Molar Volume Dependence of Static Magnetization and Ordering Temperature in bcc Solid ^{3}He

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Nuclear magnetic ordering in bcc ^{3}He has been studied by a static magnetization measurement from 10 down to 0.3 mK for a molar volume region of $V=24.14$ to $V=22.46$ cm^3/mole. The results show that the ordering temperature T_N varies in proportion to $V^{16.5\pm1}$ and that the magnetization in the ordered state is constant. The magnetization reduced by its maximum value is found to be represented by a universal function of the reduced temperature T/T_N.

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The low-temperature properties of bcc solid ^{3}He have been described in terms of two-, three-, and four-particle exchange interactions.[1] These exchange interactions, which lead to nuclear ordering in solid ^{3}He, depend strongly on molar volume as shown by the pressure measurements at higher temperatures.[2] The spin structure in the ordered state proposed by Osheroff, Cross, and Fisher[3] suggests that the four-particle exchange process plays an important role in the antiferromagnetic ordering in solid ^{3}He. The magnetic properties of solid ^{3}He near the ordering temperature have been studied by several groups.[1] All of their results, however, have dealt with the solid for a narrow molar volume region near the melting curve. In order to investigate the molar volume dependence of the exchange interaction, we have measured the static magnetization of bcc solid ^{3}He through the ordering temperature for the molar volume region from 24.14 cm^3/mole ($T_N=1.01$ mK) to 22.46 cm^3/mole ($T_N=0.29$ mK).

The experimental arrangement was almost the same as that of Prewitt and Goodkind.[4] A sample of solid ^{3}He was formed in the pores of a sintered silver sponge (nominal 700 Å). Cooling was achieved by nuclear demagnetization of copper (22 mole). The magnetization of solid ^{3}He was measured statically with a SQUID magnetometer in the trapped field of 26 mT. The cell temperature was determined from the susceptibility of Pt wires by means of a pulsed NMR at a resonant frequency of 250 kHz. This NMR thermometer was calibrated against the superfluid ^{3}He-B transition (Helsinki scale).

A sample of solid ^{3}He (^{4}He less than 3 ppm) was made by a blocked-capillary method at about 2 K and annealed for one day. The molar volume of the samples was determined from the pressure in the cell with use of the data of the P-V-T relation on the melting curve.[5] For the cell pressure measurements, we made use of the system of ^{3}He magnetization measurements. The magnetic flux running through the pickup coil changes linearly with pressure in the cell because of the elastic expansion of the cell wall (1.5 mm thickness). No hysterisis was observed in the relation between SQUID output and the pressure up to 100 bars. With use of this measuring system, the molar volume was determined within an accuracy of ±0.02 cm^3/mole. Although the SQUID system picks up the change of pressure as well as magnetization of solid ^{3}He, the two signals can be separated by the temperature region where they change predominantly. Below 10 mK, the total pressure increase due to the thermal expansion and the nuclear ordering transition of solid ^{3}He gives an error less than 1% for our magnetization measurements.[6]

The magnetization of solid ^{3}He was measured at a stationary state. In the course of cooldown or warmup, the cell temperature was held constant until thermal equilibrium was established and then both the temperature and the magnetization of solid ^{3}He were recorded.

Figure 1 shows the magnetization of solid ^{3}He as a function of inverse temperature for six molar volumes. It was obtained by subtracting the magnetization of the cell body from the raw data so that the magnetization of solid ^{3}He obeys the Curie-Weiss law above 5 mK.[7] For each sample, the magnetization increases linearly with inverse temperature in the paramagnetic region and drops suddenly at the ordering transition temperature T_N. In the ordered state, it appears to increase slightly with decreasing temperature. But, the behavior in the ordered state will be considered later.

At the transition, thermal equilibrium could not be obtained because of a very long thermal relaxation time (several days) which is related to a latent heat of the first-order phase transition.

