Low-field phase boundaries of bcc solid ^{3}He

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The low-field phase boundaries of bcc solid ^{3}He at V=24.18 cm^3/mole were determined from isochoric pressure measurements. We clarified that the critical magnetic field B_N of the transition line between the low-field and the paramagnetic phases was described as $B_N \sim \sqrt{T_N(0) - T_N(B_N)}$ as a function of the critical temperature T_N. The sharp kink at the triple point along the low-field phase boundary indicates that the transition between the high-field and paramagnetic phases is first order near the triple point.

The structure of low-density solid ^{3}He is simple, with nuclear spin $\frac{1}{2}$ on a body-centered cubic structure. In spite of its simplicity, a wide variety of experiments, since the first observation of nuclear ordering,[1] have found that solid ^{3}He possesses multiple magnetic phases in the millikelvin temperature range. A sample of solid ^{3}He with the melting density in zero magnetic field undergoes a first-order phase transition from the paramagnetic phase (PP) to the low-field phase (LFP) with the *uudd* antiferromagnetic spin structure at 0.93 mK.[2] The transition temperature slightly decreases upon increasing the applied field. As the field is raised beyond about 0.4 T, there exists a high-field phase (HFP). The magnetic structure of the HFP is believed to be a canted normal antiferromagnetic type.[3] The rich magnetic phase diagram of solid ^{3}He mentioned above is now generally understood to arise from multiple exchange interactions among ^{3}He atoms.[4,5]

There still remains controversy about the order of the HFP-PP transition. The transition in the higher-field region has been generally accepted as second order, but the order, near the triple point (TP) in the lower-field region, has not yet been conclusively determined. Measurements of the NMR frequency shift by Osheroff,[6] the specific heat and the melting pressure by Sawada *et al.*,[7,8] and the melting pressure by Okamoto *et al.*[9] indicate that the transition is first order or most likely first order near the TP. On the other hand, studies of the static magnetization by Prewitt and Goodkind,[10] the melting pressure for fields up to 0.495 T by Tang and co-workers,[11] and the specific heat at 0.6 and 1 T by Greywall and Busch[12] suggest that the transition is second order. Recent work of the melting pressure by Xia, Ni, and Adams[13] reported the transition is first order up to the tricritical point near 0.6 T.

There exist two methods of investigating the order of the HFP-PP transition. One is the direct approach of measuring the change of a physical quantity across the phase line. The other is the indirect procedure of examining how the other two phase lines connect at the TP. The slopes of the phase line between the LFP and the other phase are described by the magnetic Clausius-Clapeyron equation as follows:

$$\frac{dB_{C1}}{dT_{C1}} = -\frac{S_L - S_H}{M_L - M_H} \quad , \quad \frac{dB_N}{dT_N} = -\frac{S_L - S_P}{M_L - M_P} \ . \tag{1}$$

Here S and M are the entropy and the magnetization, respectively, and the suffixes L, H, P, $C1$, and N refer to the LFP, HFP, PP, LFP-HFP transition, and LFP-PP transition, respectively. If the HFP-PP transition is first order, dB_{C1}/dT_{C1} does not equal dB_N/dT_N at the TP. This implies that the LFP boundary has a kink where it intersects the HFP-PP line. Presently there are four experiments[6,8,9,11] in the literature which discuss the order of the HFP-PP transition on the basis of the behavior of dB_{C1}/dT_{C1} and dB_N/dT_N at the TP. Two of them[8,11] do not have enough data to verify a slope discontinuity and cannot deduce the functional forms of two phase lines. The work by Okamoto *et al.*,[9] with much data, uses melting pressure as a parameter corresponding to temperature. Unfortunately, as the melting pressure depends on both temperature and magnetic field, the properties of the LFP boundary remain ambiguous. Therefore few experiments exist which contain enough data covering the LFP boundary, including both the LFP-HFP and the LFP-PP transitions, to conclusively determine the order of the transition.

