Observation of Ferromagnetic Ordering in hcp ^{3}He

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High-density bcc and hcp ^{3}He have been cooled into the magnetically ordered state by direct nuclear demagnetization. Pressure changes during precooling and demagnetization indicated the magnetic state of the system. For a bcc sample with $v = 20.10$ cm^3/mole, the two well-known magnetic transitions were observed. For hcp samples precooled to 614 μK in a field of 2.5 T, for which the spin entropy is almost zero, the transition to the ferromagnetic state was indicated by a decrease in pressure near the end of the demagnetization. For hcp samples with $S \geq 0.46R \ln 2$ ferromagnetic ordering did not occur. [S0031-9007(96)00626-6]

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Magnetism of solid ^{3}He has been of interest for over 40 years because of the unique opportunity that it provides for studying the effects of multiple-exchange interactions. Antiferromagnetic ordering near 1 mK in the low-pressure bcc phase, resulting from competing effects of several exchange interactions (two-, three-, four-particle, and higher), was first observed by Halperin *et al.* [1] in 1974.

In the high-pressure hcp phase, the predominant exchange process is expected to involve three particles because of the triangular arrangement of atoms in the basal plane [2]. Symmetry arguments show that three-particle exchange would produce ferromagnetic ordering [3].

A number of experiments in the paramagnetic phase above the ordering temperature have shown that ferromagnetic interactions are dominant [4–6]. These include magnetization measurements showing a positive Weiss theta [4,5] and an increase in pressure upon cooling in an applied field [6]. The ferromagnetic transition temperature for the highest molar volume is expected to be a few 10 μK [4–6]. However, cooling ^{3}He to such a low temperature using nuclear refrigeration of copper has been precluded because of the large Kapitza resistance between the ^{3}He and the heat exchanger.

In the last few years, attempts have been made to cool the ^{3}He nuclear spins directly as a second stage of nuclear cooling. Okamoto *et al.* [7] have demonstrated success with this technique in cooling bcc ^{3}He into its ordered states as indicated by increases in pressure. Near the end of the demagnetization, a rapid drop in pressure occurred caused by heating accompanying flux exclusion from their superconducting indium O ring. For their hcp samples a decrease in pressure was also seen at low fields and was attributed to heating. They did not identify a transition signature for hcp samples, but took the absence of a pressure increase as evidence of ferromagnetism.

We have used the isochoric pressure, measured with a capacitive strain gauge, to indicate the magnetic state of the ^{3}He during precooling and subsequent demagnetization [8–10]. A number of improvements in the experiment have allowed us to make the first clear observation of magnetic ordering in the hcp phase. We avoided the heating problem by sealing the cell with epoxy (Stycast 2850FT) rather than an indium O ring and took other precautions to assure that demagnetizations were adiabatic. Most significantly, we performed several demagnetizations with different values of the spin entropy, allowing us to distinguish magnetic ordering in low-entropy samples from the behavior of higher-entropy samples which did not order.

The sample cell used in this work, shown schematically in Fig. 1, must meet a number of special requirements because of the changing magnetic field. The compressed silver-powder heat exchanger, connected to the copper first stage by a slotted pure silver rod to minimize eddy current heating, provides thermal contact for precooling the ^{3}He. Coin silver was used for the strain gauge because of its low heat capacity and desirable mechanical properties [11].

In order to minimize problems with eddy current heating in the cap, silver wires were welded to it and screwed to the base flange at the copper stage. Slots to reduce

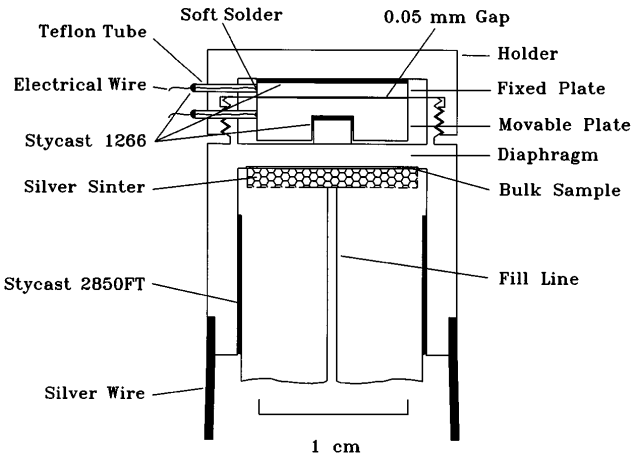

FIG. 1. Schematic drawing of the experimental cell (see text for details).

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eddy current heating were machined in the capacitor plates and the housing for the fixed plate. Threads machined in the cap and fixed-plate housing held them together with a spacing of 0.05 mm between the plates.

Possible heating by the capacitance bridge during demagnetizations was reduced by using an excitation voltage of 2.1 V (peak to peak) applied for only 90 sec every 10 min until the field reached 0.1 T. Then the time interval was reduced to 5 min, with continuous excitation only below about 0.02 T. Changes in capacitance of the empty cell were measured as a function of temperature and field (including ramping). There was no change with temperature in the range of these measurements. However, there was a small field dependence which was subtracted from the raw data.