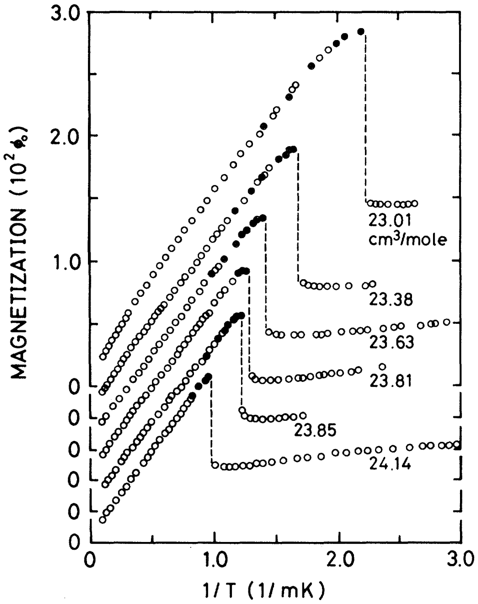

FIG. 1. The magnetization of solid ^{3}He as a function of inverse temperature for six molar volumes. The constant magnetization of the cell body has been subtracted on the assumption that the magnetization of solid ^{3}He obeys the Curie-Weiss law above 5 mK. The large drops of the magnetization show the antiferromagnetic ordering transitions.

Therefore, the ordering temperature was determined as a mean value of the temperatures where magnetization decreases abruptly during cooldown and where it increases during warmup. This transition width was about 20 μK.

Figure 2 shows the ordering temperature as a function of molar volume on a logarithmic scale. In this region, the ordering temperature varies in proportion to $V^{16.5\pm1}$, which is in reasonable agreement with the previous measurement at high temperatures.[2]

In the present method of static measurements, the output of the SQUID magnetometer inevitably involves all the magnetization signals as well as those of bulk solid ^{3}He. As the sample is confined in very small pores of sintered silver, the amount of ^{3}He forming the first few layers on the cell wall cannot be neglected (about 2–3% of the total ^{3}He). We took this surface effect and the impurity atoms in the cell body into account as the source of background magnetization. To see these effects, magnetization measurements have been made under the various conditions of ^{3}He in the cell. Figure 3 shows the results. The magnetization of the cell body is constant below 2 mK. Therefore, all of the temperature-dependent part of the signals comes from ^{3}He. The Curie-Weiss behavior of the low- and high-pressure liquid is probably related to the so-called boundary magnetism.[8] We would like to notice that the magnitude of this excess susceptibility has considerable pressure dependence. Four measurements have

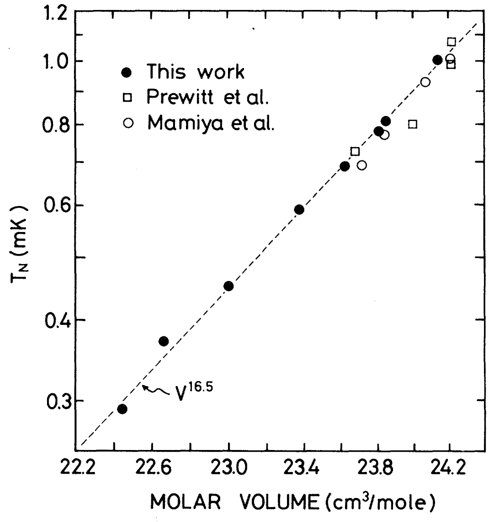

FIG. 2. Molar volume dependence of the ordering temperature plotted in logarithmic scale. The broken line shows the $V^{16.5}$ dependence. Solid circles, this work. Open squares, Prewitt and Goodkind (Ref. 4). Open circles, Mamiya *et al.* (Ref. 6).

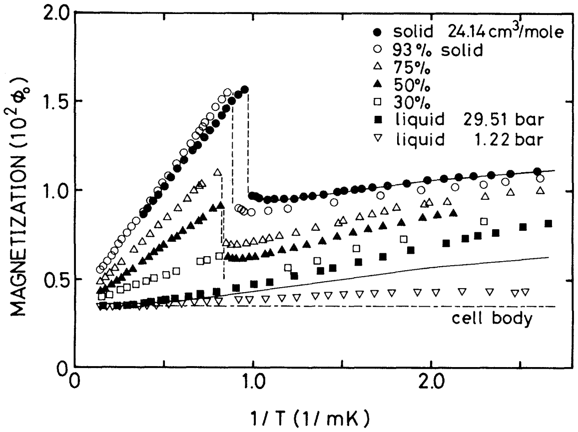

FIG. 3. The raw data of magnetization of ^{3}He as a function of inverse temperature under various conditions of the sample. The magnetization of the cell body was measured while it is evacuated. The magnetization curves of the mixtures of liquid and solid ^{3}He are shown for the molar fractions of solid in the cell from 30% to 93%. The broken lines show the ordering in solid ^{3}He. The estimated background magnetization is shown by a solid line.

been made for the mixture of liquid and solid ^{3}He. In this temperature region, the ratio of liquid and solid in the cell remains essentially constant when the temperature is changed along the melting curve. The antiferromagnetic transition occurs for all mixtures of liquid and solid as well as 100% solid. The magnetization curves for three samples (30%, 50%, and 75% molar fraction solid) are parallel for a broad temperature interval below T_N, and their temperature dependence is almost the same as that of the high-pressure liquid. As the difference of solid fraction resulted in the constant magnetization difference below T_N, we concluded that the apparent temperature dependence is due to the background magnetization of the liquid near the surface layers, and that the magnetization of the bulk solid ^{3}He is constant below T_N.