Our purpose in this paper is to determine the order of the HFP-PP transition near the TP in bcc ^{3}He and to give more conclusive evidence to resolve the controversy discussed earlier. In previous experiments[6,8,9,11,13] a ^{3}He sample at melting density was employed, the molar volume of which changes with both temperature and magnetic field. We decided to measure the isochoric pressure in which the magnitudes of the exchange interactions are constant. In addition to this, there is another advantage in this measurement compared with melting pressure and specific-heat measurements. Solid ^{3}He adhered to the surface of substances, strongly influenced by the surface potential, may have different properties from bulk ^{3}He. It is known that in specific-heat and static magnetization measurements, solid ^{3}He in the pores of a heat exchanger contributes to the measurement and is never negligible. On the other hand, in pressure measurement only the solid ^{3}He sample between a diaphragm and a heat exchanger contributes to the pressure measurement,

the solid in a heat exchanger does not.[14]

At first we measured the isochoric pressure across the HFP-PP transition to investigate the existence of the pressure jump.[15] However, in one measurement a jump in the unexpected opposite direction was observed; in another no jump at all was seen. We suppose now that these phenomena seem to be related to the hysteresis of the first-order phase transition. In this paper, using isochoric pressure measurements, we investigate both the LFP-HFP and the LFP-PP transitions, present a precise magnetic phase diagram in the LFP, and determine the order of the HFP-PP transition from the behavior of the LFP boundary at the TP.

We used a nuclear-demagnetization refrigerator[8] with a 30 mole copper stage and a 9 T magnet. At present we can maintain the temperature below 1 mK for about 4 months after the demagnetization. This cryogenic platform enables us to make experiments on solid ^{3}He under as close to a thermal equilibrium condition as feasible and to determine a more precise magnetic phase diagram of ^{3}He. At temperatures higher than 0.93 mK, we have employed a ^{3}He melting-curve thermometer on the basis on the Greywall scale,[16] with which we calibrated a ^{195}Pt-pulsed NMR thermometer that determined temperatures less than 1 mK. In order to check our NMR thermometer at temperatures below 1 mK, we measured the ^{3}He melting pressure in zero magnetic field. This measurement was consistent with the results of Osheroff and Yu.[17]

Our pressure cell was a typical Straty-Adams-type strain gauge.[18] The body of the cell and the diaphragm were made, respectively, of 99.99% silver and beryllium copper. The solid ^{3}He sample was formed in a bulk, albeit thin, cylindrical space (26 μm thickness and 8 mm diameter) which was located between the diaphragm and the heat exchanger. The heat exchanger was made of sintered-silver powder[19] with a large specific surface area of 4.9 m^2/g, which minimized the thermal relaxation time of the solid ^{3}He sample. This time constant was about 8 h near the HFP-PP transition. As the resultant volume correction factor was small (about 3%), we present the raw data without correction. The homogeneity of the magnetic field at 0.5 T, for the sample cell, was 3×10^{-3} over 1 cm on the central axis.

High-purity ^{3}He with less than 1 ppm ^{4}He impurity was prepared.[20] The ^{3}He sample was annealed for about 24 h at a temperature just below the melting point before any measurements were made. This procedure is dramatically effective in reducing the pressure noise by minimizing density inhomogeneity. The resultant molar volume of our sample was determined using the compressibility of solid ^{3}He and the difference between the melting pressure and the pressure of our all-solid sample at the melting density Néel temperature, 930 μK. All the data we present in this paper were taken on a single sample with 24.18 cm^3/mole which was never remade, because it is difficult to remake just the same density sample.

A magnetic field sweep method was used to determine the LFP-HFP transition points, which is an effective way to examine such a weakly temperature-dependent phase line. Figure 1 indicates the pressure change of the LFP-HFP transition as a function of magnetic field at 353 μK.

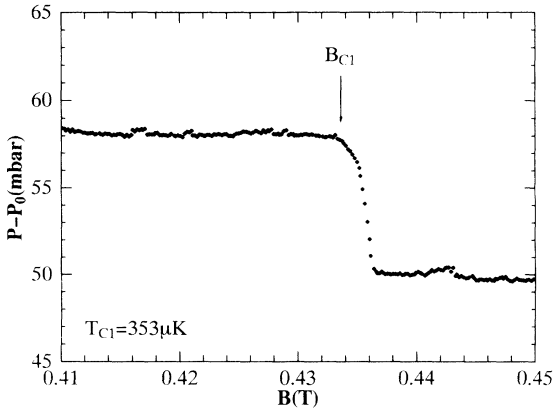

FIG. 1. The pressure vs magnetic field near the LFP-HFP transition at 353 μK. P_0 is the pressure at the high temperature.

The pressure drop ($\Delta P_{C1} \sim 8$ mbar) accompanying the first-order phase transition is obviously observed. Our continuous field sweep method (not stepwise sweep) increases the apparent transition width due to the latent heat at the transition point. All the LFP-HFP data were taken only in one direction, increasing the magnetic field from the LFP to the HFP. Therefore we regard the beginning of the pressure drop as the transition point, as shown by the arrow in Fig. 1. This way of determination is reasonable because we made sure that the transition point obtained by the continuous sweep method excellently agreed with the one by the stepwise method at the same temperature. The field sweep rate is always about 2.3×10^{-3} T/h.