Samples were formed by the blocked capillary technique, with an initial pressure of about 14 MPa applied to the liquid near 3.4 K. After the solid had formed upon cooling, the samples were annealed for 72 h near the melting point in order to minimize density gradients. After annealing, hcp samples were slowly cooled through the bcc-hcp transition [12] over a period of approximately one week.

Samples were prepared with a particular initial entropy S for subsequent demagnetization by precooling to temperatures between 550 μK and 3.99 mK in a field of 2.485 T. The initial entropy was calculated from the precooling temperature and field. Changes in pressure during precooling indicated the degree of polarization that was achieved.

The 173-mole copper nuclear refrigerator for the first-stage cooling, which can maintain temperatures below 500 μK for many days, has been described elsewhere [13]. A homemade magnet capable of producing 2.5 T was used for second-stage cooling of the ^{3}He nuclei.

We tested our procedure for cooling the nuclei to the ordered state by first studying a bcc sample ($v = 20.10$ cm^3/mole) which has well-known signatures of the two transitions [8,9]. This sample was precooled to 500 μK in a field of 2.485 T by the copper stage over a period of 40 days during which it achieved almost 100% polarization ($S = 0.02R \ln 2$). It was then demagnetized at a constant rate to zero field in 34 h. During the relatively short demagnetization, the Kapitza resistance provides adequate thermal isolation from the copper stage. The possibility of heating of the ^{3}He by the copper stage was reduced further by demagnetizing the copper stage to less than 200 μK just before the ^{3}He demagnetization began.

Our results for demagnetization of the bcc sample, shown in Fig. 2, are quite similar to those of Okamoto *et al.* [7] with one important exception: We do not find a drop in pressure near the end of the demagnetization which they saw and attributed to heating. The absence of heating is crucial for understanding the results for hcp samples, discussed below. The gradual increase in pressure near $B = 0.5$ T indicates the transition from the

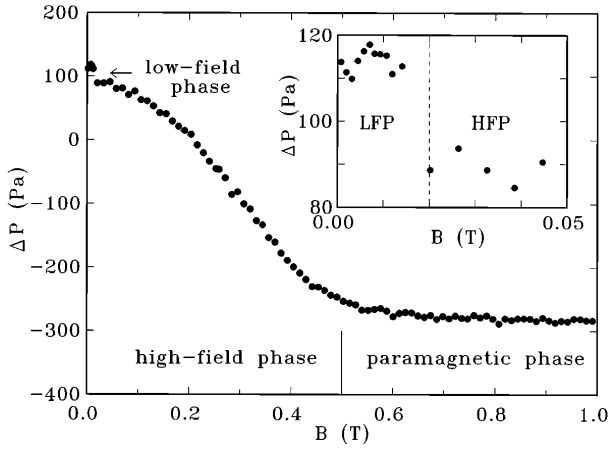

FIG. 2. Pressure change relative to the unpolarized state during demagnetization of a bcc sample with $v = 20.10$ cm^3/mole and $S \simeq 0$. The second-order transition from the paramagnetic phase to the high-field phase occurs at $B \simeq 0.5$ T and the first-order transition to the low-field phase, as shown on an expanded scale in the inset, occurs at $B = 0.02$ T.

paramagnetic phase (PP) to the high-field phase (HFP) while the step at 0.02 T, shown enlarged in the inset, indicates the transition into the low-field phase (LFP).

It is useful to examine several adiabatic demagnetization paths on the B-T phase diagram for different spin entropies, determined by the initial conditions of field and temperature. These are shown in Fig. 3 for the bcc sample with $v = 20.10$ cm^3/mole. The phase diagram was constructed for this volume by scaling from other data [9,14,15]. The adiabatic paths, which join smoothly at high temperature to $B/T = \text{const}$ lines for noninteracting spins, were constructed from the entropy data of Ni, Xia, and Adams [16].

For the $S = 0.10R \ln 2$ case, the PP-HFP transition occurs at 0.3 T, a slightly lower field than we observe for $S = 0.02R \ln 2$, as would be expected. As the HFP-LFP transition takes place near 0.03 T, the temperature increases slightly to 26 μK because of the entropy discontinuity (at constant B or T) at this transition. For $S = 0.30R \ln 2$, the sample is not cooled into the LFP, but remains on the PP-LFP phase boundary to zero field. This behavior of the demagnetization paths for various isentropes for the bcc solid is of importance in understanding demagnetizations of the hcp solid.

In the hcp phase we have studied two samples with nearly the same volumes of 19.61 and 19.65 cm^3/mole. In a series of precoolings and demagnetizations, these samples were precooled to various initial temperatures between 614 μK and 3.99 mK in fields of 2.485 T, corresponding to spin entropies from $S/R \ln 2 = 0.02$ to 0.85. Our results for P vs B during the demagnetizations are shown in Fig. 4.

