Magnetization and ac Susceptibility of hcp Solid ³He near the Nuclear Ordering Temperature

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The magnetization and the ac susceptibility of hcp solid ³He have been measured for molar volumes V from 18.34 to 19.29 cm³/mole at temperatures down to 38 μK. The Weiss temperature obtained from the magnetization is proportional to $V^{18.4 \pm 0.6}$. The Zeeman-exchange relaxation time obtained from the ac susceptibility indicates the appearance of the critical phenomenon for the sample with 19.29 cm³/mole.

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Experimental studies of hcp solid ³He have been made by NMR relaxation-time measurements[1] at temperatures near 1 K and by magnetization measurements by Takano *et al.*[2] below 0.4 mK. Takano *et al.* have obtained the Weiss temperature and have indicated that the dominant spin interactions are ferromagnetic. Their result is consistent with the theory.[3] However, there is an inconsistency in the molar-volume derivative of the spin interaction obtained between the NMR and the magnetization measurements. In order to resolve this inconsistency, we have measured the static magnetization and the ac susceptibility in the temperature range from 38 μK to 6 mK. The Weiss temperature was obtained from the magnetization at molar volumes from 18.34 to 19.29 cm³/mole, and the coefficient of the next-higher-order term in the high-temperature expansion was obtained at 19.29 cm³/mole. The Zeeman-exchange relaxation time was also obtained from the ac susceptibility.

Our main idea is as follows: Both the static magnetization and the ac susceptibility are measured with a SQUID magnetometer at the same time. Because the SQUID magnetometer measures only flux changes, the static-magnetization measurements are subject to flux jumps caused by electric and mechanical disturbances. On the other hand, the ac susceptibility is stable and reproducible before and after the disturbances.[4] One can correct the flux jumps using values of the ac susceptibility, which enables us to measure the static magnetization accurately over a wide temperature range.

The solid ³He sample was contained in the pores of a sintered-silver sponge and was cooled with a double-stage nuclear demagnetization refrigerator of PrNi₅ and copper in series. The sample was in a constant magnetic field of 1.9 mT trapped with a titanium superconducting cylinder. The ac susceptibility was interpreted from the mutual inductance between the primary coil and the secondary coil of an astatic pair on the cell. The mutual inductance was measured by an alternating field of 0.13 μT (rms) at frequencies from 16 to 33 Hz in parallel with the constant field. The static magnetization was interpreted from the static flux change in the secondary coil. The sample cell design and the measurement tech-

nique have been described elsewhere.[4] The pressures of the sample were monitored with a capacitive strain gauge on the copper nuclear stage. The molar volumes were determined from the hcp-bcc phase transformation pressure or the melting pressure of hcp ³He.[5] The uncertainty in the determination of the molar volumes was ± 0.01 cm³/mole.

The measurements were made with gradually increasing temperature, after the samples were cooled down to the lowest temperature. We determined the thermal relaxation time between the sample and the cell by observing the magnetization change after a heat pulse was applied. The relaxation time was proportional to T^{-3}, and was 2900 sec at 60 μK and 620 sec at 100 μK for 19.29 cm³/mole. Temperatures were measured with a platinum pulsed NMR thermometer[6] at a frequency of 125 kHz at time intervals of 10 times the longitudinal relaxation time of platinum. The NMR thermometer was calibrated against a ³He-melting-curve thermometer from 0.9 to 3 mK using the temperature scale of Fukuyama *et al.*[7] The ³He-melting-curve thermometer also provided thermometry above 0.9 mK. With the NMR thermometer the temperature was determined with a precision of 2%.

We obtained the magnetization and the susceptibility of hcp ³He by subtracting the empty-cell data from those with the sample present. The empty-cell data were obtained in the temperature range from 40 μK to 20 mK. They obey the Curie law, and the magnetization change over the measurement temperature range is 4% of that with the sample. This change may be explained by the copper nuclear magnetization in the cell body. The arbitrary zero in the magnetization of the sample was determined by extrapolation of the data above 1 mK where the magnetization obeys the Curie law.

