Calorimetry of bcc Solid ^{3}He through the Nuclear Magnetic Ordering Temperature in a Magnetic Field

A. Sawada,[a] H. Yano, M. Kato,[b] K. Iwahashi, and Y. Masuda[c]

Department of Physics, Nagoya University, Nagoya 464, Japan

(Received 17 December 1985)

The nuclear magnetic specific heat of bcc solid ^{3}He of molar volume 24.15 cm^3/mole has been measured as a function of temperature T and magnetic field B for 0.6 mK $< T <$ 40 mK and $B <$ 8.0 T. The discontinuous change in entropy following the high-field to low-field phase transition has been also measured. The magnetic phase diagram has been determined. The observed specific-heat peak at the ordering temperature T_N is discussed in connection with the nature of the phase transition.

PACS numbers: 67.80.Gb, 67.80.Jd

Solid ^{3}He presents a wide variety of interesting properties as a quantum solid. Especially its nuclear magnetism is full of interest. The magnetic susceptibility, pressure, NMR, and melting pressure of solid ^{3}He through the nuclear ordering temperature T_N have been already measured.[1] These measurements revealed that three phases exist in bcc solid ^{3}He at low temperatures: a paramagnetic phase (PP) for temperature higher than 1 mK, a low-field antiferromagnetic phase (LFP) for temperature lower than 1 mK with tetragonal symmetry, and a high-field magnetically ordered phase (HFP) for magnetic field higher than 0.4 T with a cubic structure which has not been determined. The phase transitions from the PP to the LFP and from the HFP to the LFP are first order. The nature of the PP to HFP transition is expected to be first order[2] but there was still uncertainty.[3-5] The magnetic properties of solid ^{3}He can be described with the use of a four-spin exchange Hamiltonian, in which two-spin exchange interactions up to third neighbors (J_n) and two kinds of large planar and folded cyclic four-spin exchange interactions (K_P and K_F) are taken into account. It is expected that these exchange Hamiltonians will explain the complex magnetic phase diagram of solid ^{3}He found experimentally.

Measurements of the specific heat of solid ^{3}He through T_N as a function of temperature at different magnetic fields are expected to provide valuable information on the nuclear magnetic properties. For example, whether new magnetic phases exist or not is significant to investigate. So far the investigated temperature range of specific heat, however, has been limited to temperatures above T_N because of the long relaxation time [6-10] and the measurements have been also limited to zero field. We have cooled the ^{3}He sample down to 0.6 mK and present here the first measurement of the specific heat of bcc solid ^{3}He in the temperature range through T_N in magnetic field. Zero-field results of our work have been reported elsewhere,[11] as well as some preliminary results in magnetic fields up to 1.2 T.

The construction of the cryostat was shown in Fig. 1 of Ref. 11. Cooling was performed by means of an adiabatic nuclear demagnetization of $PrNi_5$. The $PrNi_5$ stage in the present cryostat differed somewhat from that in the Jülich one.[12] We removed the central thermal link which connected the mixing chamber of the dilution refrigerator with the $PrNi_5$ stage, and instead, we used cooper wires, tied to the $PrNi_5$, to connect the mixing chamber with a ^{3}He-sample stage directly. This construction has the merit of an improvement on the packing factor of $PrNi_5$. We worried about the appearance of the heat leak due to the eddy current which flowed along the large electrical loop; however, no such large heat leak was detected. Temperatures were measured by a pulsed Pt-NMR thermometer[13] located on the ^{3}He cell. The steady magnetic field for the

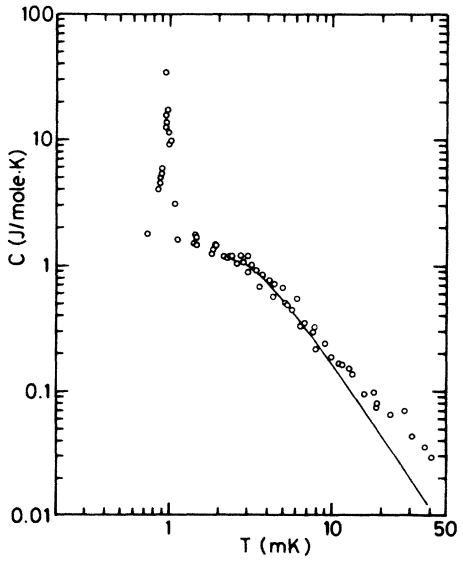

FIG. 1. Specific heat vs temperature in zero field. Solid line shows the calculated result with $e_2 = 9.5$ mK^2 and $e_3 = 15.4$ mK^3.

