

SIGN AND MAGNITUDE OF THE EXCHANGE INTERACTION IN SOLID  $^3\text{He}$   
FROM MAGNETIC SUSCEPTIBILITY MEASUREMENTS\*

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The nuclear magnetic susceptibility of solid  $^3\text{He}$  was measured between temperatures of 5.3 and 800 mK for several molar volumes. In each case, the susceptibility followed a Curie-Weiss law with a negative Weiss constant, hence a negative exchange interaction  $J$ . Magnitudes of  $J$  as a function of molar volume agree with previous measurements.

We report here for the first time a study of the magnetic susceptibility of solid  $^3\text{He}$  in which, for every molar volume investigated, the magnetic behavior was like that of an antiferromagnet at temperature  $T$  well above the Néel temperature. The measurements also gave a direct determination of the magnitude of  $J$  as a function of molar volume. Susceptibility measurements by earlier workers<sup>1-5</sup> led to inconclusive results regarding the sign and magnitude of the exchange interaction  $J$ . Previous measurements were made at high temperatures so that the best high-precision measurements gave only upper limits. Furthermore  $^4\text{He}$  impurities may have influenced some of these measurements.

One can expect that when  $T \gg J/k$ , the magnetic susceptibility will follow to good approximation a Curie-Weiss law

$$\chi = C(T - \theta)^{-1}, \quad (1)$$

where  $C$  is the Curie constant. The Curie-Weiss constant  $\theta$  can in turn be expressed as a function of  $J$ :

$$\theta = \frac{2}{3}Z(J/k)I(I+1) \quad (2)$$

with  $Z = 8$  for the number of nearest neighbors in the bcc structure, the spin  $I = \frac{1}{2}$ , and  $k$  the Boltzmann constant. Thus, it is clear that a determination of  $\theta$  from the susceptibility as a function of temperature will yield both the sign and magnitude of  $J$ . Since  $^3\text{He}$  is a very compressible solid and  $J$  has a very large molar-volume dependence the application of pressure will lead to large changes in the magnitude of  $J$ .

The  $^3\text{He}$  sample was contained in an epoxy cell with 4300 No. 50 Formvar-insulated copper wires feeding through an epoxy pressure seal. Thermal contact to the  $^3\text{He}$  was established with the wires and the cell was designed so that none of

the sample was more than 0.003 cm from the nearest cooling surface in order to minimize the relaxation times.<sup>6</sup> The Cu wires from the pressure cell led to a tin heat switch which was attached to the bottom of a 600-g cerium-magnesium-nitrate salt. Temperatures of 5.3 mK were realized at the sample by magnetically cooling the salt. A dilution refrigerator was used to cool the salt to 0.1 K before demagnetization and to perform the role of the usual "guard salt" at 90 mK. With this arrangement we could maintain temperatures below 10 mK for more than a week.

Temperatures were measured by cw NMR on  $^{63}\text{Cu}$  and  $^{195}\text{Pt}$  using a frequency modulated Robinson circuit. The  $^{63}\text{Cu}$  sample was part of the 4300 wires in the cell and the  $^{195}\text{Pt}$  sample was 0.12 g of 5- $\mu$  powder interspersed among the Cu wires. The temperature was assumed to be proportional to the ratio of a calibrator signal to the height of the NMR signal. The proportionality constant was determined at 0.4 K by using a  $^3\text{He}$  vapor pressure thermometer. Between the temperatures of 5.3 and 395 mK the  $^{195}\text{Pt}$  and  $^{63}\text{Cu}$  thermometers were found to agree within 5%.

Thermal equilibrium times were checked by turning on the tin heat switch and quickly cooling the sample from about 100 mK to the lower temperature of the salt. The entire lower stage below the tin heat switch cooled to 10 mK in 4 min and reached a lowest temperature of 7 mK in 30 min. Within the 5% precision of these measurements there was no disequilibrium between the thermometer and the  $^3\text{He}$ . The cooling rate was limited by thermal contact with the salt.

