Solid-^{3}He Magnetic Susceptibility by High-Precision NMR Measurements

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(Received 22 August 1983)

A high-precision NMR technique was used to measure the magnetic susceptibility of bcc solid ^{3}He at several molar volumes over a temperature range well above the ordering temperature. The results resolve a long-standing inconsistency between high-magnetic-field pressure measurements and earlier magnetic susceptibility measurements, give new values of the Curie-Weiss temperature θ nearly a factor of 2 smaller than previous values, and suggest that θ follows a power-law volume dependence.

PACS numbers: 67.80.Jd, 67.80.Gb, 75.30.Cr

The magnetic behavior of solid ^{3}He is believed to be a consequence of quantum exchange of whole atoms with a nuclear magnetic moment. This situation is unusual and interesting compared to other solids where magnetic effects rely on quantum exchange of electrons. Despite considerable theoretical and experimental efforts to understand magnetism in solid ^{3}He, a surprising number of unresolved problems still challenge our basic knowledge of this quantum solid. This Letter describes an experiment where new and unexpected results have been found using high-precision NMR measurements to study the magnetic susceptibility of solid ^{3}He. The results resolve an inconsistency between previous high-magnetic-field pressure measurements[1,2] and magnetic susceptibility measurements,[3,4] show that the Curie-Weiss temperature θ is about a factor of 2 smaller than the values found in earlier susceptibility work, and provide evidence in support of a power-law volume dependence for θ. Samples were investigated at various molar volumes (21 to 24 mL/mole). The measurements were done in the zero-field limit with a low magnetostatic field (17.1 mT), covering a temperature range of ≈ 16 to 514 mK.

A model-independent spin-exchange Hamiltonian is useful for developing a high-temperature series expansion of the magnetic susceptibility χ. In the limit of zero magnetic field, the inverse susceptibility can be written as

$$\chi^{-1}=C^{-1}(T-\theta+B/T), \tag{1}$$

where higher-order terms in temperature T have been excluded.[5] The Curie constant C and θ are familiar quantities; B, on the other hand, is a coefficient related to θ by $B=\theta^2-\alpha_2/8$, where α_2 is a second-order coefficient occurring in the high-T expansion of the Helmholtz free energy.

Accurate values of θ have not been available from past susceptibility experiments.[3,4] One group of experiments[3] were done at high temperatures (≈ 10 to 500 mK) where the precision was insufficient to determine accurate values of θ because of very small deviations from Curie-law behavior. Another group of experiments[4] were done at much lower temperatures (≈ 1 to 15 mK) near the ordering temperature where significant higher-order deviations from Curie-Weiss behavior developed. However, because of correlations among higher-order terms in Eq. (1), values of θ could easily be misinterpreted from the data since high-temperature behavior was not studied.

One can also compare susceptibility measurements with high-magnetic-field pressure measurements $P(T,H)$.[1,2] The latter are sensitive to the volume derivatives of θ, which can be intergrated to yield values of θ. However, since these values are obtained indirectly, questions concerning analytical procedure can arise.[6] The recent high-precision $P(T,H)$ work[1] and the older $P(T,H)$ work,[2] although mutually consistent, yield values of θ much different from those obtained from early χ measurements.[3,4] This inconsistency has been known for sometime,[7] but its explanation has never been clearly identified.

The data reported in this Letter were obtained by a high-precision NMR *susceptibility* technique involving a sample chamber with two nearly identical cells, to form two different-density solid ^{3}He samples. Thermal contact was achieved by solidifying the samples in the interstitials of copper wires tightly packed inside each cell. Half of the wires were used to thermally couple the two samples, and the rest were used to establish thermal contact with CMN and LCMN thermometers[8] and the mixer of a dilution refrigerator. Typically, one sample was formed at a high density (≈ 21 mL/mole) where the susceptibility closely followed Curie-law behavior. The other sample, which was formed at a lower density, followed Curie-Weiss behavior.[9]