As shown in Fig. 1, the magnetization of hcp ³He obeys the Curie-Weiss law for all molar volumes except 19.29 cm³/mole. In the inset to Fig. 1, we show the deviation of the magnetization for 19.29 cm³/mole from the Curie-Weiss equation obtained by fitting the data above 50 μK. The magnetization deviates below 50 μK by several times as much as the standard deviation. In or-
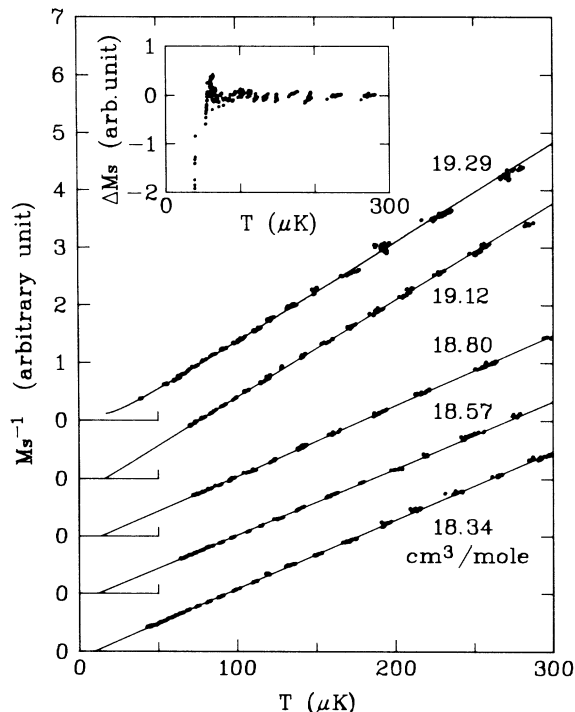
FIG. 1. Inverse magnetization of hcp solid ^{3}He as a function of temperature. The solid curves are the fits by Eq. (1) with the first three terms for 19.29 cm^3/mole and with the first two terms for other molar volumes. Inset: Deviation of the magnetization for 19.29 cm^3/mole from the Curie-Weiss equation obtained by fitting the data above 50 μK.

der to explain this deviation, we introduce the third term of the high-temperature series expansion of the magnetization M_s,

$$(M_s/H)^{-1}=C^{-1}(T-\theta+B/T+\cdots),\qquad(1)$$

where H is the applied magnetic field, C is the Curie constant, and θ is the Weiss temperature. Consequently, we can obtain θ and B by fitting the data with the first three terms of Eq. (1). We also obtain θ from 18.34 to 19.12 cm^3/mole by fitting the data with the first two terms of Eq. (1). The values of θ and B so obtained are listed in Table I. Incidentally, the magnetization data could not be fitted by Eq. (1) over all the measurement temperatures, because the data undulated several percent at 0.6 mK. Hence, the temperature ranges for fitting are shown in Table I.

The Weiss temperature θ as a function of molar volume is shown in Fig. 2. The molar-volume dependence of θ is assumed to obey a power law

$$\theta=A(V/19)^{\gamma},\qquad(2)$$

where V is the molar volume. The result of this fit is that A and γ are 16.3 ± 0.7 μK and 16.1 ± 2.3, respectively. Figure 2 shows that θ at 19.12 cm^3/mole is small
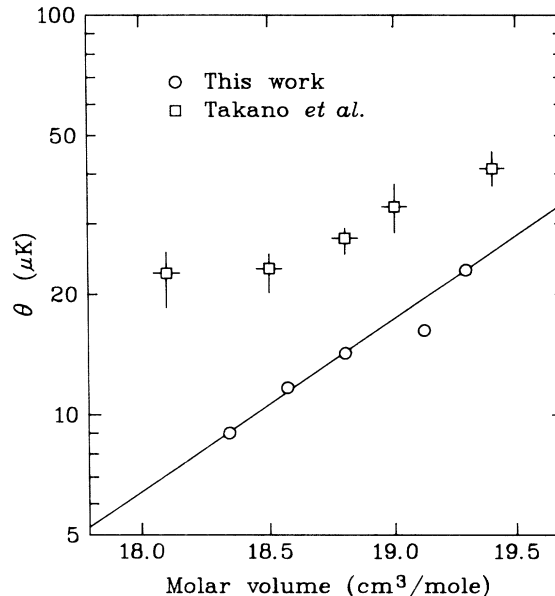

FIG. 2. Weiss temperature of hcp solid ^{3}He as a function of molar volume: circles, this work; squares, Ref. 2. Errors for our data are estimated to be 0.2 to 0.6 μK. The solid curve is the fit by a power law $V^{18.4}$.