A Rollin circuit in conjunction with a Varian No. C-1024 signal averager was used to measure the  $^3\text{He}$  susceptibility at 714 G. The linewidth was broadened by the superconducting magnet inhomogeneity to 0.25 G which is much larger than

the natural linewidth. Each signal height from the  $^3\text{He}$  was compared with a calibrator signal which allowed compensation for drifts in the gain of the measuring equipment and drift in the  $Q$  of the tank circuit.  $^3\text{He}$  signals at power levels 0.1, 1, and 4 nW were taken to check for saturation and heating effects. Only below 10 mK was saturation observed. At the lowest temperature and the highest rf power level the saturation was 2.3% for 24.0-cm $^3$ /mole, 9.5% for 23.1-cm $^3$ /mole, 18% for 22.0-cm $^3$ /mole, and 25% for 21.0-cm $^3$ /mole solids. At the lowest power level and temperature, we never exceeded 1% saturation even for the 21.0-cm $^3$ /mole solid.

In Fig. 1 we have plotted against temperature the inverse susceptibility  $\chi^{-1}$ , in arbitrary units. Measurements were taken on solids that were formed at molar volumes 24.0, 23.1, 22.0, and 21.0 cm $^3$ /mole by the blocked capillary technique. As shown in the figure, the typical scatter in the data, which comes mainly from the thermometer, is 5% or less. The linear curves drawn through the data points<sup>7</sup> were fitted by the method of least squares and in each case extrapolate with an intercept on the negative temperature axis. We interpret the negative intercepts to mean that solid  $^3\text{He}$  will order as an antiferromagnet. Recent theoretical work<sup>8-10</sup> has predicted antiferromagnetic behavior. It would be difficult to interpret the data otherwise in consideration of the knowledge that solid  $^3\text{He}$  has no electronic structure to produce internal electric and magnetic fields to complicate the interpretation of the magnetic sus-

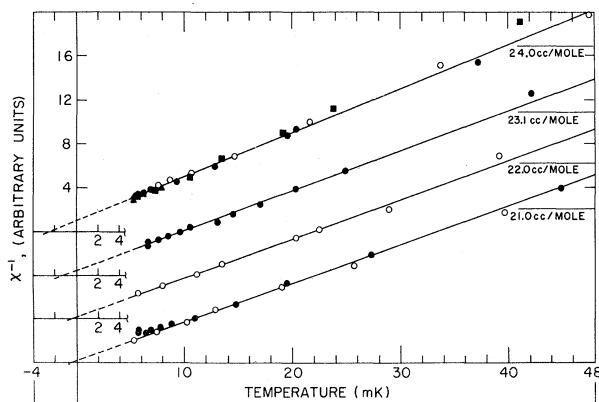


FIG. 1. Inverse susceptibility versus temperature for various molar volumes. Closed circles, squares, and triangles are for samples with 125-ppm  $^4\text{He}$  impurity. Open circles are for samples with  $\approx 2$ -ppm  $^4\text{He}$  impurity. For 24.0-cm $^3$ /mole solid, the closed circles, squares, and triangles distinguish measurements from different runs.

ceptibility as in other solids. The values of the intercepts on the temperature axis gave the Curie-Weiss constants  $\theta = -2.9 \pm 0.7$  mK,<sup>11</sup>  $-1.3 \pm 0.3$  mK,  $-0.48 \pm 0.16$  mK, and  $-0.44 \pm 0.30$  mK for the respective molar volumes 24.0, 23.1, 22.0, and 21.0 cm $^3$ /mole. The errors quoted for  $\theta$  were obtained from an error analysis of the deviations of the data from the best straight-line fits. Included in the errors are any curvature effects from a higher order expansion of  $\chi$  in  $J/kT$ . Using the relation of Baker et al.<sup>12</sup> and Eq. (2) the above  $\theta$ 's would give Néel temperatures  $T_N = 2.0 \pm 0.48$ ,  $0.89 \pm 0.21$ ,  $0.33 \pm 0.11$ , and  $0.30 \pm 0.21$  mK, respectively.