A phase-coherent pulsed-NMR spectrometer was used to compare the magnetic behavior of the low-density sample with the high-density sample by simultaneously measuring the free-induction decay (FID) of both samples. The FID's were Fourier transformed to produce the frequency-domain spectrum of each sample. A small gradient field (0.7 G/cm) was used to separate the spectra. The area under each absorption spectrum was proportional to χ since the magnetostatic field was small. This simultaneous-measurement technique made it possible to use the susceptibility of the high-density sample to measure the temperature of the low-density sample. The procedure was accomplished by referring to Eq. (1) and writing two similar equations for each sample. Upon elimination of the temperature between the equations and neglecting terms higher than second order, we get

$$\chi_H/\chi_L = C_H/C_L[1-\Delta\theta(\chi_H/C_H)+A(\chi_H/C_H)^2], \qquad (2)$$

where $\Delta\theta=\theta_L-\theta_H$, $A=B_L+\theta_H{}^2$, and χ_H and χ_L represent the measured susceptibilities. Subscripts H and L refer to high- and low-density samples, respectively. The B_H coefficient was neglected. Constants C_L and C_H were determined at a high-temperature reference point[10] where Curie-law behavior was applicable. By determining the ratio of the two measured quantities, χ_H/χ_L, thermal fluctuations and the spectrometer's drift and gain changes cancel to first order, increasing the relative accuracy by at least an order of magnitude. Additional experimental details are described elsewhere.[11]

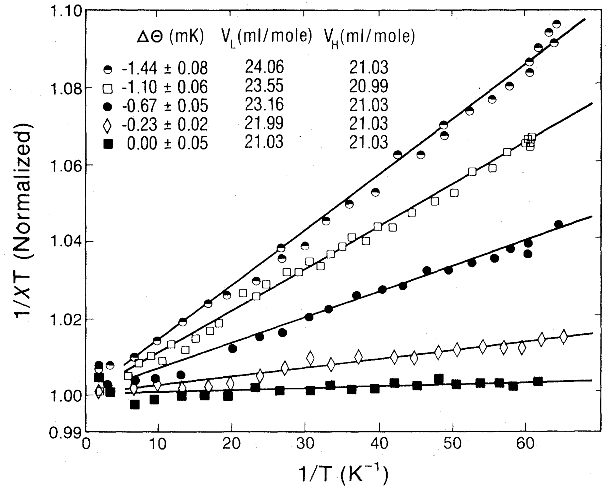

FIG. 1. Solid-^{3}He susceptibility, of several molar-volume samples, plotted as $1/\chi T$ vs $1/T$.

With the high-density sample held at a fixed molar volume in one cell, the susceptibilities of several density samples formed in the other cell were measured. The results are shown in Fig. 1 plotted as $1/\chi T$ vs $1/T$. The data along the ordinate is the ratio χ_H/χ_L, normalized to 1 at $1/T=0$. The abscissa, labeled in inverse kelvin units, comes from the χ_H data $[T^{-1}=\chi_H/\chi_H(\text{ref})T_{\text{ref}}$, where "ref" refers to the reference point[10]$]$.

The data shown in Fig. 1 were analyzed in general by a univariate curvilinear regression to Eq. (2), with use of orthogonal polynomials. Drawn through the data of each sample are the resulting straight-line fits whose slopes determine values of $\Delta\theta$. These values are listed in Table I. Typically, the data scattered $\leqslant\frac{1}{3}\%$ over the entire temperature range. The errors assigned to the values are the accumulated errors from T_{ref} ($\approx 1\%$), $\chi_H(\text{ref})$ ($\approx 3\%$), and the standard errors of the slope ($\approx 1\%$) and the intercept ($\approx 0.1\%$). The possibility of systematic errors and irreproducible effects were carefully investigated. For example, high- and low-density samples were interchanged between cells, the magnetostatic field shifted by 0.2%, the gradient field increased by 10%, and the receiver bandwidth doubled. None of these changes (including others we tried) caused significant effects and the data remained consistent to within 8%.

Attempts to observe curvature effects from the second-order term were done by collecting a larger number of points than usual for two samples. With the present accuracy, only upper and lower limits can be placed on B; viz., $B=0\pm1$ mK^2 at 24.13 mL/mole, and $B=-1.24\pm0.65$ mK^2 at 23.55 mL/mole.

