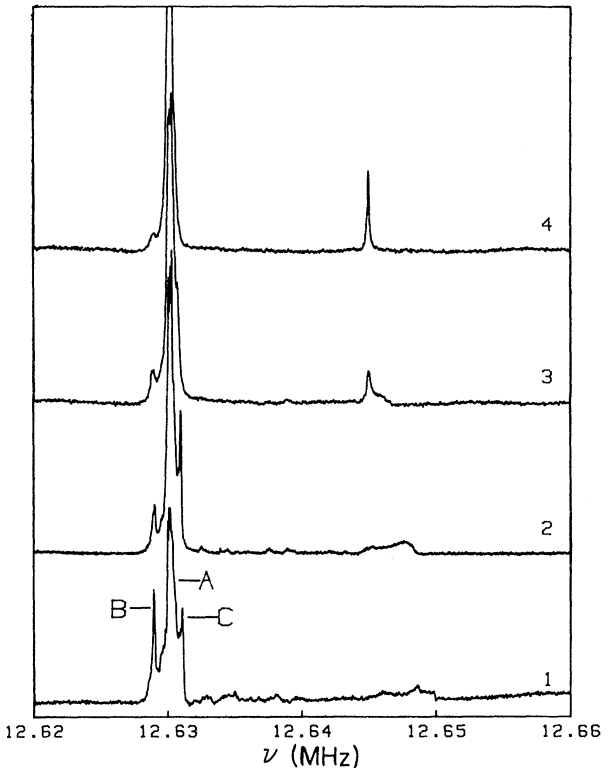

FIG. 1. NMR spectra, intensity vs frequency (MHz), for $B_0 = 390$ mT. The various spectra show the evolution after reaching the ordered phase at the following times: No. 1, 13 min; No. 2, 29 min; No. 3, 43 min; and No. 4, 56 min (the compression lasted 8 min in the ordered phase). The small liquid signal is hidden under the solid signal. Features A, B, and C are explained in the text.

The shift of this peak relative to $\nu_0 = \gamma B_0/2\pi$ was -1.1 ± 0.2 and -0.8 ± 0.2 kHz at $B_0 = 270$ and 390 mT, respectively. We were not able to follow it to lower fields.

As the compression continued after reaching T_c, a number of "bumps" began to appear at $\nu > \nu_0$. These grew in intensity and became narrower for some time after the compression ended, as shown by the sequence in Fig. 1. (In most cases the narrowing was not as pronounced as shown, spectrum No. 2 or No. 3 being typical.) On different compressions at the same field, different patterns of the $\nu > \nu_0$ peaks were seen. However, they were always distributed over a given region of frequency for a given field. The maximum shift $\nu - \nu_0$ for the bumps as a function of field is shown in Fig. 2. The intensity of the bumps decreased with field so that at 60 mT they were barely detectable. (Our NMR coil was designed for high frequencies up to 100 MHz.)

The high-frequency bumps are likely a manifestation of the $\nu > \nu_0$ antiferromagnetic resonance of the ordered solid. Resonances in different crystallites with frequencies dependent on orientation of the crystal axis relative to B_0 would produce the observed bump pattern.[6] With use of a field gradient along the sample to provide spatial resolution,[7] it was shown that several crystallites formed during the compression. The sharpening of the peaks with time could be an annealing effect. The magnitude of the frequency shifts indicates a large anisotropy energy, which is incompatible with the cubic magnetic lattice of most theoretical models.[8]

We interpret our lack of observed temperature dependence of the frequencies of the antiferromagnetic resonances as support for a first-order transition at low fields, with a discontinuous jump in ν at T_c (see discussion of paragraph 3). This is consistent with the results of Osheroff[9] who has studied the temperature dependence of the antiferromagnetic resonances for $B \lesssim 70$ mT. He finds the maximum frequency given by[10] $\nu^2 = \nu_0^2 + \nu_{AF}^2$, where $\nu_{AF}(T_c) \approx 500$ kHz. As shown in Fig. 2, this relation gives shifts near those which we observe; however, it has a faster field dependence than our results.