There exists a large latent heat between the LFP and the PP which makes the thermal equilibrium time strikingly long near the transition. It is rather difficult to determine the transition points precisely. Consequently it is easy to mistake them. The transition temperature differences among the previous data by other groups[6,8,10,11] appear to be attributable to the thermal equilibrium problem of the ^{3}He sample. Therefore the LFP-PP data were taken as close to complete thermal equilibrium as possible after changing the temperature. Especially near the transition, temperature was changed by a narrow step of about 10 μK so that when we write $T_N = 850$ μK we mean that the transition happens at a temperature between 845 and 855 μK. Figure 2 shows the pressure change as a function of temperature at the fixed magnetic field of 0.3663 T. The solid circles denote the thermal equilibrium points which, near the transition, were taken by keeping the temperature constant within 1 μK for a few days and ensuring pressure equilibrium. T_N at this field is 828 ± 5 μK. The same transition temperature was obtained when both increasing and decreasing the temperature.

The magnetic phase diagram shown in Fig. 3 was obtained by repeating both the temperature and the field sweeps mentioned above. As expected from the antiferromagnetic spin-wave theory, the LFP-HFP transition field B_{C1} at the temperature T_{C1} is described as

$$B_{C1}(T_{C1}) = B_{C1}(0) + \alpha T_{C1}^4 + \beta T_{C1}^8 + \cdots , \qquad (2)$$

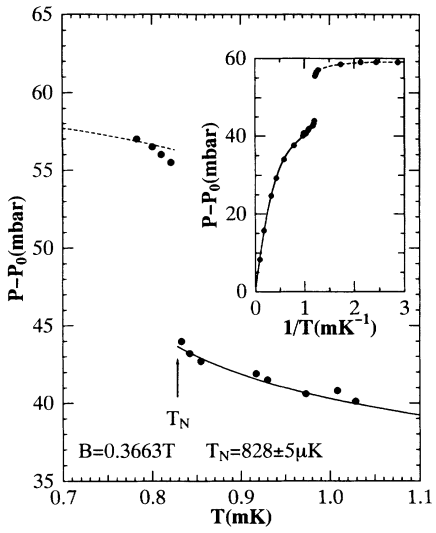

FIG. 2. The pressure vs temperature near the LFP-PP transition for B=0.3663 T. The inset shows an overall pressure change as a function of inverse temperature over a wider temperature range. The broken line below T_N indicates a least-squares fit of $P(T) = P(0) + aT^4$. The solid line above T_N shows $P - P_0 = A_1/T + A_2/T^2 + A_3/T^3$.

where α and β are constant coefficients. The solid line in Fig. 3 is the least-squares fit of this form to all the LFP-HFP transition data. The βT_{C1}^8 term is so small that its contribution is negligible even near the TP. Fukuyama *et al.*[21] have already reported the same relation (2) as ours. Their least-squares fitting of this form was done only to the data at temperatures well below the TP. Our work shows that the LFP-HFP data can be well fitted even at temperatures near the TP. The $B_{C1}(0)$ value extrapolated to zero temperature is 0.438 T. Substituting our

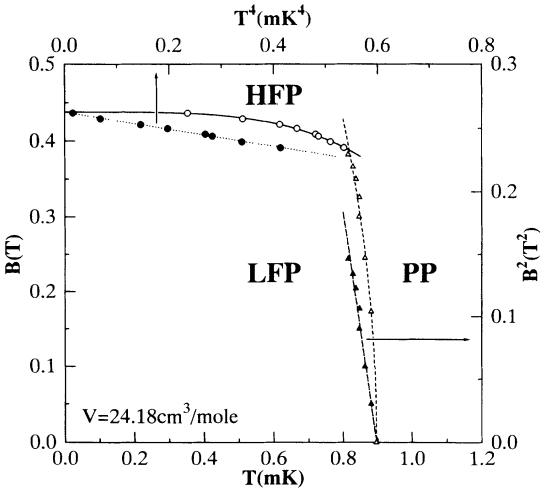

FIG. 3. The resultant magnetic phase diagram of bcc ^{3}He. The LFP-HFP transition fields B_{C1} are plotted as functions of T_{C1} and T_{C1}^4 (the dotted line), and the LFP-PP transition temperatures T_N as functions of B_N and B_N^2 (the dashed line). The temperature deviation of the LFP-PP transition points (open triangles) is almost equal to ± 5 μK.

molar volume into their volume dependence equation of $B_{C1}(0)$,[21] we get $B_{C1}(0)$=0.444 T.