A comparison of our results with previous χ and $P(T,H)$ measurement is shown in Fig. 2. Our

TABLE I. Values of $\Delta\theta$ and θ for several molar volumes. EST denotes an estimate of θ_H.

V (mL/mole)	$\Delta\theta$ (mK)	θ (mk)
24.13	-1.37 ± 0.07	-1.48 ± 0.07
24.13	-1.28 ± 0.13	-1.39 ± 0.13
24.06	-1.44 ± 0.08	-1.55 ± 0.08
23.55	-1.10 ± 0.06	-1.21 ± 0.06
23.16	-0.67 ± 0.05	-0.78 ± 0.05
21.99	-0.23 ± 0.02	-0.34 ± 0.02
21.03	0.00 ± 0.05	(-0.11)EST

values of $\Delta\theta$ are plotted in Fig. 2 as a function of molar volume V (i.e., $\ln|\Delta\theta|$ vs $\ln V$). Line a, as determined by a computer fit, corresponds to a power-law volume dependence, $\Delta\theta \approx V^{\gamma}$, where $\gamma = 19 \pm 2$. Although there is no satisfactory theoretical reason for a simple power-law dependence,[6] we believe that it is significant that the data demonstrate this dependence since similar behavior has been observed recently in several other types of measurements.[12] The strong volume dependence of $\Delta\theta$ suggests that θ_H is small; therefore we estimated its value to be -0.11 mK by extrapolating line a to 21.03 mL/mole. This extrapolation procedure tends to overestimate θ_H (i.e., give too large a negative value); nevertheless, we add -0.11 mK to each $\Delta\theta$ value in Table I to obtain values of θ. These new values of θ differ from those found in earlier χ work[3] by nearly a factor of 2; however, they are in good agreement with values from the $P(T,H)$ measurements.[1,2] Line b drawn through the points is a computer fit to both the $P(T,H)$ and the present χ data. The slope of this line corresponds to a power of 16.9 ± 0.7. Line c has the same slope as line b, but it is displaced upward by a factor of 2 to fit (by eye) the early χ measurements. This discrepancy reflects the quality of the early experiments and the systematic errors originating from temperature measurements and from large statistical deviations of the data, which can be significant when θ is determined by plotting χ^{-1} vs T, as was done.

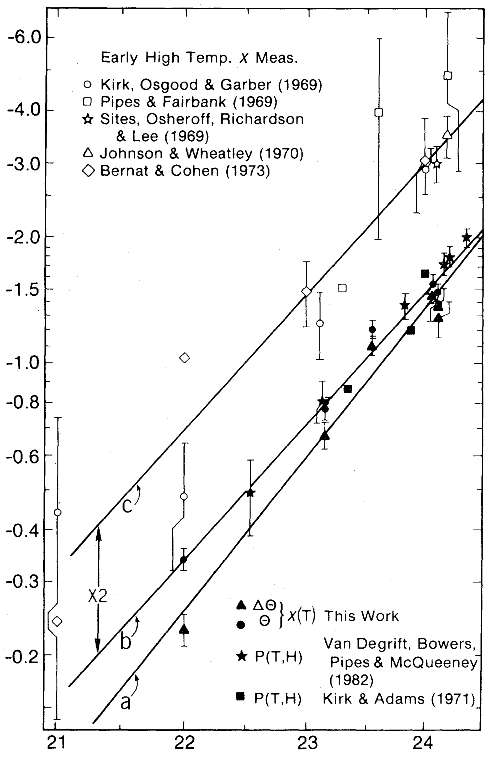

FIG. 2. Comparison of θ's from $P(T,H)$ measurements (Refs. 1 and 2) and early χ measurements (Ref. 3) with present work (solid circles and triangles), as a function of molar volume. See text for discussion of lines a, b, and c.

