

## Novel Susceptibility of bcc Solid $^3\text{He}$ through the Nuclear-Ordering Temperature

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We measured for the first time simultaneously the ac susceptibility and the static magnetization of high-density bcc solid  $^3\text{He}$  through the nuclear-ordering transition. We discovered a sharp peak in the real part of the ac susceptibility and an abrupt depression in the imaginary part near the transition temperature. The transition temperature indicated by the ac susceptibility is higher than that indicated by static magnetization. We interpreted the novel behavior in the susceptibility in terms of the spin relaxation between the Zeeman system and the exchange system.

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Nuclear magnetism of solid  $^3\text{He}$  is due to a unique direct exchange of atoms which have large zero-point motion. Previous studies of entropy [1], magnetization [2], pressure [3], specific heat [4], and NMR [5] have established that the nuclear ordering of bcc solid  $^3\text{He}$  is a first-order phase transition. The direct exchange interaction strongly depends on the spacing between atoms or density of solid  $^3\text{He}$ . By the use of a new technique we shed light on the phase transition of bcc solid  $^3\text{He}$  having high density and low ordering temperatures.

In this Letter, we report on the first measurement of the ac magnetic susceptibility of bcc solid  $^3\text{He}$  at low frequencies. The experimental apparatus and method are the same as our previous work [6,7]. Briefly, our unique hybrid sample cell with a superconducting quantum interference device magnetometer allows us to measure ac susceptibility and magnetization simultaneously without excess heating at  $\mu\text{K}$  temperatures. The sample of bcc solid  $^3\text{He}$  contained in the pores of a silver sponge was cooled with a double-stage nuclear demagnetization apparatus, capable of cooling samples down to  $44 \mu\text{K}$ . An oscillating magnetic field ( $0.13 \mu\text{T}_{\text{rms}}$ ) at a variable frequency from 16 to 33 Hz was applied to the sample parallel to the static field of 1.9 mT. Subsequent to attaining minimum temperature, the measurements were performed by raising the temperature in stepwise fashion using externally applied heat pulses. The warmup around the transition temperature, however, was allowed to occur with only the residual heat leak ( $\sim 0.5 \text{ nW}$ ). Thermometry was provided by platinum NMR thermometers which were calibrated against a  $^3\text{He}$  melting curve thermometer. The precision of thermometry is 2%.

Figure 1 shows the real part  $\chi'$  of the susceptibility at frequency 19 Hz for molar volumes 21.51, 20.97, and  $20.58 \text{ cm}^3$  as a function of temperature. The imaginary part  $\chi''$  of the susceptibility is also shown for only the molar volume of  $20.97 \text{ cm}^3$ . The displayed susceptibility is the net susceptibility which was computed by subtracting the background susceptibility from the total susceptibility. One series of runs at a given molar volume takes typically one month to complete the temperature sweep. The noise in Fig. 1 arises mainly from temperature measure-

ment. In the high-temperature paramagnetic region, the behavior of the ac susceptibility is similar to the static magnetization (not shown). In marked contrast to the paramagnetic regions, a sharp peak in  $\chi'$  and an abrupt depression in  $\chi''$  occur for all the investigated samples. We identify the newly discovered features as the paramagnetic- to ordered-state transition of bcc solid  $^3\text{He}$ . The peaks in  $\chi'$  appear with both increasing and decreasing temperature. For a given sample and frequency the height of the peaks is independent of (1) the rate of temperature change from  $+0.03$  to  $-0.06 \mu\text{K/h}$  and (2) the amplitude of the oscillating field from 0.13 to  $0.26 \mu\text{T}_{\text{rms}}$ . The frequency of the oscillating field, on the other hand, produces large changes in  $\chi'$  and  $\chi''$ .

