Magnetization of Solid Helium-3 from 0.2 to 11 T

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We have used the depression of the ^{3}He melting pressure with magnetic field to determine the magnetization of the ordered solid phases in the $T=0$ limit from 0.2 to 11 T. In the U2D2 phase we find that $\chi=(5.41\pm0.08)\times10^{-6}$ cgs unit, independent of field, while in the canted antiferromagnetic phase we are able to extrapolate the magnetization for the first time to full ferromagnetic alignment at $H_{c2}=21.7\pm1$ T. This field is of particular importance, as it is the only point in the ordered phases at which the results of mean-field theories, used to calculate physical properties from the Hamiltonian, become exact.

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Nuclear-spin ordering in solid ^{3}He is believed to arise from a combination of atom-atom exchange processes, including at least two-particle exchange, and three- and four-particle ring exchanges.[1,2] There is competition between these exchange processes, for instance two-particle exchange favors antiferromagnetism while three-particle exchange favors ferromagnetism; and this competition is thought responsible for the existence of the very different ordered phases observed above and below a lower critical field $H_{c1}=0.45$ T at melting pressures. Experiment and theory suggest that the low-field phase had a U2D2 magnetic structure, consisting of uniformly oriented spins in planes normal to the [100] direction, with spins on adjacent planes pointing two planes up, then two down, repeated throughout the crystal. Above 0.45 T, a highly polarized phase with a cubic magnetic symmetry exists. It is believed to have the canted antiferromagnetic (CNAF) structure: two interpenetrating simple cubic ferromagnetic sublattices, nearly orthogonal to each other at fields just above H_{c1}. The angle between the sublattice magnetizations decreases as the field is raised. In the multiple-spin-exchange model, complete alignment is achieved at a finite upper critical field H_{c2}. During this process the transition temperature, T_{N}, which rises from about 1 to about 3 mK, should drop rapidly to zero; and the solid magnetization should rise from $0.6M_{\mathrm{sat}}$ to M_{sat}, where $M_{\mathrm{sat}}=0.2672$ cgs unit is the saturation magnetization.

Despite the richness of the ^{3}He phase diagram, it has been difficult to accurately estimate the magnitude of the exchange processes by fits to experimental properties using the multiple-spin-exchange Hamiltonian. Exact high-temperature expansions can be obtained above T_{N} in the paramagnetic phase. Here, however, the demands on experimental precision are severe, especially well above T_{N}, where the series can be truncated at just a few terms. The properties of the ordered phases have only been calculated in the mean-field approximation and it has been shown that some of them can be strongly affected by quantum corrections.[3] As the field approaches H_{c2}, however, mean-field theory becomes exact.[4] One can therefore use a measurement of the upper critical field as an accurate means to determine a combination of the exchange frequencies. However, prior to our results the only experimental indication of that field had been based on measurements of the depression of the ^{3}He melting pressure in fields to 7 T by Godfrin *et al.*[5] This work, while in good agreement with our new results, is not of sufficient accuracy nor does it extend to sufficiently high fields to reliably estimate H_{c2}. Calculations of H_{c2} based on estimates of the exchange parameters obtained by fits to other experimental data have suggested[4] $H_{c2}=15.7$ T, just within the reach of current superconducting-magnet technology. By extrapolation we are able to show that $H_{c2}=21.7\pm1$ T, well beyond current experimental accessibility, but in itself a rigorous test of estimates of the exchange Hamiltonian.

Our measurements were obtained with a powerful demagnetization cryostat consisting of a copper wire nuclear stage which could be precooled to 6.5 mK, providing a nuclear heat capacity in excess of 100 J/K. With this stage it was possible to remain below 1 mK for over two months, and to sweep the sample field from 0.2 to 11 T and back without warming above 3 mK. It was assumed that the temperature of the ^{3}He followed the temperature of the nuclear stage above about 0.4 mK, and the temperature of the nuclear stage was itself calibrated against the melting curve above 1 mK in a field of 0.5224 T. The high heat capacity and intrinsic reversibility of the nuclear stage then allowed us to accurately determine ^{3}He temperatures directly from the magnet current, once the ambient heat leaks had been accounted for. Below about 0.6 mK we could accurately fit the melting pressure, P_m, as $P_0-\alpha T^4$, and by extrapolating that behavior, we were able to show that our limiting ^{3}He temperature was 0.38 mK at 0.5 T, and 0.41 mK at 1 T. Previous work has shown that we can reach below 0.8 mK at 11 T with this apparatus, and perhaps well below that temperature. In these particular measurements, however, accurate thermometry has not been necessary.

