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MAGNETIC SUSCEPTIBILITY MEASUREMENTS IN VERY DILUTE SOLID SOLUTIONS OF He^4 IN He^3 †

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Measurements of the magnetic susceptibility of very dilute solid solutions of He^4 in He^3 as a function both of molar volume and of He^4 concentration in the α phase have been made between 1 and 0.04°K by steady-state nuclear-resonance techniques. The most dilute solution (5×10^{-7} He^4) followed Curie's law over the range in molar volume (23.0-21.0 cc) measured. The less dilute solutions (1×10^{-4} and 3×10^{-4} He^4) followed a Curie-Weiss law with relatively large, antiferromagnetic Weiss constants.

Much experimental[1-9] and theoretical work[10-16] over the past ten years has been directed at the problem of the magnetic behavior of solid He^3. The motivation for this work has been in large part the possibility of studying nuclear spin ordering at experimentally realizable temperatures. Solid He^3 is the best candidate for such a study as its large zero-point motion results in relatively large exchange effects.

Earlier experimental studies[1,2,5] of the magnetic susceptibility of solid He^3 have been hampered by the unexpectedly large and poorly understood effects of small amounts of He^4 impurity. In order to separate these effects from the behavior of the pure solid and to begin to study the influence of this impurity on the magnetic susceptibility of the solid, we have made nuclear-resonance absorption measurements on solid He^3 in two separate apparatus.

The first of these was used by one of us[17,18] to make measurements between the temperatures of about 1 and 0.04°K at He^4 impurity concentrations of $(5\pm2)\times10^{-7}$ and $(1.0\pm0.4)\times10^{-4}$ and at molar volumes between 23.0 and 21.0 cc. The thermometry in this apparatus was accomplished by means of carbon resistors in the range between 1 and 0.3°K and by copper nuclear-resonance thermometers[19] between 0.5 and 0.04°K. The resistors were calibrated against the vapor pressure of He^3. This apparatus was specifically designed to insure that equilibrium conditions were achieved. The precautions taken included having two copper thermometers. One of these cooled before the sample. The other was cooled by the sample. Thus, the temperature of the sample lattice was bracketed. In addition, relaxation-time measurements were made by the saturation method; the sample remained at the lowest temperature reached for times of 5-10 h to eliminate the possibility of further relaxation; and measurements were made on both the warming and cooling portions of the demagnetization cycle without observing any hysteresis.

The second apparatus was used to make measurements between the temperatures of about 1.2 and 0.4°K, at a He^4 impurity concentration of $(3\pm1)\times10^{-4}$ and at a molar volume of 19.9 cc. Obtaining equilibrium at this molar volume and He^4 concentration and over this range in temperature presented no difficulty as the relaxation times are known[8] to be relatively short over most of this range and extrapolate to even shorter values over the rest of the range. As an independent check of this point, at the lowest temperature, where the relaxation times should be longest, no change in susceptibility was observed when the apparatus remained at constant temperature for 24 h. Direct He^3 vapor pressure measurements were used for thermometry in this apparatus.

Our results show that in the purest solution, at a He^4 impurity concentration of 5×10^{-7}, at all molar volumes measured (23.0 ± 0.1, 22.4 ± 0.1, and 21.0 ± 0.1 cc), the magnetic susceptibility follows Curie's law with limits on the Weiss constant of $\pm3\times10^{-3}$ °K. These measurements were made in the first apparatus described above and extend down to about 0.04°K. They are shown in Fig. 1 and appear to be characteristic of the pure solid. These data are consistent with the latest calculations of Nosanow and co-workers,[16] as well as with the earlier ones,[15] with the relax-

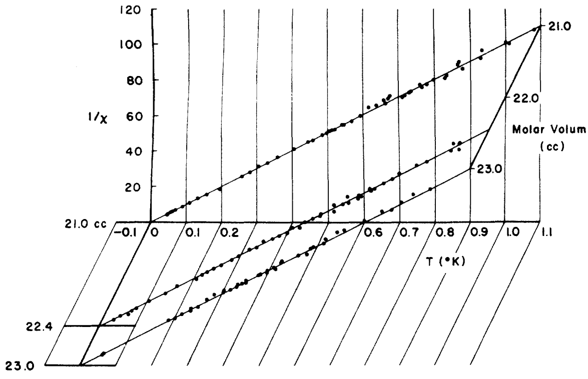

FIG. 1. The inverse of the susceptibility of solid He^3 as a function of temperature at a He^4 impurity concentration of 5×10^{-7} and at molar volumes of 23.0 ± 0.1, 22.4 ± 0.1, and 21.0 ± 0.1 cc. The data are plotted on a three dimensional graph with the inverse of the susceptibility along the vertical axis, the temperature along the horizontal axis, and the molar volume along an axis perpendicular to the plane of the page.