NMR thermometer was supplied by a superconducting solenoid with a superconducting shield.[14] Accordingly, this field was not influenced by the demagnetization field, nor by a magnetic field applied to the ^{3}He cell for the measurement of the specific heat up to 8.0 T. The NMR thermometer was calibrated down to 4 mK against a nuclear-orientation thermometer of ^{60}Co in hcp ^{59}Co single crystal. The ^{3}He cell was connected to the $PrNi_5$ stage via a superconducting tin heat switch. The cell was machined from a silver rod of 99.99% pure metal and was packed with a sintered silver sponge composed of 700-Å particles.[15] The available volume was 1.701 cm^3 with a surface area of 40 m^2.

The ^{3}He used had a ^{4}He content of 2.7 ppm. Samples were formed at constant volume by use of the blocked-capillary method. They were solidified very slowly for 7 h and then were annealed at a few millikelvins below their melting temperature for 3 h. Heat capacity was measured by the heat-pulse method which was controlled by a computer. The magnitude of heat pulse ΔQ was chosen to limit the temperature change $\Delta T/T$ to less than 5%. The heat capacity resulting from the empty cell was measured at the beginning of the experimental run in the respective magnetic field. It was about $\frac{1}{30}$ of the total capacity at zero field; however, it became appreciably large in high magnetic fields, for instance about $\frac{1}{5}$ at 3.0 T, which was larger than the value expected from the heat capacities of the nuclei, electron, and phonon contributions in copper and silver. The net heat capacity of solid ^{3}He was derived by deduction of the heat capacity of the addenda from the total measured value.

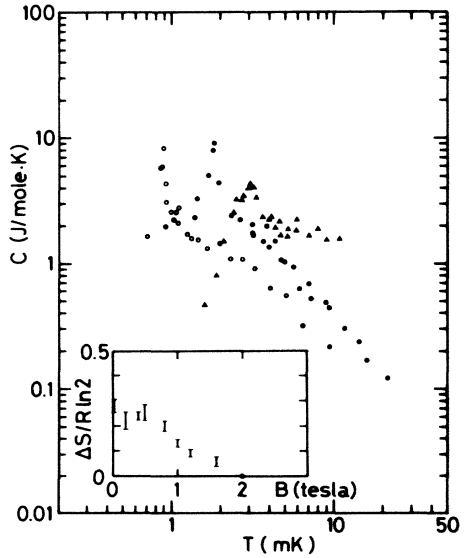

FIG. 2. Specific heat vs temperature in magnetic fields [0.4 T (open circles), 2.0 T (solid circles), 8.0 T (triangles)]. Inset: The entropy reduction at T_N vs the magnetic field.

The experimental results for specific heat of bcc solid ^{3}He of molar volume 24.15 cm^3/mole in magnetic fields up to 8.0 T are shown in Figs. 1 and 2. As can be seen from Fig. 1, the specific heat shows a sharp peak at 0.96 mK in zero field. Corresponding to this peak, the thermal relaxation curve following a heat pulse becomes essentially flat. This peak is thought to arise from the latent heat, which means the nature of this transition is first order. However, the specific heat does not diverge completely at T_N but has a finite temperature width of less than 10 μK, because the density of the sample is thought not to be perfectly uniform.

The specific heat in a magnetic field of 0.4 T is expected to show two transition points, i.e., the one at the lower temperature occurs at the LFP to HFP transition, the other at the HFP to PP transition.[2,4,5] However, in the present experiment only the HFP to PP transition was detected. The variation of phase boundary between the LFP and the HFP against temperature is extremely small. Therefore this flatness of the phase boundary makes difficult its detection by sweeping of the temperature at the fixed field. In order to investigate the LFP to HFP transition in detail, it is thus necessary to sweep the field adiabatically and to measure the magnetic fields at which the temperature changes discontinuously with the LFP to HFP transition and vice versa. The existence of this discontinuous temperature change reveals that the LFP to HFP transition is first order with the discontinuous change

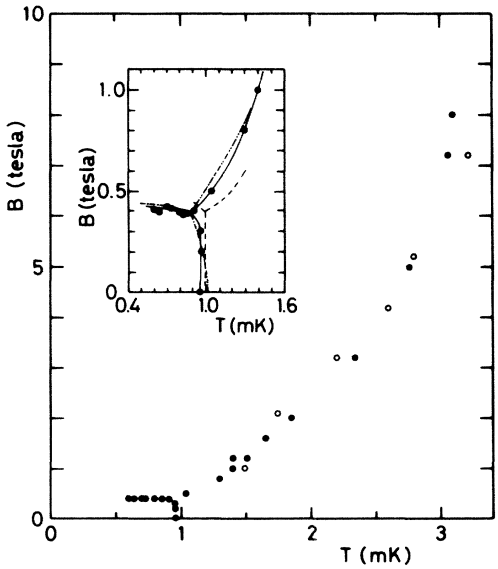

FIG. 3. Phase diagram. Solid circles are the present data and open circles those of Godfrin *et al.* (Ref. 16). Inset: The low-field data. The results of Osheroff (Ref. 2) (dot-dashed line), Kummer, Mueller, and Adams (Ref. 3) (double dot-dashed line), and Prewitt and Goodkind (Ref. 4) (dashed line) are also shown for comparison.

in entropy. The magnetic phase diagram thus obtained is shown in Fig. 3, as well as the specific-heat results.

