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Solid ^{3}He Magnetic Phase Diagram from Static Magnetization Measurements

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The magnetization of solid ^{3}He has been measured at a density of 24.2 cm^3/mole for fields between 0.8 and 5.8 kG, and for temperatures from 0.55 to 2.0 mK. The measurements reveal the magnetic properties of the phases previously observed by Kummer, Mueller, and Adams, and are in qualitative agreement with the phase lines obtained by them. A new second-order transition has been observed for a narrow range of fields above 4.1 kG.

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In recent years several experiments have shown that solid ^{3}He undergoes a first-order magnetic phase transition at 1.0 mK.[1-6] Kummer, Mueller, and Adams[2] have found that in a magnetic field this transition remains first order up to 4.1 kG and then changes character to a second-order transition for higher fields. In order to study this unanticipated result, we have measured the static magnetization, M, versus temperature in fields, H, from 0.8 to 5.8 kG, and at temperatures down to 0.55 mK. Our measurements reveal the behavior of the magnetization of the previously observed phases, and also show a new transition which exists in a narrow range of fields above 4.2 kG.

A sample of solid ^{3}He was cooled by means of adiabatic demagnetization of a bundle of copper wires. The ^{3}He was contained within a sintered silver sponge with a surface area of 1.0 m^2 and an available volume of 0.1 cm^3. A small portion of the ^{3}He (roughly 10%) occupied a region outside the silver sponge which was left as a consequence of the sponge fabrication. This region had a typical thickness of 0.1 mm. Magnetic fields were applied to the ^{3}He by trapping the required current in a superconducting solenoid wound directly onto the sample container. Accuracy in the setting of magnetic fields was estimated to be 3%. The field was stabilized against drifts by a thin niobium-titanium cylinder between the ^{3}He and the solenoid. The ^{3}He magnetization was sensed by a SQUID magnetometer connected to the sample container by a superconducting transformer. Because of the poor thermal conductiv-

ity of solid ^{3}He, a thermometer was not immersed directly in the ^{3}He. Instead, the temperature of the sample container was measured with a cw NMR platinum thermometer attached to the silver body of the sample container. The thermometer was calibrated against a cerium-magnesium-nitrate mutual-inductance thermometer which had been calibrated against the ^{4}He vapor pressure. The thermometer calibration was checked against the temperature of the ^{3}He-*A* to -*B* superfluid transition at 29 atmospheres. The temperature of that transition, extrapolated to zero magnetic field, was found to be 2.30 ± 0.04 mK which is in excellent agreement with the value 2.313 mK quoted by Paulson *et al.*[7] Absolute accuracy of the thermometer was estimated to be 5% while the precision was 5 μK at 1 mK.

Solid ^{3}He samples were prepared by freezing liquid ^{3}He at the desired density under conditions of constant volume. A molar volume of 24.2 cm^3/mole was chosen for all measurements presented here. It was necessary to reform the sample each time the magnetic field on the ^{3}He was changed. The precision in resetting the ^{3}He molar volume was estimated to be ± 0.05 cm^3/mole. The measurements were begun at a temperature between 16 and 20 mK. In order to remove the arbitrary zero of the SQUID magnetometer the ^{3}He magnetization at the starting temperature was assumed to be given by a Curie-Weiss law with a Weiss temperature of 2.5 mK. The selection of a different Weiss temperature (say 3.5 mK) would alter the absolute size of the measured low-temperature magnetizations by less than 1%.

Data were collected by following the ^{3}He magnetization as sensed by the SQUID during demagnetizations and subsequent warmups. Periodically the temperature of the sample container was adjusted so as to bring it into equilibrium with the ^{3}He it contained and at that time the temperature of the sample container and the ^{3}He magnetization were recorded. Points were taken both cooling and warming. A slight hysteresis in the temperature for a given magnetization was noted and attributed to the longer thermal time constant of the small amount of ^{3}He outside the silver sponge. The magnetization of the sample container materials was measured by collecting data with the container filled with liquid ^{3}He pressurized to the minimum of the melting curve. Since the magnetization of liquid ^{3}He is very nearly temperature independent over the range of temperatures measured, these measurements served to indicate the amount of magnetization signal that was added to that of the solid ^{3}He by the materials of the sample container. Total corrections to the solid ^{3}He signal amounted to less than 3% except at the lowest temperatures and fields where they were 5%.