When we examine the paramagnetic- to ordered-state transition region in detail, we observe that the signature of transition occurs at a temperature higher in suscepti-

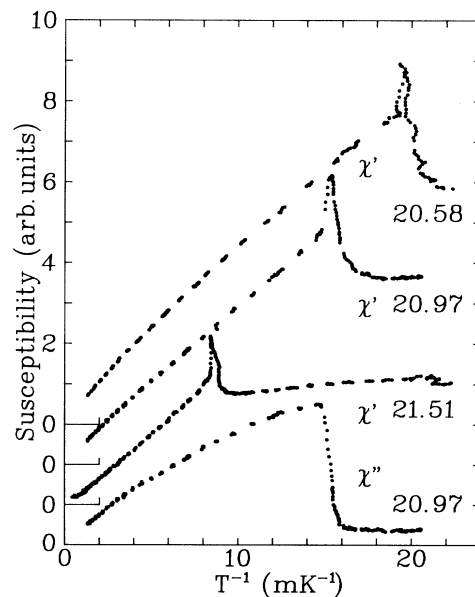


FIG. 1. Real and imaginary parts of the ac susceptibility  $\chi'$  and  $\chi''$ , respectively, as a function of inverse temperature. The numbers beside the plots indicate molar volume.

bility than in magnetization. Figure 2 shows an expanded plot near the transition region for the sample of 20.97 cm<sup>3</sup>/mol. With decreasing temperature in Fig. 2, the transition of  $\chi'$  and  $\chi''$  begins to occur at point *A* where the temperature is higher than the indication of the beginning point *B* of the magnetization. The difference between temperatures indicated by points *A* and *B* is 1.7, 2.0, and 0.4  $\mu$ K for molar volumes of 21.51, 20.97, and 20.58 cm<sup>3</sup>, respectively. The assertion that the temperature of the <sup>3</sup>He spin indicated by  $\chi'$  and  $\chi''$  is equal to the temperature of the <sup>3</sup>He spin indicated by the magnetization  $M_s$  can be supported by the following argument. We analyze  $\chi'$  and  $\chi''$  using the Casimir-Du Pre theory [8] to find the susceptibility at zero frequency  $\chi_0$ . We find that  $\chi_0$  derived from measured  $\chi'$  and  $\chi''$  is very similar to the observed magnetization  $M_s$  over the paramagnetic and transition region as shown in Fig. 2. We regard the similarity in detail between  $\chi_0$  and  $M_s$  as evidence for our assertion that magnetization and susceptibility measurements are made at the same temperature.

The analysis of susceptibility based on the thermodynamic theory of Casimir and Du Pre is as follows. The ac susceptibility is regarded as the response of the nuclear spins to the oscillating field parallel to the static field. When an interaction exists between two thermodynamic-equilibrium systems with an associated relaxation time  $\tau$ , the ac susceptibility is written

$$\chi' = \frac{\chi_0 - \chi_s}{1 + \omega^2 \tau^2} + \chi_s, \quad (1)$$

$$\chi'' = \frac{(\chi_0 - \chi_s)\omega\tau}{1 + \omega^2 \tau^2}, \quad (2)$$

where  $\omega$  is the angular frequency of the oscillating field and  $\chi_0$  and  $\chi_s$  are the isothermal and the adiabatic susceptibilities, respectively. In our case the energy absorbed in the Zeeman system by the oscillating field is transferred to the exchange system and then to the sample cell. The latter energy transfer is related to the Kapitza thermal boundary resistance between the solid <sup>3</sup>He and the cell wall. Since the measured relaxation time associated with this boundary resistance is several thousand seconds, the important relaxation process in the present frequency of a few tens of Hz is the Zeeman-exchange relaxation. The measured frequency dependence of  $\chi'$  and  $\chi''$  is in good agreement with Eqs. (1) and (2) with  $\chi_s = 0$  at temperatures for the paramagnetic state, as shown typically in Fig. 3(a). This fact justifies the use of the theory of Casimir and Du Pre. On the other hand, by measuring  $\chi'$  and  $\chi''$  as a function of temperature at a fixed frequency, 19 Hz, we were able to obtain the temperature dependence of  $\chi_0$  and  $\tau$  as shown in Fig. 2, using the relations  $\chi_0 = (\chi'^2 + \chi''^2)/\chi'$  and  $\tau = \chi''/\omega\chi'$ . These relations were derived from Eqs. (1) and (2) with  $\chi_s = 0$ .