While it is hard from the present specific-heat results alone to determine positively the nature of the phase transitions that we have observed in bcc solid ^{3}He, we can make some reasonable speculations based on the existence of the latent heat. The specific-heat data in magnetic fields between 0.4 and 1.6 T show the same peak as in the case of zero field, and the peak of the specific-heat curve in this field range is thought to arise from the same latent heat as in zero field. Accordingly, it is thought that the transition from the HFP to the PP is first order in this field range. The plot of the entropy reduction at T_N versus the magnetic field shows zero reduction at a certain field between 1.6 and 2.0 T, as seen from the inset to Fig. 2. This fact suggests that the first-order transition may end at this critical field.

We measured the warmup curve to confirm accurately the critical field. The measurements of the warmup curve were thought to be suitable for a detailed understanding of behavior of the heat capacity. However, a precise determination of the critical field was made difficult by the appearance of an anomaly in the warmup curve. Namely, as shown in Fig. 4, the temperature rises rapidly in the vicinity of the transition point. In the cooling curve, the same quick rise of temperature as seen in the warmup curve was observed. This result suggests that the phenomenon is irreversible. The reasons why this anomaly appears are not clear at present. In order to investigate this anomaly in detail, we repeated the measurements several times. The temperature at which the anomaly in the warmup curve appears falls when the molar volume of the sample is decreased. The field dependence is shown in the inset to Fig. 4. No anomaly in the warmup curve of the addenda alone is observed. One possible explanation for the hysteresis is that a lattice distortion of solid ^{3}He in the ultrafine silver-particle matrix occurs with the phase transition. Nobody knows, unfortunately, how the presence of a large surface area or a strain field affects the solid ordering, at present.

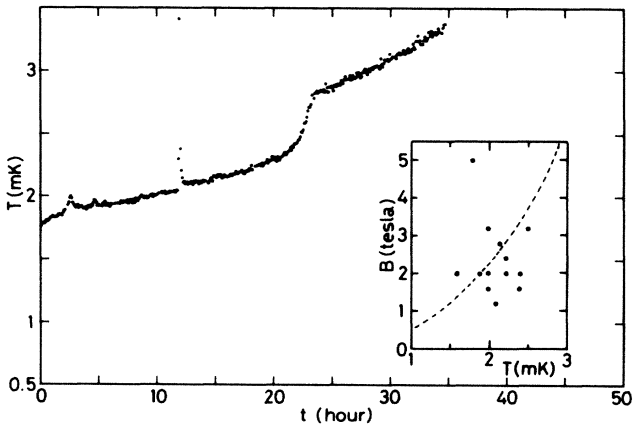

FIG. 4. Warmup curve in a magnetic field of 2.0 T. At $t = 21$ h the anomaly appears and the temperature of the cell, 2.3 mK, rises to 2.8 mK. Inset: The temperature at which the anomaly in the warmup curve appears vs magnetic field. Dashed line shows the boundary of the HFP-PP transition.

It is supposed that the nuclear magnetic relaxation time of solid ^{3}He in high magnetic fields at low temperatures is extremely long, because the energy transfer from the Zeeman system to the exchange system is very slow. The relaxation time is given by Thomlinson, Kelly, and Richardson[17] as follows:

$$T_1(\omega)/T_1(0) = \exp(\omega/\omega_e), \tag{1}$$

where ω is the Larmor frequency and ω_e is the exchange frequency. For example, in the case of a magnetic field of 8 T and a molar volume 24.15 cm^3/mole, Eq. (1) gives $T_1 = 300$ h. Contrary to this expectation, our measurements of the heat capacity of solid ^{3}He in magnetic fields reveal that the thermal relaxation time is short and the thermal resistivity is proportional to T^{-1} and field independent. Giffard *et al.*[18] suggested that the Zeeman energy is transferred to the wall by a process which does not require the intermediate step of Zeeman-exchange coupling. If we assume the spin-diffusion process, the diffusion time is estimated as 0.1 msec in the small area of the sintered silver sponge in the present case. If there is a process of energy transfer from the Zeeman system to the wall, Kapitza resistance becomes the bottleneck for the relaxation process in our system.