From Eq. (2) the magnitude of the exchange interaction,  $|J|$ , at each molar volume was computed and entered on the graph in Fig. 2. The dashed curve represents the extensive work of Panczyk and Adams (PA)<sup>13</sup> in which  $|J|$  was determined from measurements of  $(\partial P/\partial T)_V$ . Also  $|J|$  has been determined by several workers<sup>14</sup> from NMR relaxation time measurements. We are in best agreement with the work of PA<sup>13</sup> and Richardson, Hunt, and Meyer,<sup>14</sup> for all molar volumes except 21.0 cm $^3$ /mole. In the last case, the agreement was somewhat worse because we were limited by the precision of the thermometry, and possible small effects from saturation and relaxation times.

Furthermore, our data gave one other consistent result; the slopes of the straight lines in Fig. 1 were proportional to the Curie constant  $C$ . They scaled with the inverse molar volumes with-

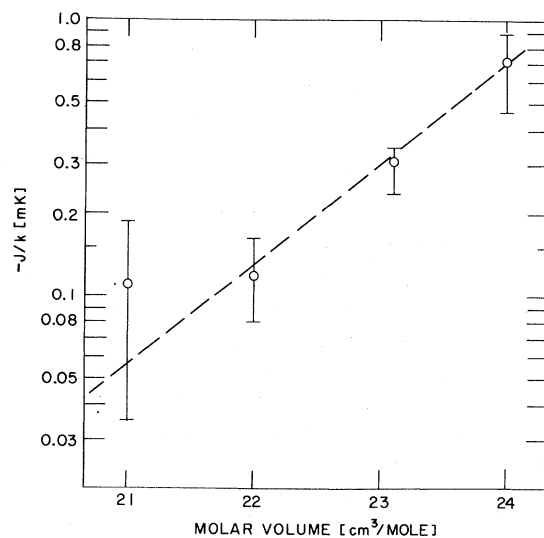


FIG. 2. The exchange interaction versus molar volume. Dashed curve of  $|J|$  from Panczyk and Adams, Ref. 13. Open circles, this work.

in 2% for all densities studied except the 23.1-cm<sup>3</sup>/mole solid which scaled to within 5.5%.

Measurements were extended to 0.8 K and the data followed a Curie-Weiss law. However, to cover the entire temperature range, it was necessary to measure the area under the absorption signals because the linewidth changed at  $\approx 100$  mK. Apparently some material in the sample cell was undergoing a magnetic transition which changed the linewidth of the <sup>3</sup>He signal about 0.1 G. This small broadening had negligible effect on the Pt and Cu since their linewidths were much broader. Below 80 mK the area and signal height data have equivalent temperature dependence. If we plot the high temperature (0.1 to 0.8 K) signal-height data versus temperature, the extrapolated straight-line fits give large negative Curie-Weiss constants of about 20 to 30 mK. Perhaps this observation is a tentative explanation for some of the unusually large Curie-Weiss  $\theta$ 's reported in earlier susceptibility measurements.<sup>4</sup> We investigated and eliminated the possibility that the <sup>3</sup>He was undergoing a change in linewidth by using the same pressure cell to measure the liquid <sup>3</sup>He susceptibility. Again we observed the linewidth change. We suspect ordering of a small iron impurity (62 ppm) in the Pt powder. There may also be some magnetic impurity in the Cu wires leading into the cell.

A limited study of the effects of <sup>4</sup>He impurities, present in concentrations of 125 and  $\approx 2$  ppm, was made to see if the impurity changed the susceptibility by the isotopic phase separation<sup>15</sup> or enhanced the exchange interaction.<sup>16</sup> In none of the molar volumes investigated except 21.0 cm<sup>3</sup>/mole did we observe a difference in  $\chi$  for the two concentrations. The 21.0-cm<sup>3</sup>/mole solid did differ at temperatures  $< 10$  mK from the 125-ppm <sup>4</sup>He, departing slightly from the Curie-Weiss law behavior, whereas the sample with  $\approx 2$  ppm <sup>4</sup>He followed the Curie-Weiss law to the lowest temperature measured.

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