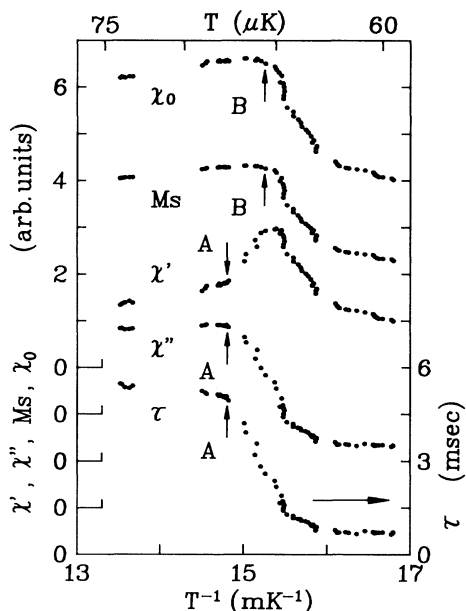


FIG. 2. An expanded plot near the transition region for the real and imaginary parts of the ac susceptibility,  $\chi'$  and  $\chi''$ , respectively, static magnetization  $M_s$ , susceptibility at zero frequency  $\chi_0$ , and the relaxation time  $\tau$  for the molar volume of 20.97 cm<sup>3</sup>. Arrow *A* shows the beginnings of the transition for  $\chi'$ ,  $\chi''$ , and  $\tau$ . Arrow *B* shows those for  $\chi_0$  and  $M_s$ . Note that zeros for  $\chi'$ ,  $\chi''$ ,  $M_s$ , and  $\chi_0$  are shifted in that order along the ordinate.

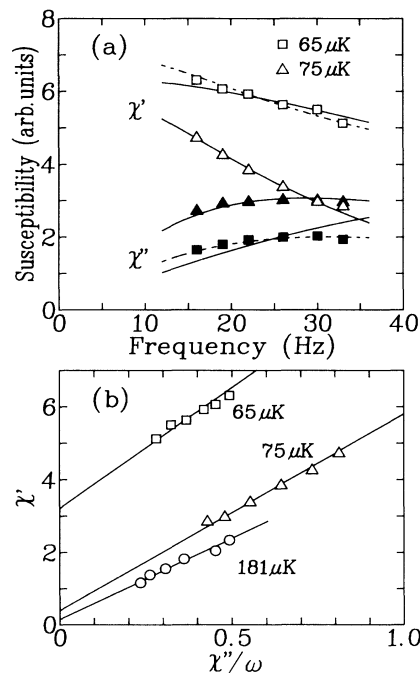


FIG. 3. (a) Susceptibility vs frequency for the molar volume 20.97 cm<sup>3</sup>. Solid lines show fits of Eqs. (1) and (2) with adiabatic susceptibility  $\chi_s = 0$ . Dashed lines show those with a finite  $\chi_s$ . (b) Real susceptibility vs imaginary susceptibility divided by frequency  $\omega$  for the molar volume of 20.97 cm<sup>3</sup>. The intercept indicates adiabatic susceptibility  $\chi_s$  and the slope an inverse of the relaxation time  $\tau$ .

The behavior of  $\chi_0$  obtained in this manner turned out to agree with that of  $M_s$ , although  $\chi_0$  was derived independently of  $M_s$ .

Detailed examination of the fit in Fig. 3(a) reveals that the existence of a nonzero  $\chi_s$  in the ordered state gives a better fit for all of the investigated molar volumes. This tendency can be seen more clearly in the plot of  $\chi'$  vs  $\chi''/\omega$  at each temperature, as shown in Fig. 3(b). The intercept and slope of the line yield  $\chi_s$  and inverse  $\tau$ , respectively. For all samples  $\chi_s$  in the paramagnetic state is nearly zero, and  $\chi_s$  in the ordered state has a nonzero value of approximately  $0.7\chi_0$ . We derived the temperature variation of  $\chi_s$  and  $\tau$  in the ordered state as follows. We observed that  $\chi_0$  is exactly proportional to  $M_s$  in the paramagnetic state. Therefore, we assume  $\chi_0$  is also proportional to  $M_s$  in the ordered state with the same proportionality ratio as in the paramagnetic state.  $\chi_s$  and  $\tau$  obtained for the ordered state are shown in Fig. 4.