Before the results are presented, we discuss two features of the experiment which complicated interpretation of the data. First, since portions of the ^{3}He sample occupied two quite different physical environments with necessarily different thermal time constants, care had to be taken to assure that the measured signal represented the behavior of the entire sample and not just one of its constituent parts. This problem was particularly troublesome in the vicinity of sharp features in the ^{3}He magnetization temperature dependence. Data were taken under a variety of different thermal histories in order to determine the true equilibrium behavior. Secondly, because it was necessary to reset the ^{3}He sample density after each change of magnetic field on the ^{3}He it was impossible to guarantee that all samples measured had precisely the same molar volume. This problem takes on added significance when one realizes that, at least for temperatures above 10 mK, the exchange parameters of ^{3}He have a volume dependence of V^{18}. An attempt was made to produce the samples in an identical fashion for each field. Nevertheless, there is evidence that the densities varied by as much as 0.4%.

We obtained data for six magnetic fields: 0.8, 4.0, 4.2, 4.6, 5.0, and 5.8 kG. In Fig. 1 the data have been plotted as an inverse effective susceptibility H/m or χ_{eff}^{-1}. This has been done to facilitate comparison of behaviors obtained in different fields by removing any linear field dependence. For the two low fields investigated, 0.8 and 4.0 kG, the behavior was found to be in agreement with that reported earlier by us.[3] A first-order transition was observed at 0.99 ± 0.01 mK. Below 2.0 mK and above the transition, the magnetization was found to be substantially above the Curie-Weiss value that would be obtained by an extrapolation of the high-temperature behavior. At the transition the magnetization drops within a temperature span of no more than 20 μK to a value of (45 ± 3)% of its maximum. The transition temperature was found to be the same in 4.0 kG as in 0.8. The lack of a magnetic field dependence for the transition temperature is inconsistent with reasonable assumptions about the latent heat and was most likely due to a discrepancy in the molar volumes of the samples measured in

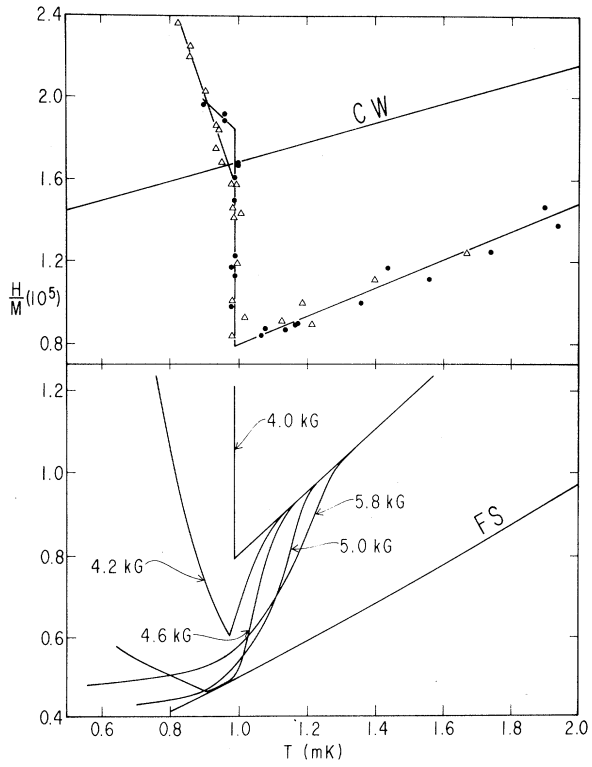

FIG. 1. The inverse effective susceptibility ($\chi_{eff}^{-1} = H/M$) for solid ^{3}He. Upper graph is for the fields (triangles) 4.0 kG and (circles) 0.8 kG. The line labeled CW is the Curie-Weiss $\chi_{eff}^{-1} = (T+\Theta)/C$ with Weiss temperature $\Theta = 2.5$ mK and Curie constant C. The lower graph (note the change in scales) is for fields 4.0–5.8 kG. Individual datum points have been omitted for clarity. Lines are free-hand fit to the data. The line labeled FS is χ_{eff}^{-1} for free spins at 5.0 kG with the ^{3}He moment and density.

0.8 and 4.0 kG.

Compared with the behavior observed at 4.0 kG and below, the measurements at 4.2 kG and above displayed behavior strongly dependent upon magnetic field. At 4.2 kG the effective susceptibility departs abruptly from the low field line at about 1.07 mK and peaks at a temperature of 0.98 mK. (See Fig. 1.) Within experimental resolution the data show that the peak is accompanied by a discontinuous change in dM/dT. The sharpness of the peak combined with the above-mentioned thermal hysteresis made it difficult to measure the precise height of the maximum, but it is at least 30% larger than the maximum value of M/H achieved for lower fields. The behavior at 4.6 kG was also found to display a peak, but one that was not as sharp and whose temperature had decreased to 0.91 mK.