From a simple calculation, it is easy for us to obtain the theoretical functional form of the LFP-PP transition line. In the range where $M = \frac{\chi_1}{\mu_0}B + \frac{\chi_3}{\mu_0}B^3 + \cdots$ is a good approximation, the free energy of the LFP and the PP can be both written in the even power series of B. On the LFP-PP transition line, we get the following equation:

$$\gamma B_N^2 + \delta B_N^4 = \Delta S\big(T_N(0),0\big)\cdot\big(T_N(0) - T_N(B_N)\big)\ . \quad (3)$$

Here γ and δ are the constant coefficients and $\Delta S\big(T_N(0),0\big)$ is the entropy change at the Néel temperature for zero magnetic field. The LFP-PP transition can be described by this functional form. From the least-squares fit of this form to our eight data points, it turned out the δB_N^4 term makes less contribution near the TP than γB_N^2, from which the LFP-PP transition line results in the simpler approximation

$$B_N \sim \sqrt{T_N(0) - T_N(B_N)}\ . \quad (4)$$

The broken line in Fig. 3 is the least-squares fit of this form. It was found in this work that the LFP-PP transition could be written in this simplified form. $T_N(0)$ at zero magnetic field is 897 μK, in excellent agreement with the results derived from the relation: $T_N(0) \sim V^{18\pm1}$.[22]

Assuming that the HFP-PP is second order, neither the higher-order spin-wave term nor the magnetization term near the TP becomes negligible. As a result, two LFP boundaries will connect smoothly at the TP. Nevertheless, the higher-order terms βT_{C1}^8 and δB_N^4 obtained from this work are so small that the boundaries never do that. Should either of two phase lines abruptly break in the vicinity of the TP, while the two phase lines connect smoothly at the TP, there must be a fourth new phase. From all of experiments so far, we can exclude the possibility of its existence. Therefore, the HFP-PP transition is first order near the TP. The intersecting point of the two functional forms can be taken as the TP (B_3 , T_3). Our value is (B_3 , T_3)=(817 μK , 0.388 T).

From this work, we have two experimental values of the slope of the phase lines at the TP, -243 and -2450 T/K for the LFP-HFP and LFP-PP transition, respectively. The slope of the LFP boundary changes by about a factor of 10, which is equal to Osheroff's results.[6] From the change of the slope at the TP, we can deduce other physical quantities. In addition to the above two values, we take 475 T/K for the dB_C/dT_C of the HFP-PP transition at TP from Ref. 11. Assuming that the entropy discontinuity of the LFP-PP transition at the TP is $\Delta S_{LP} = 0.23R\ln 2$ from Ref. 13 and substituting the above values into Eq. (1), the deduced jumps in entropy and magnetization ($\Delta S, \Delta M$) of the HFP-PP transition at the TP are $\Delta S = 0.14R\ln 2$ and $\Delta M = -0.26M_{\rm sat}$, respectively. Here $M_{\rm sat}$ is the saturation magnetization.

Here we consider some of the other experiments focusing on the order of the HFP-PP transition near the TP. There are two groups in specific-heat measurement[7,12] that arrived at opposite conclusions. It is difficult to judge the transition order from the divergence in the spe-

cific heat of a system with a long thermal equilibrium time. There also exists the notable point that they performed experiments at different magnetic fields, taking account of the result of Xia, Ni, and Adams.[13] Also, we suppose that the magnetization jump[10] disappeared because of the inhomogeneity of the sample in the sintered silver.

We conclude that the HFP-PP transition is first order in the vicinity of the TP. There has been little theoretical support for a first-order HFP-PP phase transition for some time. The recent calculation by Sun and Hetherington[23] first proposed a tricritical point where the HFP-PP transition changes from first to second order, showing the transition is first order near the TP.

After submission of this paper, similar results by Xia, Ni, and Adams[24] were published. They measured the melting pressure to determine the phase diagram including the LFP boundary. The shape of the LFP boundary obtained by them is in excellent agreement with this work. Their measured entropy jump of the HFP-PP transition of $0.13R\ln 2$ at the TP is the same as ours within experimental error.

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