By use of the high-temperature expansion, the specific heat above T_N and in low magnetic fields is described up through the T^{-3} term in the following:

$$C = \frac{R}{4}\left[\frac{e_2 + (\gamma\hbar B)^2}{T^2} - \frac{e_3 - 3(\gamma\hbar B)^2\theta}{T^3} + \cdots\right], \tag{2}$$

where e_2 and e_3 are constants independent of temperature and magnetic field. R, γ, $\hbar$, B, T, and θ are the gas constant, gyromagnetic ratio, Planck's constant, magnetic field, absolute temperature, and Curie temperature, respectively. If we fit our zero-field data by Eq. (2), e_2 and e_3 are derived as 9.5 mK^2 and 15.4 mK^3 which are almost the same values as found by Fukuyama *et al.*,[10] who also have observed an excess heat capacity beyond a T^{-2} behavior at high temperature. e_3 has a large positive value, which is consistent with our pressure data published previously,[19] but not consistent with theoretical prediction.[20] Using Eq. (2),

one can determine the value of θ, which is found to be larger than -1.5 mK.

In conclusion, we have presented the first measurement of the specific heat of solid ^{3}He through the ordering temperature in magnetic field. We have obtained the latent heat associated with the first-order phase transition in magnetic fields below about 2 T. We have also observed an anomaly in the warmup curve near the nuclear ordering temperature.

We thank D. D. Osheroff for helpful comments, S. Inoue, T. Mamiya, and T. Uchiyama for their experimental contributions, and the Plasma Technical Division of the Institute of Plasma Physics, Nagoya University, for their technical support. This work has been supported in part by the Grant-in-Aid for Scientific Research of the Ministry of Education, Science and Culture, Japan.

[a]Present address: Department of Physics, Tohoku University, Sendai 908, Japan.

[b]Present address: Canon Inc., Tokyo 160, Japan.

[c]Present address: Department of Liberal Arts, Aichi-gakuin University, Aichi 470-01, Japan.

[1]For example, see M. Roger, J. H. Hetherington, and J. M. Delrieu, Rev. Mod. Phys. **55**, 1 (1983).

[2]D. D. Osheroff, Physica (Amsterdam) **109&110B+C**, 1461 (1982).

[3]R. G. Kummer, R. M. Mueller, and E. D. Adams, J. Low Temp. Phys. **27**, 319 (1977).

[4]T. C. Prewitt and J. M. Goodkind, Phys. Rev. Lett. **44**, 1699 (1980).

[5]K. Uhlig, E. D. Adams, G. E. Haas, R. Rosenbaum, Y. Morii, and S. F. Kral, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics, Karlsruhe, 1984* (North-Holland, Amsterdam, 1984), p. 279; E. D. Adams, Bull. Am. Phys. Soc. **30**, 775 (1985), and private communication.

[6]S. H. Castles and E. D. Adams, Phys. Rev. Lett. **30**, 1125 (1973).

[7]J. M. Dundon and J. M. Goodkind, Phys. Rev. Lett. **32**, 1343 (1974).

[8]D. S. Greywall, Phys. Rev. Lett. **37**, 105 (1976).

[9]B. Hebral, G. Frossati, H. Godfrin, G. Schumacher, and D. Thoulouze, J. Phys. (Paris), Lett. **40**, L41 (1979).

[10]H. Fukuyama, Y. Miwa, A. Sawada, and Y. Masuda, J. Phys. Soc. Jpn. **53**, 916 (1984).

[11]A. Sawada, M. Kato, H. Yano, and Y. Masuda, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics, Karlsruhe, 1984* (North-Holland, Amsterdam, 1984), p. 239.

[12]R. M. Mueller, C. Buchal, H. R. Folle, M. Kubota, and F. Pobell, Cryogenics **20**, 395 (1980).

[13]A. Sawada, H. Yano, and Y. Masuda, Rev. Sci. Instrum. **56**, 480 (1985).

[14]K. A. Muething, D. O. Edwards, J. D. Feder, W. J. Gully, and H. N. Scholtz, Rev. Sci. Instrum. **53**, 485 (1982).

[15]Vacuum Metallurgical Company, Ltd., Yokota 513, Yamatake-cho, Yamatake-gun, Chiba-ken, Japan.

[16]H. Godfrin, G. Frossati, A. S. Greenberg, B. Hebral, and D. Thoulouze, Phys. Rev. Lett. **44**, 1695 (1980).

[17]W. C. Thomlinson, J. F. Kelly, and R. C. Richardson, Phys. Lett. **38A**, 531 (1972).

[18]R. P. Giffard, J. P. Stagy, W. S. Truscott, and J. Hatton, J. Low Temp. Phys. **31**, 817 (1978).

[19]T. Mamiya, A. Sawada, H. Fukuyama, Y. Hirao, K. Iwahashi, and Y. Masuda, Phys. Rev. Lett. **47**, 1304 (1981).

[20]H. L. Stipdonk and J. H. Hetherington, Phys. Rev. B **31**, 4684 (1985).