As further justification of our estimate of θ_H, we have analyzed the high-density sample using temperatures determined from the CMN and LCMN thermometers. Figure 3 shows the high-density sample in the form $1/\chi T$, plotted as the ratio $\chi_{\mathrm{CMN}}/\chi_H$ versus χ_{CMN}, expressed in inverse kelvin units. χ_{CMN} is the CMN susceptibility determined by $\chi_{\mathrm{CMN}} = C/(T - \Delta_{\mathrm{CMN}})$, where C is the CMN Curie constant and Δ_{CMN} is the "shape correction" factor used in CMN thermometry to relate magnetic temperature scales to the absolute temperature scale. Around 18 mK the data begin to depart from a linear dependence in temperature and finally deviate by 4% at 15 mK. This departure is caused by loss of thermal equilibrium between the CMN thermometer and the high-density sample at the lowest temperatures. The straight line is therefore determined by a computer fit of all data between 18 and 500 mK. From the slope of the line we find $\theta_H - \Delta_{\mathrm{CMN}} = -0.45 \pm 0.07$ mK. Unfortunately, Δ_{CMN} is not a universal constant, but it typically varies between $+0.3$ and $+0.7$ mK depending on thermometer design.[13] These values, however, imply that θ_H ranges between -0.22 and $+0.32$ mK, including the error limits. Applying a similar analysis with the LCMN thermometer and intercomparing the CMN and the LCMN thermometers show that

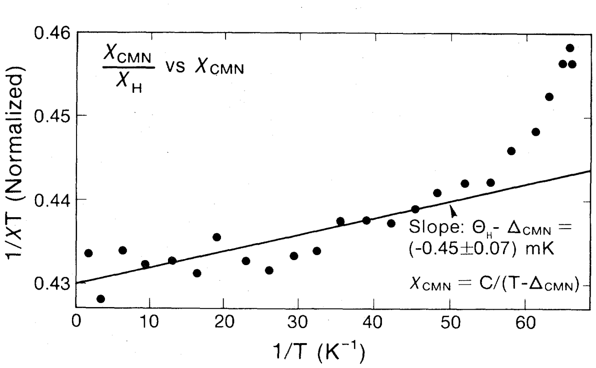

FIG. 3. A $1/\chi T$ vs $1/T$ plot of the high-density sample with use of the CMN thermometer.

$\theta_H - \Delta_{\mathrm{LCMN}} = +0.10 \pm 0.11$ mK and $\Delta_{\mathrm{CMN}} - \Delta_{\mathrm{LCMN}} = +0.55 \pm 0.04$ mK. Again, Δ_{LCMN} is not constant but ranges between -0.15 and $+0.30$ mK; this implies that θ_H lies between -0.16 and $+0.51$ mK. We conclude that there is no justification for θ_H having a rather large negative value; in other words $\theta_H > -0.22$ mK. The possibility of a small positive value cannot be ruled out, and theoretical arguments do not preclude such a possibility. In conclusion, these results give supporting evidence that θ follows a power-law volume dependence. If this conclusion is valid, then its origin could generate a significant challenge for the current theoretical model[5,6] (a two-parameter multiple-exchange Hamiltonian) believed to offer the most complete understanding of solid-^{3}He magnetism. Certainly additional high-quality experiments are needed to clarify this point.

In summary these new high-precision susceptibility measurements provide new values of θ at several molar volumes and resolve a longstanding inconsistency between $P(T,H)$ and χ measurements.

We wish to thank Dr. J. Parpia for useful discussions on shape corrections and Professor J. Hetherington (and Willard) for insightful remarks. One of us (Z.O.) wishes to thank the Charles F. Squire Fellowship Fund for a summer fellowship. This work has been supported by the National Science Foundation through Grant No. DMR-8205902.

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[8] CMN (cerous magnesium nitrate) and LCMN (lanthanum-diluted CMN) susceptibility measured with a SQUID.

[9] Molar volume values were determined, to within $\pm 0.05\%$, from the solidification pressures [E. R. Grilly, J. Low Temp. Phys. 4, 615 (1971)] with capacitive strain gauges connected to each cell.

[10] $T_{\mathrm{ref}} = 513.7 \pm 5.0$ mK, from an NBS SRM 767 device.

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[13] The possibility of measuring Δ_{CMN} by direct comparison with the NMR susceptibility of the copper wires in the cells was considered. However, because of the very short duration of the copper FID it was not possible to achieve the requisite accuracy.