The value of  $\tau$  in the paramagnetic state is in approximate agreement with the  $T_2$  value in the high-temperature NMR [9]. The relaxation time  $\tau$  is considered to be equal to the spin-spin relaxation time  $T_2$  if the Zeeman energy is much smaller than the exchange energy.  $T_2$  in the paramagnetic state is expressed in terms of the exchange energy  $J$  as  $J = 1.258\tau(\gamma^2\hbar/a^3)^2$ , where  $\gamma$  is the gyromagnetic ratio of  $^3\text{He}$  and  $a$  is the lattice constant [10]. Thus  $\tau$  in the paramagnetic state is nearly constant with temperature. In the first-order phase transition from the paramagnetic to the antiferromagnetic state, a drastic change in spin configuration occurs. Therefore it is possible that the relaxation time and susceptibility drastically change at the transition as in Fig. 4. On the other hand, in the second-order phase transition of ferromagnetism,

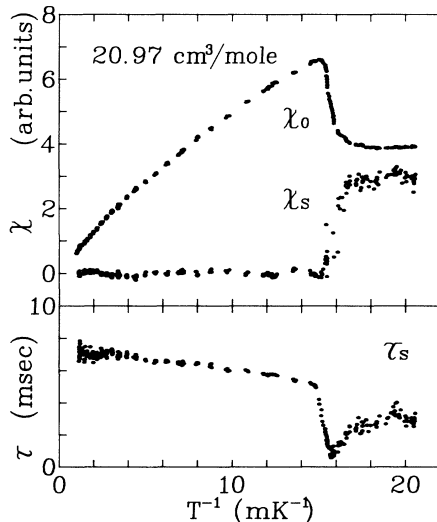


FIG. 4. Isothermal susceptibility  $\chi_0$ , adiabatic susceptibility  $\chi_s$ , and relaxation time  $\tau$  vs inverse temperature for the molar volume of  $20.97 \text{ cm}^3$ .

the spin configuration in a magnetic field changes gradually in the same direction, and as a result the relaxation time changes continuously as shown by the behavior near the transition in the hcp phase of solid  $^3\text{He}$  [7].

In the paramagnetic state  $\chi_s$  is nearly zero from the experimental results.  $\chi_s$  is expressed as  $\chi_s = (C_M/C_H)\chi_0 = \chi_0 - T(\partial M/\partial T)_H^2/C_H$  in terms of the specific heat at constant magnetization and constant field,  $C_M$  and  $C_H$ , respectively. Thus  $\chi_0$  is nearly zero if  $M$  has a Curie-Weiss susceptibility. On the other hand, the finite  $\chi_s$  nearly equal to  $\chi_0$  comes from the fact that the magnetization in the antiferromagnetic state is approximately constant with temperature. Note that at absolute zero,  $\chi_s$  should approach  $\chi_0$  according to the above equation.

Since  $\chi_s$  turns out to be finite for the present range of frequency between 16 and 33 Hz, there should be further depression of  $\chi'$  above 33 Hz. This indicates that in the ordered state there occurs another path of the energy flow from the nuclear spins excited by an oscillating field to the exchange energy, i.e., the reservoir of the spin waves at higher frequencies. The energy absorbed by  $^3\text{He}$  nuclear spins,  $Q$ , is estimated as  $1 \times 10^{-16} \text{ W/cm}^3$  from the expression  $Q = \omega\chi''H_1^2/2\mu_0$ , where  $H_1$  is the amplitude of the oscillating field and  $\mu_0$  is the magnetic permeability of vacuum. The heat capacity of the spin wave,  $C$ , is estimated to be  $10^{-2} \text{ J/Kcm}^3$  from the expression  $C = (4\pi^2/15)k_B(k_B T/\hbar)^3/v_s^3$  [11], where  $v_s$  is the spin-wave velocity which is proportional to the exchange energy or the ordering temperature. To raise the temperature of the spin-wave reservoir, for example, by  $1 \mu\text{K}$ , would take approximately 1000 d. Thus the spin wave is regarded as a thermal reservoir in this mechanism.