Our 16-cm^3 sample was contained within an all-silver variable-volume cell, the body of which was thermally isolated from the 66-m^2 silver heat exchanger, and heat sunk separately to the opposite end of the nuclear stage. Solid ^{3}He was grown in a nylon tail piece at the bottom of the cell, and pressures were measured with a beryllium copper capacitive pressure transducer[6] mounted above the cell in the gradient region of the sample magnetic field. The magnetization was determined from a differential measurement of the depression of P_m through the magnetic Clausius-Clapeyron equation:

$$dP_m/dH = (M_s V_s - M_l V_l)/(V_s - V_l). \qquad (1)$$

Here M_s and V_s are the magnetization and molar volume of the solid, and M_l and V_l are the same quantities for the liquid. The fact that we are working in the limit of negligible entropy has simplified expression (1) as well as our experimental task.

To utilize (1) we have assumed $\chi_l \equiv M_l/H = 1.115 \times 10^{-7}$ cgs unit,[7] and that $V_s = 24.22$ cm^3/mole and $V_l - V_s = 1.314$ cm^3/mole.[8] Although these quantities were not measured in the field or temperature limits of our experiment, we estimate that the probable changes in these properties with field and temperature are negligible. The largest deviations result from the slight rise in the susceptibility of the liquid in the superfluid A phase,[7] which would increase M_l by $0.0002 M_{\text{sat}}$ at 11 T, and from the increase in the molar volume of the ordered solid. To estimate the change in V_s we use the Cornell University data on the solid pressure at constant volume across the para → U2D2 transition,[9] as no data exist in the CNAF phase. This suggests a decrease in $V_l - V_s$ of about 0.17%. These corrections have not been applied to our data.

The evening before a measurement of dP_m/dH, the nuclear stage was cooled to between 0.3 mK ($H < 2$ T) and 0.9 mK ($H > 8$ T), and 0.05 cm^3 of polycrystalline solid ^{3}He was grown at the base of the nylon tail piece. The next morning the temperature was lowered another 20% to check that the melting curve was essentially flat. This procedure was adequate to ensure negligible temperature corrections to the magnetization only above 2 T. At lower fields we extrapolated to $T=0$ the dependence of the magnetization measured between 0.6 and 0.4 mK by NMR, assuming it to vary as $M(0) - \alpha T^2$. We have found it necessary to increase our measurement of M_s by $0.011 M_{\text{sat}}$ at 0.5 T, and by $0.004 M_{\text{sat}}$ at 1 T. For other $H > H_{c1}$, we have assumed a correction proportional to $1/HT_n$, which fits our data but has no physical significance. In the U2D2 phase, the temperature correction to M_s at 0.4 T was only 0.1%.

The measurement process consisted of the determination of the melting pressure with a resolution and stability (over several hours) of 1 μbar before and after the field was swept upward and later downward by typically 40 mT. Below 2 T, fields were determined by NMR on the liquid ^{3}He signal, while at higher fields we had to rely on a measurement of the magnet current and the coil constant we had determined below 2 T. With the NMR we found a 0.6% higher coil constant by measuring dH/dI than we found from H/I. Presumably, the difference is due to flux redistribution after the 40-mT sweeps, but in the end we have averaged the constants found using the two methods. The field was swept at a rate of 0.012 mT/sec to avoid eddy-current heating, a problem which disappeared in high fields because of the increasing transverse magnetoresistance. We then waited 1–2 h after each sweep to ensure temperature and pressure equilibrium. Sweeps were always performed in both directions, and the values averaged. Differences in the two directions were always less than 0.3%. One final correction was necessary because of the force on the strain gauge due to the gradient in the sample magnetic field. This force tended to decrease the apparent pressure as the field was raised, and was measured below 1 mK just below the melting pressure and also at 4 mK with the cell empty. It necessitated a decrease in the values of dP_m/dH varying from 0.48% at 11 T to almost zero below 1 T. In the end, we believe our measurements of magnetization in the high-field phase are accurate to ±0.5%, with a scatter and reproducibility better than 0.3%. In the U2D2 phase dP_m/dH was so low that our errors are probably dominated by the 1.5% scatter in our results.