When the molar fraction of solid is increased to 93%, the temperature dependence below T_N changes from those of lower molar fractions, and coincides with the behavior of 100% solid near the melting curve. We see here that the behavior of the background magnetization changes when the liquid portion in the cell decreases below a certain value. Furthermore, we would like to point out that the ordering temperature for three lower molar fractions of solid are about 15% higher than that of 100% solid on the melting curve. For 93% solid, T_N falls on a value in between. This 15% higher ordering temperature corresponds to the melting curve of ^{3}He depressed by about 1 bar in this restricted geometry.

From the above measurements, we estimated the background magnetization of the solid as follows. The temperature-dependent part of the magnetization for 100% solid below T_N presumably comes from a few layers of solid near the wall surface. We took this as the background below T_N. Although its contribution above T_N is not clear, it will tend to a small value at high temperatures. Assuming that this background magnetization is not affected by the phase transition at T_N, and that it vanishes where the magnetization of liquid ceases to have temperature dependence, we smoothly extrapolated it to higher temperature with the same slope as that of the curve below T_N. The estimated background magnetization is shown by a solid line in Fig. 3. We also assume that this background does not change for the samples of different molar volumes, and subtracted this background from all the raw data for the molar volume region of 23.01 to 24.14 cm^3/mole.

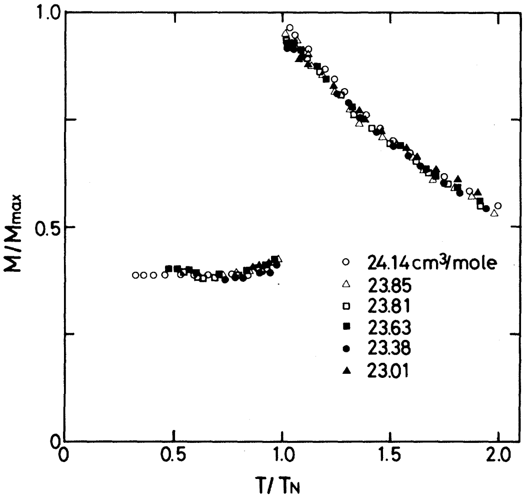

FIG. 4. The reduced magnetization M/M_{max} as a function of reduced temperature T/T_N for six molar volumes.

For each sample, the magnetization starts to deviate from high-temperature Curie-Weiss behavior at $T \simeq 5T_N$ and rise above the curve. At T_N, the magnetization drops to 39% of its maximum value M_{max}. Here, M_{max} stands for the magnetization just above the transition. To obtain M_{max} for each molar volume, we corrected the roundness near the transition. The ratio of the magnetization drop at T_N remains constant for all molar volumes. On the basis of this fact, we analyzed all data to express the reduced magnetization M/M_{max} as a function of reduced temperature T/T_N. The results are shown in Fig. 4.

Figure 4 shows that the reduced magnetization M/M_{max} can be represented by a universal function of T/T_N for all the measured molar volumes. This result implies that the magnetic properties of solid ^{3}He can be characterized by only one parameter T_N for the temperature below $2T/T_N$. If three- and four-particle exchange interaction play a major role in the magnetic properties near the phase transition, the present result leads to the similar molar volume dependence between them, which is consistent with the recent calculation by Roger.[9] Another possible reason is that only one exchange process plays a major role in the phase transition. This reasoning was proposed by Avilov and Iordansky.[10] According to their calculation, if some kind of exchange interaction is more important than others for some value of molar volume, the same kind of exchange interaction dominates at all molar volumes.

We also found that the inverse of $M_{\max}$ is proportional to $V^{16.5\pm1}$ which is the same relation as T_N. Therefore, temperature and molar volume dependence of the magnetization $M(T, V)$ may be expressed in the form

$$M(T, V) = M_{\max} f(T/T_N) \propto V^{-\gamma} f(T/T_N).$$

Here, $\gamma = 16.5 \pm 1$. The functional form of $f(x)$ must be determined by theory.

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