The relaxation time at higher frequencies can be estimated in two ways. The disturbance of the nuclear spin is transferred through a dipole-dipole interaction to the spin-wave reservoir in a time constant determined by the spin-wave velocity and the pore dimension. This time constant  $\tau_A$  is expressed as  $\tau_A = L/v_s(E_{ex}/E_d)^{1/2}$  [12], where  $L$  is the pore diameter of approximately  $2800 \text{ \AA}$ ,  $v_s$  is  $0.8 \text{ cm/sec}$ , and  $E_{ex}$  and  $E_d$  are the exchange energy  $100 \mu\text{K}$  and the dipole energy  $1 \mu\text{K}$ , respectively, for the sample of  $20.97 \text{ cm}^3/\text{mol}$ . From this expression, we get  $\tau_A = 0.1 \text{ msec}$ . Second, according to the theory which calculates the spin relaxation rates due to the three- and four-magnon processes in the  $u2d2$  phase [13], the relaxation rate for the present frequency is 3 to 6 kHz depending on the mode, temperature, and angle between the l-vector and the static magnetic field. This indicates that the relaxation time is on the order of  $0.1 \text{ msec}$ . The above two time constants are in agreement with the time constant suggested to exist at higher frequencies than in the present experiment. Thus the finite value of  $\chi_s$  and the possible smaller time constant are related to the nature of the ordered state.

Here, we remark that the measured susceptibility and relaxation times are a bulk property. The reason is three-

fold. First, the measured relaxation times are in agreement with the values which were obtained in NMR measurements for the bulk solid  $^3\text{He}$  at higher temperatures [9]. Second, the molar volume dependence of the ordering temperature  $T_N$  which was determined from the peaks of the real susceptibility indicates  $T_N \propto V^{2/3}$  [14]. This is compared with the exchange energy  $J \propto V^{-1/3}$  obtained from  $T_1$  and  $T_2$  measurements for the bulk solid  $^3\text{He}$  [9]. Third, the phenomenon associated with the surface in electronic materials and superconductive substances often results from the shielding of electronic charges at the surface. However, in this experiment of solid  $^3\text{He}$ , such a situation does not occur.

Thus we analyzed the novel phenomenon of susceptibility of solid  $^3\text{He}$  in terms of the Casimir-Du Pre theory. However, this phenomenon should be analyzed from first principles in terms of microscopic theory. A naive picture shows that novel phenomena should occur because of  $\omega\tau \approx 1$  in the paramagnetic and transition regions. In the ordered state there is almost no loss because of  $\chi_s \approx \chi_0$ , which suggests a possible higher relaxation rate due to the spin-wave reservoir.

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- [1] W. P. Halperin, C. N. Archie, F. B. Rasmussen, R. A. Buhrman, and R. C. Richardson, Phys. Rev. Lett. **32**, 927 (1974). References [1] to [5] show the first observation for each kind of measurement.
- [2] T. C. Prewitt and J. M. Goodkind, Phys. Rev. Lett. **39**, 1283 (1977).
- [3] T. Mamiya, A. Sawada, H. Fukuyama, Y. Hirao, K. Iwahashi, and Y. Masuda, Phys. Rev. Lett. **47**, 1304 (1981).
- [4] A. Sawada, M. Kato, H. Yano, K. Iwahashi, and Y. Masuda, Phys. Rev. Lett. **56**, 1587 (1986).
- [5] D. D. Osheroff, M. C. Cross, and D. S. Fisher, Phys. Rev. Lett. **44**, 792 (1980).
- [6] H. Yano, T. Uchiyama, T. Kato, Y. Minamide, S. Inoue, Y. Miura, and T. Mamiya, J. Low Temp. Phys. **78**, 165 (1990).
- [7] H. Yano, H. Kondo, T. Suzuki, Y. Minamide, T. Kato, Y. Miura, and T. Mamiya, Phys. Rev. Lett. **65**, 3401 (1990).
- [8] H. B. G. Casimir and F. K. Du Pre, Physica (Utrecht) **5**, 507 (1938).
- [9] R. A. Guyer, R. C. Richardson, and L. I. Zane, Rev. Mod. Phys. **43**, 532 (1971).
- [10] A. Abragam and M. Goldman, *Nuclear Magnetism: Order and Disorder* (Oxford Univ. Press, New York, 1982).
- [11] D. D. Osheroff and C. Yu, Phys. Lett. **77A**, 458 (1980).
- [12] T. Kusumoto, O. Ishikawa, T. Mizusaki, and A. Hirai, J. Low Temp. Phys. **59**, 269 (1985).
- [13] T. Ohmi and M. Tsubota, J. Low Temp. Phys. **83**, 177 (1991).
- [14] S. Abe, T. Takeuchi, H. Kondo, T. Suzuki, H. Yano, and T. Mamiya (to be published).