All our data, corrected for the small systematic effects described above, are shown in Fig. 1. In the U2D2 phase, we have measured $\chi \equiv M/H$ equal to 5.49×10^{-6}, 5.35×10^{-6}, and 5.39×10^{-6} cgs unit in fields of 0.20, 0.40, and 0.42 T, respectively. We therefore conclude that χ is field independent in the U2D2 phase, with a $T=0$ value of $(5.41 \pm 0.08) \times 10^{-6}$ cgs unit. This is

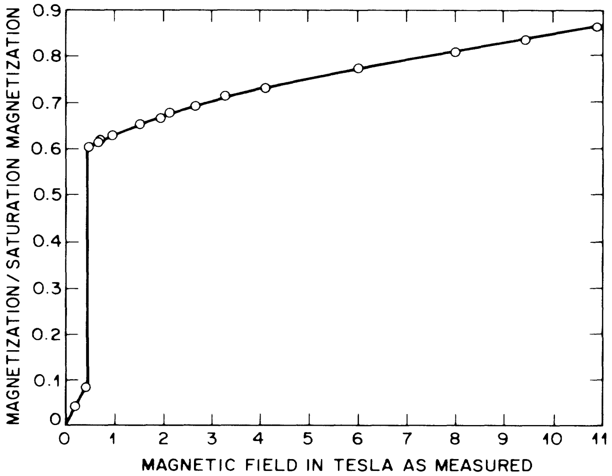

FIG. 1. The $T=0$ magnetization of solid ^{3}He from $H=0.2$ T to $H=11$ T at melting pressures.

about 30% higher than previous values[10] which have been reported; however, we can conceive of no systematic errors of this magnitude which would result in an apparent χ with no field dependence. If one uses the depression of the melting pressure at the superfluid A transition to estimate the solid magnetization at that temperature, and then uses that value to calibrate the measurements of the solid magnetization by Hata *et al.*[11] one finds a value very similar to ours. Using our value, one calculates the $T=0$ antiferromagnetic resonant frequency, Ω_0, in the U2D2 phase with no corrections for quantum fluctuations to be 1002 kHz, compared to the experimental value of 825 kHz. The ratio of these values, 0.823, is close to the renormalization factor for zero-point fluctuations (0.85) which one might have guessed from previous spin-wave calculations on antiferromagnetic states with simple Hamiltonians.[2] This suggests that one can indeed calculate Ω_0 with reasonable accuracy, in particular, more accurately than the difference in Ω_0 between the U2D2 and U3D3 structures.

The transition between the low-field and high-field phases is clearly first order. We have measured this lower critical field at 0.48 mK (0.44478 T) and 0.36 mK (0.44922 T) from which we extrapolate assuming $dH_{c1}/dT \propto T^3$ to find $H_{c1}(T=0)=0.4513\pm0.0005$ T. We see no hysteresis in the transition, but reproducible widths of 0.55 mT (0.36 mK) and 0.71 mT (0.48 mK). These widths do not appear to be caused by thermal gradients, and may be due to strain fields resulting from slightly different lattice deformations in the two ordered phases.

To analyze the high-field data, we first correct for the substantial depression of the melting pressure with field, which leads to an increase in the solid molar volume, and hence to the exchange frequencies. To make our data consistent with constant-volume data at the zero-field melting pressure, we have decreased all magnetic fields assuming that $J \propto V^{18}$. This correction amounts to 3.5% at $H=11$ T, and is nearly linear in magnetic field. The resulting data are plotted in Fig. 2.

To estimate H_{c2} we must extrapolate our data to the field at which $M=M_{\text{sat}}$. We have considered several extrapolation procedures, ranging from a simple linear extrapolation to polynomial fits described below. Values of H_{c2} obtained by such extrapolations range from 18.5 to 23 T, although the extrapolations which yield such extreme values contain no apparent physical motivation. For example, we find the data can be very accurately represented by the expression $M/M_{\text{sat}}=0.5337+0.09766{*}H^{1/2}$ with an rms deviation of $0.003M_{\text{sat}}$ and $H_{c2}=22.8$ T. More realistically, we consider an expansion of $H-H_{c2}$ in powers of $(1-m)$ where $m=M/M_{\text{sat}}$, taking advantage of the fact that $(1-m)$ is small. To estimate the probable fitting errors, we have repeatedly added noise of rms magnitude $0.003M_{\text{sat}}$ to the data and recalculated the fit; and we successively discarded the remaining data point with the worst deviation and recalculated the fit. From such an analysis we conclude that

$$H=(21.18\pm0.3)-(85.6\pm2)(1-m)+(83.7\pm3)(1-m)^2\ \text{T}.$$

The rms deviation of our data to this form of fit is $0.0025M_{\text{sat}}$, and $0.0015M_{\text{sat}}$ if we exclude the three worst points. For this fit $H_{c2}=21.18\pm0.4$ T, and $M_0/M_{\text{sat}}=0.581\pm0.003$, where $M_0=M(H=0)$.