compared with the line fitted for θ at other molar volumes. The value of θ at 19.12 cm^3/mole is thought to be underestimated, because the data for 19.12 cm^3/mole were fitted by the simple Curie-Weiss equation in a narrower temperature range than that used for other molar volumes, as shown in Table I. Fitting θ for all molar volumes except 19.12 cm^3/mole by Eq. (2), we conclude $A=17.4\pm0.2$ μK and $\gamma=18.4\pm0.6$. The Weiss temperature obtained by Takano et al.[2] is also shown in Fig. 2. Our values of θ are half of their values, and our result for γ is 1.7 times larger than their result $\gamma=11.1\pm1.0$. In contrast to our result, their value of θ deviates from the power law as the molar volume decreases. Our result for γ is in good agreement with $\gamma=20$ of the effective exchange frequency obtained from NMR relaxation measurements.[1]

According to the theory of Roger, Suaudeau, and Bernier,[8] the exchange frequencies of hcp ^{3}He can be es-

TABLE I. Values of θ and B obtained at several molar volumes.

V (cm^3/mole)	Temperature range of the fit (μK)	θ (μK)	B (μK^2)
18.34	45–340	9.0±0.2	
18.57	64–400	11.7±0.3	
18.80	71–310	14.2±0.2	
19.12	70–230	16.2±0.4	
19.29	38–240	23.0±0.6	211±35

timated from θ and B by

$$\theta = 3\tilde{J}_1 + 3\tilde{J}_2 + 9K , \qquad (3)$$

$$B = 3\tilde{J}_1^2 + 3\tilde{J}_2^2 + \tfrac{3}{2}\tilde{J}_1 K + \tfrac{3}{2}\tilde{J}_2 K + \tfrac{27}{4} K^2 . \qquad (4)$$

Here $\tilde{J}_1$ and $\tilde{J}_2$ are the effective pair-exchange frequencies including the three-spin exchanges in and out of the basal plane, and K is the square four-particle exchange frequency. $\tilde{J}_1$ and K are computed to be 6.1 ± 0.5 μK and -1.5 ± 0.4 μK, respectively, from θ and B for 19.29 cm^3/mole, if one assumes $\tilde{J}_1 = \tilde{J}_2$. The obtained relation of $K = -0.25\tilde{J}_1$ indicates that the four-particle exchange interaction is as important in the nuclear magnetism of hcp ^{3}He as the three-particle exchange interactions. The magnetic phase transition temperature T_c for 19.29 cm^3/mole is expected to be 12 μK, using the relation $T_c = \theta/2$ obtained for $K = -0.2\tilde{J}_1$.[8]

The ac susceptibility measurements were influenced by eddy currents in the cell body produced by the ac magnetic fields. The ac susceptibility χ is given from a complex mutual inductance M by $\chi = M(D/\alpha)e^{i\phi}$. Here D is a coefficient determined by the configuration of the cell and the coils, and α and ϕ are functions of the frequency of the ac field. Because of the difficulty in experimentally determining α and ϕ, we computed them by the finite-difference method.[9] For example, α and ϕ are 0.985 ± 0.003 and 6.4 ± 1.6 deg, respectively, at a frequency of 19 Hz.

The ac susceptibility $\chi = \chi' - i\chi''$ in a field of 1.9 mT is shown in Fig. 3 as a function of frequency. According to the thermodynamic theory of Casimir and Du Pré,[10] the ac susceptibility measured by the ac field parallel to the
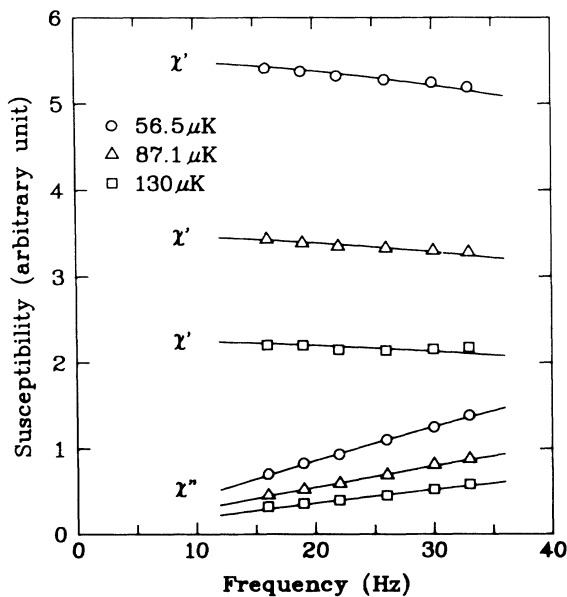
static field is given by[11]

$$\chi' = \frac{1}{1 + \omega^2 \tau^2}(\chi_0 - \chi_s) + \chi_s , \qquad (5)$$

$$\chi'' = \frac{\omega \tau}{1 + \omega^2 \tau^2}(\chi_0 - \chi_s) . \qquad (6)$$