For fields $B \geq 430$ mT, the spectra were entirely different, with typical behavior shown in Fig. 3(a). At T_c the resonance began a continuous temperature-dependent shift to higher frequen-

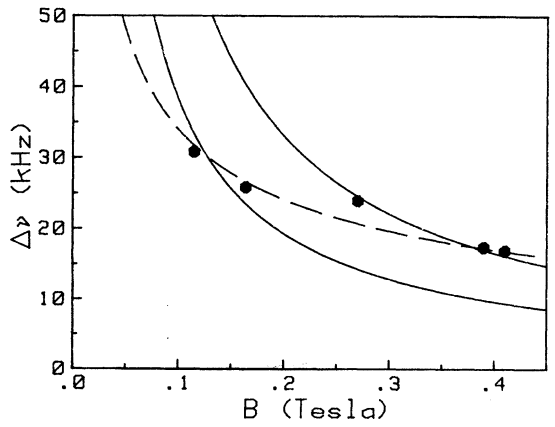

FIG. 2. Maximum frequency shift from $\nu_0 = \gamma B_0/2\pi$ vs field. Circles are present data. The solid curves follow the equation $\nu^2 = \nu_0^2 + \nu_{AF}^2$ with $\nu_{AF} = 650$ for the top curve and $\nu_{AF} = 500$ for the bottom curve. The dashed curve, without theoretical basis, is given by $\Delta\nu = 10.75 \times B^{-1/2}$.

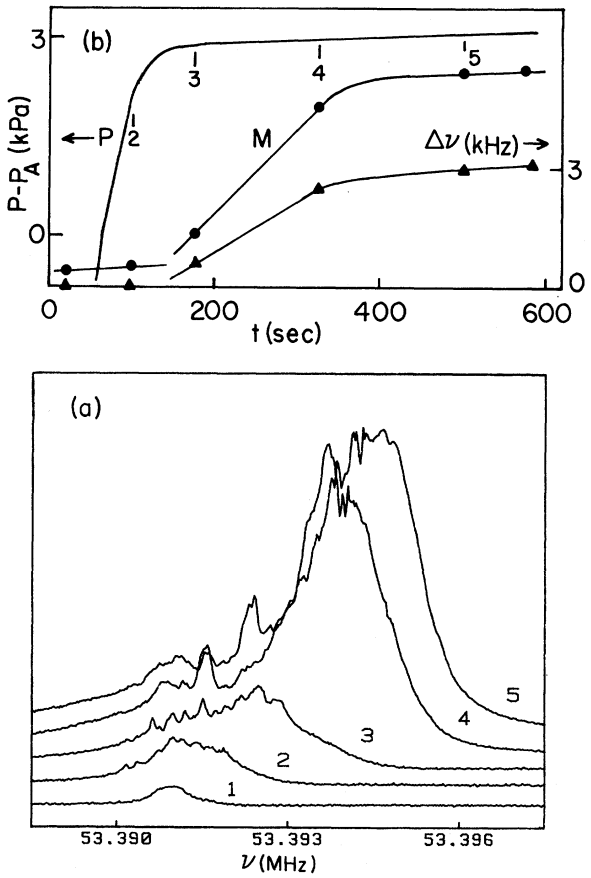

FIG. 3. (a) NMR spectra, intensity vs frequency (MHz), for $B_0 = 1.67$ T. The different spectra are: No. 1, all liquid before compression; Nos. 2–5, are as shown along the pressure trace in (b). (b) Behavior of the melting pressure, magnetization, and frequency vs time during compression through T_c.

cies. The time behavior of the spectrum (also see discussion of slow compression, below) indicated that newly formed cold solid had the largest shift, with the lower-frequency portion arising from previously formed warmer solid. The frequency shift was easily determined as a function of melting pressure. However, accuracy of melting pressure thermometry below T_c is insufficient for determining $\nu(T)$. Figure 3(b) shows the frequency shift, the magnetization, and a trace of the ^{3}He pressure versus time, with the numbers corresponding to those designating the spectra. We can conclude from the rapid onset of the frequency shift that a second-order phase transition is occurring at the higher fields.