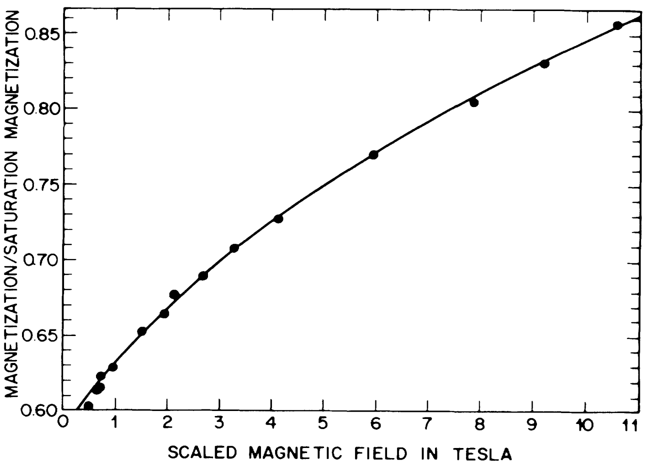

FIG. 2. The $T=0$ magnetization of the CNAF phase of solid ^{3}He corrected to constant volume at the zero-field melting pressure. This solid line is a best fit to the data described in the text.

In what we believe to be a sounder approach, we express the magnetic free energy in the form: $E=-mH+a_1m^2+a_2m^4+a_3m^6$. In the approximation that $\langle\hat{\sigma}_1\cdot\hat{\sigma}_j\rangle=\langle\hat{\sigma}_i\rangle\langle\hat{\sigma}_j\rangle$, and assuming the high-field phase is the CNAF phase, we can identify a_n with all terms in the multiple exchange Hamiltonian containing exactly n products of two-spin operators on different sublattices, and any number of two-spin operators on a single sublattice. Within our approximation, this approach is not an expansion, and accurately accounts for all exchange processes up to and including six-particle ring exchange. Then, by setting $dE/dm=0$, we obtain $H=Am+Bm^3+Cm^5$. Fitting our data, and repeating the above error analysis, we find

$$H=-(9.1\pm0.6)m+(23.8\pm2)m^3+(7.6\pm2)m^5, \qquad (2)$$

with $H_{c2}=22.3\pm0.4$ T and $M_0/M_{\text{sat}}=0.587\pm0.002$. While this expression does not fit the data quite as well as the expansion about H_{c2}, the difference ($0.0018M_{\text{sat}}$ vs $0.0015M_{\text{sat}}$) is probably not significant. If the differences in the results of these fitting procedures are representative of our fit-dependent errors, we can expect

$H_{c2}=21.7\pm1$ T and $M_0/M_{\text{sat}}=0.583\pm0.006$, independent of our extrapolation procedures.

While it is highly desirable that magnetization data be taken which extends closer to H_{c2}, we feel that our current results can already be used to estimate certain combinations of exchange frequencies, or conversely, can be used to test existing estimates of those frequencies. For example, Ceperley and Jacucci[12] have recently used Monte Carlo path-integral techniques to estimate exchange frequencies in solid ^{3}He including, most important two-, three-, four-, and six-particle ring exchange processes. When their values are adjusted to $V=24.22$ cm^3/mole, they obtain $H_{c2}=20.5$ T, in excellent agreement with our determination. We have also attempted to determine the coefficients in (2) from the published exchange frequencies of Ceperley and Jacucci. Using their values and stated error bars, we find

$$H=-(6.0\pm1.4)m+(19.7\pm2)m^3+(6.8\pm1.8)m^5.$$

This expression is reasonably close to (2), except that the magnitude of the first coefficient is significantly lower. This may be due to the crudeness of our approximations, the limited range of our data, or the existence of significant exchange processes not considered by Ceperley and Jacucci. In particular, we point out that while only six- and higher-order ring exchange processes will contribute to the coefficient of m^5 in (2), virtually all exchange processes contribute to the m^1 coefficient, due to the occurrence of products of spin operators on a single sublattice in the terms in the Hamiltonian resulting from those exchange processes.

We conclude by pointing out that our procedure of measuring $m(T=0)$ to observe H_{c2} has many advantages over every other technique of which we are aware. In particular, if our assumptions are correct, dm/dH should be discontinuous at H_{c2}, a signature which should be far more easily measured than the subtle change in entropy at T_{N} as $H\rightarrow H_{c2}$. In addition, our procedure does not require high-resolution thermometry, although field stability is essential. Unfortunately, it appears the H_{c2} lies outside the range of current superconducting-magnet technology, which will make it much more difficult to study solid ^{3}He in the vicinity of H_{c2}.

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