Volume Dependence of the Lower Critical Field in Nuclear-Spin Ordered bcc ^{3}He

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The zero-temperature lower critical field $B_{c1}(0)$ between the two nuclear antiferromagnetic phases of bcc ^{3}He has been determined as a function of molar volume from pressure measurements down to 120 μK. In a volume range between 22.45 and 24.06 cm^3/mol, the Grüneisen parameter for $B_{c1}(0)$ is anomalously small compared to typical values for other physical quantities associated with the exchange interactions. The result suggests that the two-spin exchange frequency decreases more rapidly with decreasing volume than three- and four-spin exchanges in the terms of the multiple-spin exchange model.

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The bcc phase of solid ^{3}He provides a model system of the spin-$\frac{1}{2}$ antiferromagnet. The system can be considered as weakly interacting hard spheres, and the nuclear magnetic properties at millikelvin temperatures are dominated by purely isotropic exchange interactions between neighboring atoms due to the zero-point motion [1,2]. We can easily obtain ultrahigh-purity samples [3] and can ideally change the lattice spacing by 7%, which introduces a factor-of-30 change in the exchange frequencies. These features offer unique opportunities to investigate extensively the antiferromagnetic ordering phenomena and the microscopic mechanism of the atom-atom exchange processes, even comparing experimental results quantitatively with first-principles calculations [4,5].

In spite of the structural simplicity, bcc ^{3}He has a rather rich magnetic phase diagram at the melting density. Below about 1 mK, there are two antiferromagnetic ordered phases with quite different magnetic properties separated by a lower critical field (≈ 0.4 T) [6]. The spin structures are most likely identified as the u2d2 phase [7] and the canted normal-antiferromagnetic phase [1] for the low-field phase (LFP) and the high-field phase (HFP), respectively. To understand the magnetism in this system, several antiferromagnetic and ferromagnetic multiple-spin exchange processes J_x, at least up to four spins, have been assumed to exist, i.e., the multiple exchange model (MEM) [1], which is applicable to other nearly localized fermion systems with many-body interactions.

Although the MEM can explain most of the qualitative properties of the magnetic phase diagram at the melting density as a result of fortuitous competition among the J_x [8], there is a serious question about its applicability to bcc ^{3}He. Because of the importance of steric effects between neighboring atoms during the exchange processes, the MEM predicts a certain variation in the volume dependences of the various J_x, as shown by first-principles calculations [4]. However, all physical quantities X measured so far show a remarkably similar volume dependence, which suggests unexpectedly similar volume dependences for all J_x. X includes the Weiss temperature Θ, $(e_2)^{1/2}$, the Néel temperature T_N measured in low fields, etc. [9–13]. Here e_2 is the coefficient of the leading term in the high-temperature series expansion of the free energy in the paramagnetic phase (PP). These molar volume (V) dependences are represented empirically by a simple power law with a Grüneisen parameter $\Gamma \equiv d\ln X/d\ln V = 18 \pm 1.5$. If all $\Gamma(X)$ were almost identical, some additional physics, such as *local melting* [2], dominating all the exchange processes might be hidden and the current MEM should be reexamined from that microscopic point of view.

The magnetic phase diagram, especially the LFP-HFP transition field B_{c1}, is expected to be extremely sensitive to small differences in $\Gamma(J_x)$ [2]. This is because B_{c1} is determined by a delicate energy balance between the two almost degenerate phases with completely different spin symmetries. In this Letter, we present the first experimental study of the volume dependence of the zero-temperature lower critical field $B_{c1}(0)$ in bcc ^{3}He. We find that isochoric pressure measurements down to 120 μK show $\Gamma\{B_{c1}(0)\} = 14.5$, which is substantially smaller than the ordinary Grüneisen parameter (≈ 18). This finding is consistent with the prediction of the MEM and removes the long-standing question about the applicability of this model.