The maximum frequency shift observed, about 3 kHz, was almost independent of field for 430 $< B <$ 2980 mT. This shift corresponds to an internal field of $\approx$0.1 mT, which is about the magnitude of the dipolar field if all spins are aligned. In the rapid compressions from a low starting temperature (12 mK), the solid should be reaching a state of very high polarization (low entropy). Thus the 3-kHz shift may be near the $T = 0$ value.

The spectra shown in Fig. 3 were obtained in a rapid compression. In slow compressions at T_c, a small new peak appeared at $\nu > \nu_0$, grew in intensity, and shifted to progressively higher frequencies as the compression continued. Eventually this high-frequency feature was the dominant one, with much of the ν_0 peak having also shifted toward higher frequencies. This behavior is understandable in terms of the large quantity of disordered solid formed above T_c being slowly cooled below T_c as new solid is formed there. It is consistent with the behavior in fast compressions in which a large quantity of solid is rapidly formed below T_c.

Since the amount of solid in the NMR coil is changing with time, obtaining quantitative data on magnetization versus temperature is difficult. However, with a constant compression rate qualitative conclusions can be drawn from the behavior of the magnetization (area under curve) versus time. For $B \gtrsim 410$ mT, upon reaching T_c the rate of growth of magnetization dM/dt increased sharply as shown in Fig. 3(b), whereas for $B \lesssim 400$ mT there was a decrease in dM/dt. The ratio of the maximum magnetization of the ordered solid to that of the liquid was ~90 for a field of 625 mT, but was only ~30 for a field of 390 mT. For high fields, M began to decrease immediately at the end of the compression, but for low fields, M continued to increase before decreasing. These

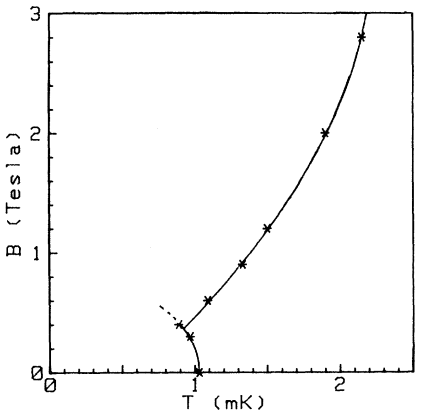

FIG. 4. Magnetic phase diagram as determined from this work and that of Kummer, Mueller, and Adams (Ref. 2) and of Schuberth, Bakalyar, and Adams (Ref. 12).

results, and the frequency shifts, are consistent with identification of the high-field phase as weakly ferromagnetic,[11] with most of the spins being aligned parallel to the field at the lowest temperature reached in the compression.

With use of the above discussed changes in the spectrum to determine the occurrence of the phase transition and with use of the $T(p)$ data of Kummer, Mueller, and Adams[2] and of Schuberth, Bakalyar, and Adams[12] to relate the temperature and pressure, we obtain the phase diagram shown in Fig. 4. Several compressions were made for $B \approx 400$ mT to study the change from low- to high-field behavior in this vicinity. Spectra for fields $390 < B < 410$ mT showed some features of both types of ordering. For example, peak C in Fig. 1 behaves like the high-field resonance while the other features are characteristic of the low-field spectrum. This suggests that for fields $390 < B < 410$ mT some of both phases were produced. This would be the case if there is a triple point at $B \approx 390$ mT with the low-field phase extending to higher fields as suggested by the dashed line in Fig. 4.

In fields of 1.67 and 2.98 T we have measured the magnetization during fast compressions in which the backstep, reported by Schuberth, Bakalyar, and Adams[12] was observed. The results are consistent with the formation of underpolarized solid until the superfluid A transition occurs.[13] Because of space limitations, the details of these measurements will be reported elsewhere.

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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-