Here ω is an angular frequency of the ac field, τ is the relaxation time, and χ_0 and χ_s are the isothermal and the adiabatic susceptibility, respectively. By analogy with the NMR relaxation of solid ^{3}He,[1] the energy generated by the ac field is transferred from the Zeeman system to the exchange system and then to the cell at ultralow temperatures. Equation (6) shows that the imaginary part χ'' appears at frequencies near the inverse relaxation time. In our frequency range, τ may be considered the Zeeman-exchange relaxation time or the spin-spin relaxation time in the Zeeman system, because the relaxation time between the sample and the cell is several hundreds of seconds as discussed in the preceding paragraphs. Furthermore, the Zeeman-exchange relaxation time is equal to the spin-spin relaxation time in fields where the Larmor frequency is sufficiently lower than the exchange frequency.[1] The magnetization generated by the ac field parallel to the static field cannot change more rapidly than the spin-spin relaxation time, so that the ac susceptibility is zero at frequencies where $\omega\tau \gg 1$. As a result, the ac susceptibility in nearly zero field can be given by Eqs. (5) and (6) with $\chi_s = 0$. Thus the Zeeman-exchange relaxation time for 18.34 cm^3/mole is determined to be 1.28 msec at 56.5 μK, 1.30 msec at 87.1 μK, and 1.31 msec at 130 μK by the least-squares fits seen as the solid lines in Fig. 3. The uncertainty in the relaxation time is ± 0.3 msec due to the error of ϕ. The heat absorption due to χ'' is estimated to be 0.3 fW at 50 μK,

FIG. 3. ac susceptibility $\chi = \chi' - i\chi''$ of hcp solid ^{3}He for 18.34 cm^3/mole as a function of frequency. The solid curves are the fits by Eqs. (5) and (6) with $\chi_s = 0$.
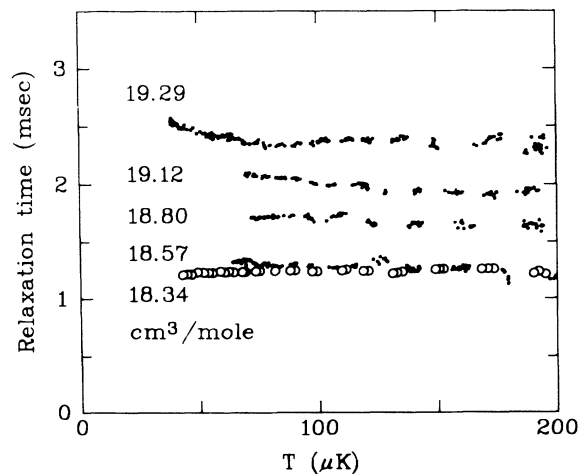

FIG. 4. Zeeman-exchange relaxation time of hcp solid ^{3}He as a function of temperature: open circles, for 18.34 cm^3/mole; solid circles, for other molar volumes. Some data for 18.34 cm^3/mole have been omitted for clarity.

which causes the temperature difference of 4 nK between the sample and the cell estimated from the boundary resistance of bcc ^{3}He.[12]

In Fig. 4 we show the temperature dependence of the Zeeman-exchange relaxation time obtained from the ac susceptibility at a fixed frequency of 19 Hz by using Eqs. (5) and (6) with $\chi_s = 0$. The Zeeman-exchange relaxation time should be compared with NMR measurements[1] in high magnetic fields where the Larmor frequency is greater than the exchange frequency. Our data in nearly zero field agree with $\frac{3}{10}$ of the high-field data within several tens of percent on the basis of the ten-thirds effect,[1] although the high-field data cannot be compared directly with our data. Our result of the temperature independence of the relaxation time also agrees with the NMR measurements. The relaxation time for 19.29 cm^3/mole increases at lower temperatures with decreasing temperature. This behavior is considered a signature of the critical phenomenon occurring near the magnetic phase transition. The phenomenon implies that the phase transition occurs at a temperature of a few tens of μK.

In summary, the Weiss temperature obtained from the magnetization is proportional to $V^{18.4 \pm 0.6}$. The result of an estimate of the exchange frequencies indicates that the four-particle exchange interaction is as important in the nuclear magnetism of hcp ^{3}He as the three-particle exchange interactions. The Zeeman-exchange relaxation time obtained from the ac susceptibility indicates the appearance of the critical phenomenon for 19.29 cm^3/mole.

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