For the $S = 0.02R \ln 2$ demagnetization, a slight rise in pressure occurs until the field is reduced to about

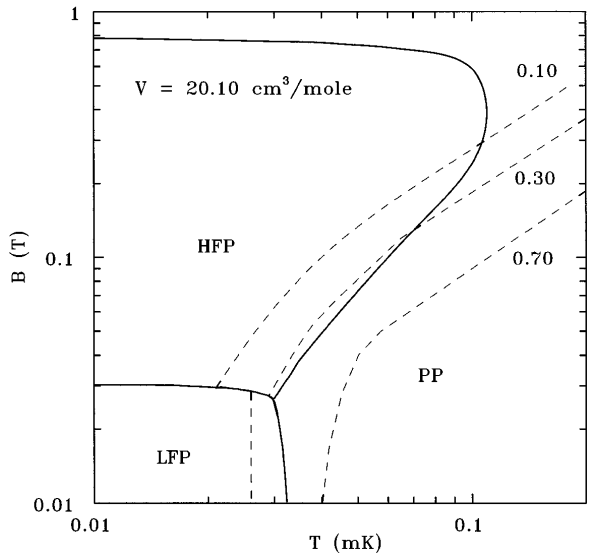

FIG. 3. The B-T phase diagram for $v = 20.10$ cm^3/mole showing various adiabatic demagnetization paths for different values of $S/R \ln 2$ (see text). The ordered state is reached only for $S/R \ln 2 \lesssim 0.5$.

0.2 T where the pressure then begins to drop. For $S/R \ln 2 = 0.72$ and 0.85, which are not expected to reach the ordered state, an increase of pressure occurs at low fields. The pressure is essentially constant for $S/R \ln 2 = 0.46$ with a slight suggestion of an upturn at the very end of the demagnetization.

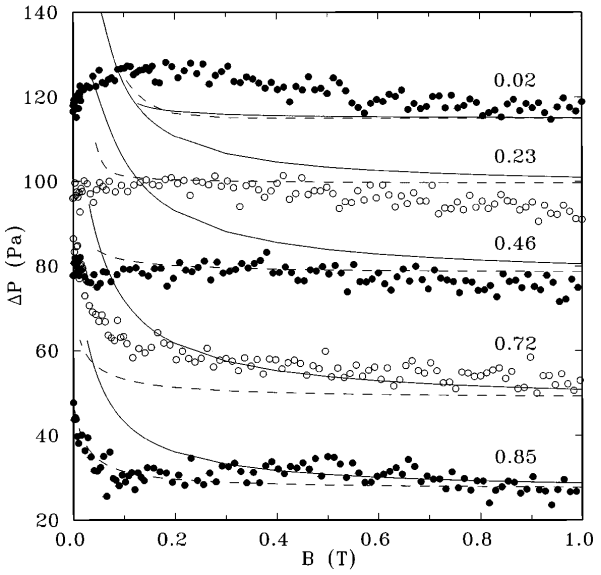

FIG. 4. Pressure changes during demagnetization of hcp samples with various values of $S/R \ln 2$. For $S \simeq 0$, ferromagnetic ordering indicated by a decrease in pressure occurs at low fields. For $S/R \ln 2 \gtrsim 0.5$, ordering does not occur. The solid and dashed lines are calculated from the high-temperature series expansion for the multiple-exchange and Heisenberg nearest-neighbor models, respectively.

As indicated by our results for bcc ^{3}He and for higher-entropy hcp ^{3}He, the decrease in pressure for low-entropy samples in our experiment was not caused by heating. In fact, Okamoto *et al.* [7] also saw a slight decrease in pressure below 100 mT, well before heating by the O ring. The lack of a heating effect and the increase in pressure for high-entropy samples indicate that the decrease in pressure at low entropy that we observe is the signature of the ferromagnetic spin state. (Antiferromagnetism produces the behavior shown in Fig. 2.)

Further support for ferromagnetic ordering is provided by the behavior of the pressure calculated from the high-temperature series expansions using model Hamiltonians. We used the expansion of the multiple-exchange Hamiltonian by Roger, Suaudeau, and Bernier [17] carried to fourth order in $1/k_BT$ and the expansion of the nearest-neighbor Heisenberg Hamiltonian (HNN) carried to tenth order in $1/k_BT$ [18]. Pressure changes relative to the unpolarized state (with a constant pressure added for each entropy in the HNN case) are shown in Fig. 4 along with the experimental results. Both expansions show an increase in pressure at low fields for all values of S, consistent with our results for high entropy. However, the expansions fail to account for the decrease in pressure that we observe for low entropy at low fields. Since the expansions fail at $T \lesssim T_c$, this supports our conclusion that the decrease in pressure is the signature of the ferromagnetically ordered state. Additional experiments, such as magnetization or magnetic resonance, will be required to demonstrate conclusively that the ground state is ferromagnetic.

In this experiment the spin temperature could not be determined during the demagnetization. However, if the relation $B/T = \text{const}$ for noninteracting spins were followed, the temperature would be 20 μK at $B = 0.10$ T where the pressure begins to decrease rapidly (see Fig. 4). The actual temperature for interacting spins is expected to be higher than this (see Fig. 3 for the bcc case).

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