The details of our experimental setup will be presented elsewhere. Briefly, a sample cell and a platinum-wire NMR thermometer are directly mounted on the nuclear stage of a copper nuclear-demagnetization refrigerator [14]. At temperatures higher than 1.7 mK, we have employed a ^{3}He melting-curve thermometer based on the new T scale [15]. It was also used to calibrate the Pt thermometer in the same temperature range. Solid ^{3}He samples were formed in a thin disk-shaped space (8 mm diameter and 0.1 mm thickness) between a sintered-powder heat exchanger and a silicon-silver diaphragm for a capacitive strain-gauge [16]. The heat exchanger (8 mm diameter and 1.5 mm thickness) consists of fine silver powder whose average particle diameter is 1100 Å after sintering (5.1 m^2/g). A pressure change of the "bulk" solid in the open space is monitored with a resolution of 10 μbar. It is believed that most of the solid within the fine matrix of the sinter hardly moves and transmits any

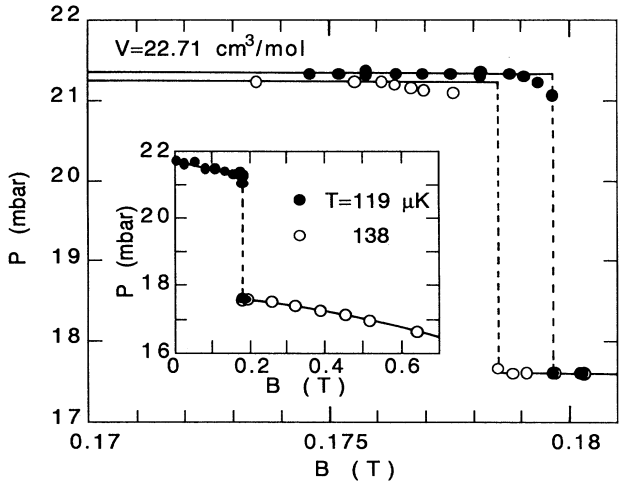

FIG. 1. Pressure changes of bcc ^{3}He as a function of magnetic field near the LFP-HFP transition. Inset: Overall pressure change in a wider field range.

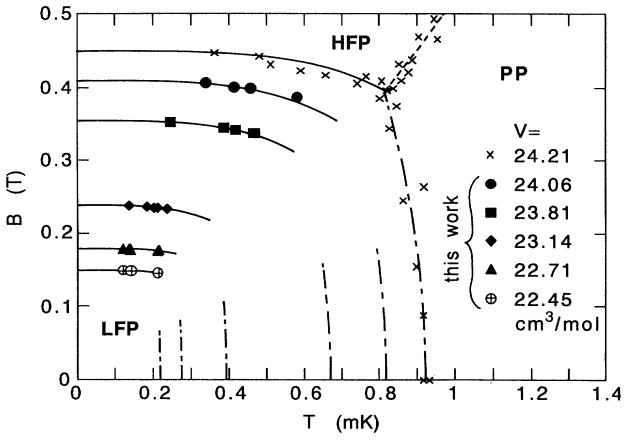

FIG. 2. The LFP-HFP transition lines in the magnetic phase diagram of bcc ^{3}He. The vertical dash-dotted lines represent the LFP-PP first-order transitions estimated from Ref. [10]. The data for the melting density (24.21 cm^3/mol) are from Refs. [6], [15], and [19–21].

pressure change to the diaphragm [12]. A volume correction necessary to obtain a truly constant-volume pressure change for our cell design is calculated to be less than 5%. Thus we made no correction here to the raw data. Such a small correction factor has been verified over a wide temperature range through T_N by comparing our $B=0$ data with those taken under nearly constant-volume conditions by other workers [11,12]. A small shielded superconducting magnet [17] with a special counterwinding and a niobium tube produces well-confined and homogeneous fields up to 0.64 T over the sample cell. The field strength was determined with an accuracy of better than 0.3% from simultaneous NMR measurements on solid ^{3}He located just below the bulk solid for the pressure measurements.

The solid sample (with ^{4}He of less than 3 ppm) was carefully prepared to obtain a uniform density distribution throughout the whole solid both inside and outside the sintered powder. At a temperature less than 10 mK below the melting point, it was annealed over night to remove any crystalline imperfections such as dislocations. The molar volume of the resultant solid was determined from the relation [18] $V=45.493P_0^{-0.1780}$. Here V is in cm^3/mol and P_0 is in bars. The value of P_0 was determined by fitting $B=0$ data in the range $30T_N \leq T \leq 100$ mK, where the pressure change is entirely dominated by the exchange interactions, with the expression $P/R=P_0/R+(de_2/dV)/8T$.