The samples studied at 5.0 and 5.8 kG showed no peak in magnetization down to the lowest temperatures reached, 0.70 and 0.55 mK, respectively. As with the samples measured at 4.2 and 4.6 kG, the behavior measured in 5.0 and 5.8 kG departs from that measured for low fields (Fig. 1). There is a characteristic temperature for this departure that depends on field, and which increases as the field is raised. The data suggest that there is a discontinuity in dM/dT at this temperature as would be expected if the phenomena is associated with the phase transition inferred from the data of Kummer, Mueller, and Adams.[2] Although the magnetization was clearly above the Curie-Weiss value at these temperatures it was never found to be significantly above the paramagnetic value. The 4.6- and 5.0-kG samples reached levels that were within the 3% measuring accuracy of the Brillouin value at 1.0 and 0.9 mK, respectively. The largest magnetization that was measured was that of the 5.8-kG sample at 0.55 mK where it was 46% of the saturation moment.

We next focus on the implications of this work upon the ^{3}He magnetic phase diagram. The experimental evidence to date has been that below 4.1 kG there is a first-order transition with a transition temperature that decreases slightly with increasing field. Above 4.1 kG a second-order transition has been found whose temperature increases rapidly as the field is raised.[2,4] Our measurements provide additional evidence for the existence of both of these transitions. Further, they show that the discontinuity in M/H associated with the first-order transition retains 85% of its low-field value at 4.0 kG. Thus, if the discontinuity is to go to zero continuously at 4.1 kG (as is usually the case for multicritical points) it must do so within a 100-G field range. A rapid field dependence is found near 4.2 kG for the second-order line originally observed by Kummer, Mueller, and Adams. We find that its temperature varies by over 90 μK in 600 G. In addition to the different order of the transition above 4.1 kG (as was observed by Kummer, Mueller, and Adams), we have found that the magnetic character of the lower temperature phase is different above 4.1 kG. It possesses a magnetization that increases as the temperature is lowered in contradiction to the decreased magnetization for the low-temperature phase below 4.1 kG. This suggests that the two low-temperature phases observed by Kummer, Mueller, and Adams may be separated by a third phase line. In fact, at 4.2

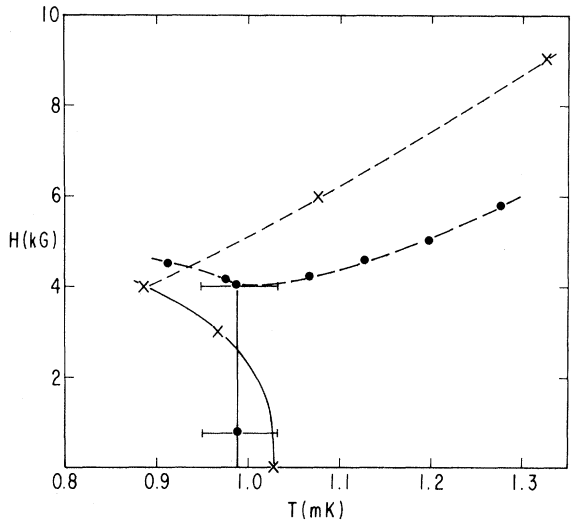

FIG. 2. The ^{3}He magnetic phase diagram as obtained by (crosses) Kummer, Mueller, and Adams, and (circles) this work. Error bars for this work (omitted for fields above 4.0 kG) represent an estimate of the uncertainties due to possible sample molar volume variations from field to field.

and 4.6 kG the cusplike discontinuous change in the slope of the magnetization versus temperature which we observe leads us to conclude that it is a second-order line. If we were merely observing a continuation of the first-order line from lower fields we could not explain how an increase of only 200 G out of 4000 could result in the disappearance of the discontinuity in M. That this phase line might have escaped detection by Kummer, Mueller, and Adams is not surprising as it would have appeared only as a discontinuity in the derivative of the quantity they measured, the ^{3}He entropy. Based on the results of this work we have constructed a magnetic phase diagram for ^{3}He as shown in Fig. 2. Also shown there is the phase diagram obtained by Kummer, Mueller, and Adams.[2] The two works agree as to the qualitative difference of the phase diagram above and below 4.1 kG. Thermodynamic analysis based on the latent heat measured by Kummer, Mueller, and Adams and the magnetization discontinuity which we have found show that the true slope of the first-order line, i.e., the transition for $H < 4.1$ kG, must lie between the two values shown. Similarly the discrepancy in transition temperatures above 4.1 kG may not be significant but due to systematic errors in the method of determining the transition temperature in one or both experiments. The new feature that we have observed is a cusp in magnetization which we have identified as a second-order transition. This transition exists in a narrow field range above 4.1 kG and lies below the temperature of the second-order transition observed by Kummer, Mueller, and Adams.

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