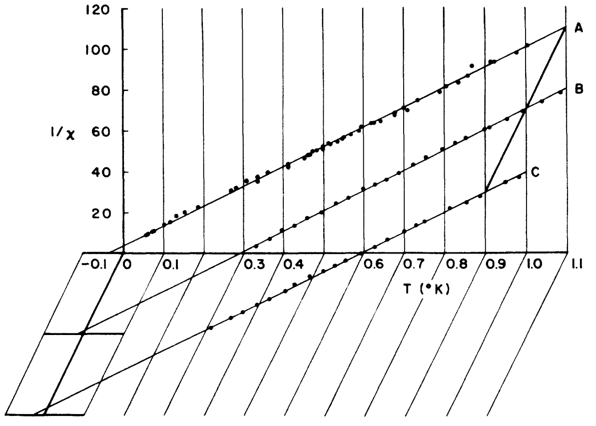

FIG. 2. The inverse of the susceptibility of solid He^3 as a function of temperature. Curve A is data taken at a He^4 impurity concentration of 1×10^{-4} and a molar volume of 21.0 ± 0.1 cc. Curves B and C are data taken at a He^4 impurity concentration of 3×10^{-4} and at a molar volume of 19.90 ± 0.05 cc. The same graphing technique as in Fig. 1 is used here except that the three runs shown are equally spaced along an axis perpendicular to the plane of the page.

ation-time measurements of a number of authors[4,6,7] and with the pressure data of Adams and co-workers.[9]

At an impurity concentration of 1×10^{-4}, the susceptibility was measured using the first apparatus at a molar volume of 21.0 ± 0.1 cc. The data are shown in Fig. 2. They followed a Curie-Weiss law over the whole temperature range measured, with a Weiss constant of $(+37\pm12)\times10^{-3}$ °K. This result agrees reasonably well with the earlier measurements of Thomson, Meyer, and Dheer[5] which are given in detail in Thomson's thesis.[20] They found a Weiss θ of $(+18\pm5)\times10^{-3}$ °K at a He^4 concentration of roughly 3×10^{-4} and a molar volume of 21.2 cc.

In addition, we made measurements in the second apparatus at an impurity concentration of 3×10^{-4} and a molar volume of 19.90 ± 0.05 cc. Two runs were taken between the temperatures of about 1.2 and 0.4°K and the data, shown in Fig. 2, followed a Curie-Weiss law in both, with Weiss constants of $(+35\pm15)\times10^{-3}$ °K and $(+20\pm15)\times10^{-3}$ °K. This result is smaller in magnitude than those of Thomson, Meyer, and Dheer[5] at roughly these densities. They found values of the Weiss θ as large as 100×10^{-3} °K at 20.17 cc/mole and 125×10^{-3} °K at 19.5 cc/mole.[20] Further, Giffard and Hatton[8] have measured the exchange-lattice relaxation time at 19.97 ± 0.03 cc/mole as a function of frequency at He^4 concentrations up to and including a concentration of 3×10^{-4}. Using the three bath model of Garwin and Landesmann,[4] they have extracted a value for the exchange frequency, $J/2\pi=0.97$ MHz. Using the mean of the Weiss θ's above and the relationship $\theta=\frac{1}{4}Z(\hbar J/k)$ valid for $T\gg\hbar J/k$, where Z, the number of nearest neighbors, is 8 for a bcc lattice, we obtain the much larger value $J/2\pi=290$ MHz.

In summary, we have shown that, with a pure enough sample, susceptibility measurements can be made, even at high densitites in the α phase, that are characteristic of solid He^3 and that these results are consistent with the current theory of the magnitude of exchange effects[15,16] and also with the results of other experiments.[4,6,7,9]

Secondly, we have seen that as the He^4 impurity content is increased nonzero Weiss constants appear. At He^4 concentrations of 1×10^{-4} and 3×10^{-4}, the positive values of θ obtained indicate a strong antiferromagnetic interaction. We have observed the sign and magnitude of θ in this concentration range in two apparatus. It agrees qualitatively though not quantitatively with that observed in earlier measurements[5] in the same concentration range.

We have also made measurements at a He^4 concentration of 3×10^{-3} and at molar volumes between 22.6 and 20.0 cc. Some of these measurements have been reported earlier.[17,18] The behavior of the susceptibility at this concentration appears to be quite complex and we are investigating it in further detail.

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