Figure 1 shows measured pressure changes from P_0 as a function of magnetic field near the LFP-HFP transitions for a 22.71-cm^3/mol sample. The data were taken in complete thermal equilibrium after changing the field stepwise at fixed temperatures of 119 and 138 μK. When the field is swept up through B_{c1} only by a narrow interval less than 10 Oe, the pressure drops abruptly by about 3.7 mbar. The sharp nature of the transition reveals that the LFP-HFP transition at such a high density is still clearly first order, as is the case at the melting one [19]. If we attribute the observed minimum transition width ($\approx$ a few Oe) to a residual density inhomogeneity in the bulk solid, it is estimated to be less than 0.01 cm^3/mol using the measured volume dependence of $B_{c1}(0)$ (see below). As the temperature increases, B_{c1} and the pressure drop ΔP_{c1} decrease, while the transition width seems to increase. A slight decrease in pressure just below B_{c1}, which was absent on the high-field side, was neglected in the determination of ΔP_{c1}. Below 0.64 T we observed no traces of extra phase transitions other than the LFP-HFP one, as shown in the inset of Fig. 1, at all the volumes studied.

Figure 2 shows measured temperature variations of B_{c1} at six different volumes between 22.45 and 24.06 cm^3/mol. From the antiferromagnetic spin-wave theory, B_{c1} is expected to behave as $B_{c1}(T)=B_{c1}(0)+aT^4$ at temperatures well below the LFP-PP and HFP-PP transitions. The solid lines in the figure are least-squares fits of this form to the experimental data. Because the $B_{c1}(T)$ lines are almost flat in the measured temperature ranges, we can extrapolate the data to $T=0$ with good accuracy.

The $B_{c1}(0)$ value determined from the above procedure is plotted as a function of volume in Fig. 3. The volume dependence can be fitted quite well by the simple power law

$$B_{c1}(0)=0.4516(V/24.21)^{14.5\pm0.3}. \quad (1)$$

Here, $B_{c1}(0)$ is in tesla and V is in cm^3/mol. The Grüneisen parameter ($=14.5\pm0.3$) is exceptionally small compared to common numbers for other physical quantities associated with the exchange interactions. The $B_{c1}(0)$ value ($=0.4516$ T) extrapolated to the melting

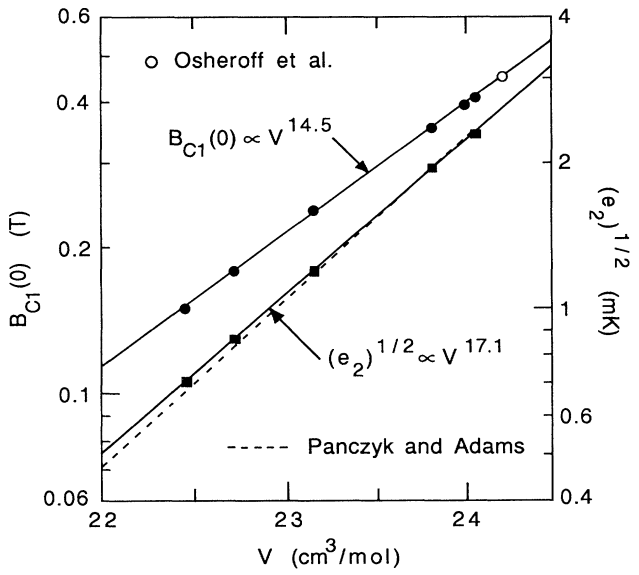

FIG. 3. Volume dependences of $B_{c1}(0)$ and the square root of e_2. The open circle is from Ref. [19] and the dotted line is from Ref. [11]. Note that the horizontal axis for V is not a linear scale but a logarithmic one.

density, 24.21 cm^3/mol, is in excellent agreement with the result ($=0.4513 \pm 0.0005$ T) of Osheroff, Godfrin, and Ruel [19]. In the figure, our $(e_2)^{1/2}$ data determined in the same manner as P_0 are also plotted, along with those of Panczyk and Adams [11]. The two measurements agree with 5% indicating the *normal* Grüneisen parameter to be 17–18.

The zero-temperature pressure jump $\Delta P_{c1}(0)$ and its volume dependence is determined in the same manner as $B_{c1}(0)$. The result is

$$\Delta P_{c1}(0) = -8.93(V/24.21)^{13.5 \pm 0.4}, \quad (2)$$

where $\Delta P_{c1}(0)$ is in mbar. The Grüneisen parameter is again unusually small but satisfies a necessary condition, $\Gamma\{B_{c1}(0)\} = \Gamma\{\Delta P_{c1}(0)\} + 1$, expected from a thermodynamic argument. Using the Clausius-Clapeyron equation, we can deduce the zero-temperature magnetization jump $\Delta M_{c1}(0)$ at $B_{c1}(0)$ from the measured $\Delta P_{c1}(0)$ and the volume dependence of $B_{c1}(0)$:

$$\Delta M_{c1}(0) = -\frac{\Delta P_{c1}(0)}{dB_{c1}(0)/dV}.$$

Substituting Eqs. (1) and (2) into the above equation, we have

$$\Delta M_{c1}(0) = 0.511 M_{\rm sat}(V/24.21)^{0.0 \pm 0.7},$$

where $M_{\rm sat}$ is the saturation magnetization. $\Delta M_{c1}(0)$ is, thus, essentially volume independent within the experimental accuracy. The large magnetization jump, exceeding a half of $M_{\rm sat}$, indicates that the HFP has a considerable ferromagnetic tendency. The deduced $\Delta M_{c1}(0)$ value ($=0.511 M_{\rm sat}$) is in excellent agreement with the result ($=0.508 M_{\rm sat}$) of Osheroff, Godfrin, and Ruel [19], who measured the melting-pressure depression with field. The remarkable consistency between the two independent determinations manifests the validity both of the measurements and then of the small $\Gamma\{B_{c1}(0)\}$ obtained in this work.

The present experiment is the first definitive observation showing that the volume dependences of the magnetic properties in bcc ^{3}He cannot be scaled with a single parameter. In terms of the MEM, this means that the different J_x must have different volume dependences [22]. At present, it is widely anticipated that there are three predominant exchanges in bcc ^{3}He, i.e., the nearest-neighbor two-spin exchange J_{1N}, the most compact three-spin exchange T_1, and the planar four-spin ring exchange K_P [4,5,8]. If we restrict ourselves to the three-parameter model, taking into account only J_{1N}, T_1, and K_P, these frequencies can be determined under three experimental constraints at any volume and then the Grüneisen parameters can be deduced [23]. We computed exchange parameters which satisfy the present $B_{c1}(0)$ data, cf. Eq. (1), as well as $\Theta = -1.6(V/24.21)^{17 \pm 1}$ mK and $(e_2)^{1/2} = 2.62(V/24.21)^{17.6 \pm 0.5}$ mK as was done by Stipdonk and Hetherington [8]. The last two constraints are mean values among several experiments [9–11] including this work. The results are

$$\Gamma(J_{1N}) = 19.7 \pm 1.0, \quad \Gamma(T_1) = 18.0 \pm 0.3, \quad \Gamma(K_P) = 17.3 \pm 0.4 \quad (3)$$

between 22.45 and 24.06 cm^3/mol. Apparently, the low value of $\Gamma\{B_{c1}(0)\}$ demands a stronger volume dependence for J_{1N} than T_1 and K_P. This seems to be reasonable because the hard-core radius becomes less important in determining the exchange frequencies at larger volumes and the ordinary two-spin exchange would be dominant there. It would be valuable to improve quantitatively this analysis by using more sophisticated techniques [24,25], which take into account quantum fluctuations, in the calculation of $B_{c1}(0)$ instead of using the mean-field approximation employed here. Nevertheless, the present semiquantitative conclusion, i.e., $\Gamma(J_{1N}) > \Gamma(T_1) \gtrsim \Gamma(K_P)$, should be correct even without exact knowledge of the quantum corrections to the antiferromagnetic ground-state energies [24].

Among the theoretical calculations of $\Gamma(J_x)$ based on first principles, Roger [4] predicted $\Gamma(J_{1N}) = 19.5$, $\Gamma(T_1) = 17.4$, and $\Gamma(K_P) = 16.2$ in the present volume range with the high-density WKB approximation taking account of the dynamical displacement of $N \approx 16$ atoms during the exchange processes. These values of $\Gamma(J_x)$ are consistent with Eq. (3), and the low $\Gamma\{B_{c1}(0)\}$ value has been predicted based on them [25]. In his calculation, the large $\Gamma(J_{1N})$ is interpreted as the result of a larger tunneling barrier potential than that for T_1 or K_P. On the other hand, another WKB calculation [26] consider-

ing smaller numbers of moving atoms predicts $\Gamma(J_{1N}) = 21.4$, $\Gamma(T_1) = 23.1$, and $\Gamma(K_P) = 21.7$, which do not agree with our result. For further testing of the MEM, it is highly desirable to improve the precision of computational works such as the path-integral Monte Carlo calculation [5] so that the calculated $\Gamma(J_X)$ can be compared with our experiment.

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