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Magnetic Susceptibility of ^{3}He in Grafoil at Ultralow Temperatures

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Measurements of the NMR susceptibility of ^{3}He in Grafoil indicate that the surface layer exhibits a Curie-Weiss magnetization tending towards ferromagnetism. The surface magnetization has a magnitude approximately equivalent to the number of spins in one monolayer. At temperatures between 0.43 and 1 mK the susceptibility has a strong magnetic field dependence. The data are consistent with a simple Landau-Ginzburg model indicating a phase transition at 0.38 mK with a lower than expected magnetization.

We present here measurements on the magnetic susceptibility, χ, of ^{3}He confined within Grafoil[1] at temperatures extending down to 0.43 mK. The temperature dependence of χ indicates that it tends towards a ferromagnetic transition near 0.38 mK, but we also find a striking magnetic field dependence to χ at temperatures below 1 mK.

The high-temperature ($T>1$ mK) behavior of χ follows a Curie-Weiss law, as found by Ahonen *et al*,[2] on Mylar and lamp black (amorphous) carbon. They were able to fit their data with an equation of the form

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

where the constant term A was attributed to the susceptibility of the bulk liquid while the temperature-dependent term was assumed to be due to the ^{3}He next to the adsorbent walls. However, we find that the more uniform substrate of Grafoil results in a substantially different interpretation of the amount of ^{3}He involved in the temperature-dependent susceptibility and that the low-temperature behavior ($T<1$ mK) indicates that χ tends to saturate in surprisingly low fields.

Our sample cell consisted of two epoxy cylinders which incorporated NMR coils. The cylinders were tightly packed with Grafoil with the principal orientation either parallel or perpendicular to the applied magnetic field. The Grafoil had been prepared by baking *in vacuo* at 1000°C for several hours. Liquid ^{3}He (with $\lesssim 40$ ppm ^{4}He; about 2% of a monolayer) entirely filled the spaces in and around the Grafoil and also filled a tube extending down from the sample chamber to the main chamber of a copper nuclear demagnetization cooled cell.[3]

Temperature has been measured using the NMR susceptibility χ_p of Pt powder. Our sample of Pt showed a small temperature dependence of T_1/χ_p where the Pt T_1 measurement was made using 90°-90° pulse sequences. Typically T_1/χ_p rises with temperature about 4% between 2 and 10 mK. We obtained average values for our calibration using several T_1 measurements between 4 and 10 mK. We then adjusted the temperature scales of each run to agree at T_c where more than one run was made at a given pressure. Our final temperature calibrations cause T_c to agree well with recent results of Paulson *et al.* using lanthanum-diluted cerium magnesium nitrate.[4]

The susceptibility of the ^{3}He sample was determined using pulsed NMR techniques with tipping pulses of between 0.5° and 1°. For all data the Q of the NMR coils limited their ringing time constant to <0.05 msec. The relative values of the susceptibility were found from an extrapolation back to zero time of the free-induction decay amplitudes measured between 0.25 and 1 msec. We have empirically determined that the free-induction decay amplitude $M(t)$ is more accurately represented by $M(t) = M(0)\exp(-\alpha t^{1.5})$ than by an exponential in the first power of t at temperatures below the superfluid transition. The constants $M(0)$ and α were determined for each trace using a least-squares fit. On selected traces after subtracting the coil ringing this shape has been shown to fit to within 2% at times $\geqslant 0.12$ msec after the pulse. In subsequent work with different Grafoil samples, modified electronics allowed us to confirm that a similar line shape was indeed followed to within 0.05 msec after the pulse.

Figures 1(a) and 1(b) show typical data for χ^{-1} vs T. The actual data runs spanned temperatures up to 20 mK. The solid lines in Fig. 1 are fits by Eq. (1) assuming $\theta = 0.38$ mK and excluding points below the superfluid transition (T_c). A least-squares analysis of these data only considering points for $T > T_c$ and allowing θ to be a free parameter gives values of $\theta = 0.38 \pm 0.05$ mK. Below T_c we see the apparent value of χ drop below the fitted curve because of the reduction of the liquid susceptibility.[5] (In the 26.0-bar data no measurements could be made in the A phase because of the complicated beat structure in the free-induction signal caused by A-phase frequency shifts.) Although we can make an approximate correction to the liquid term A in Eq. (1) we will restrict the analysis to the regions $T \geqslant T_c$ and $T \ll T_c$.

Figure 2 shows the observed transverse relaxation time T_2^* (which was measured as the $1/e$ time of the free-induction decay signal) of the ^{3}He for the data in Fig. 1(a). Magnetic field inhomogeneity contributed ~1 msec^{-1} to $1/T_2^*$. The increase in T_2^* below T_c (1.45 mK) is in part due to the rapid decrease in the contribution of susceptibility of liquid in the connecting tube to the cell, thus decreasing the effect of field inhomogeneity. More importantly we can see that T_2^* appears to decrease rapidly below 1 mK as we approach θ.

The amount of sample which participates in the Curie-Weiss law behavior is of primary interest in this experiment. Ahonen *et al.* found that approximately five monolayers were involved on their Mylar substrate while perhaps five to ten layers may follow this law in the lamp black sample. In our measurements, by using the limiting value of the susceptibility of the normal liquid at temperatures well below the Fermi temperature T_F, we can deduce from the ratio B/A the ratio of the amounts of sample having the two different susceptibilities of Eq. (1). For example, with $P = 3.74$ bars the limiting value for the susceptibility of the liquid[6] is $3.37C$ where C is the Curie constant and thus our B/A ratio

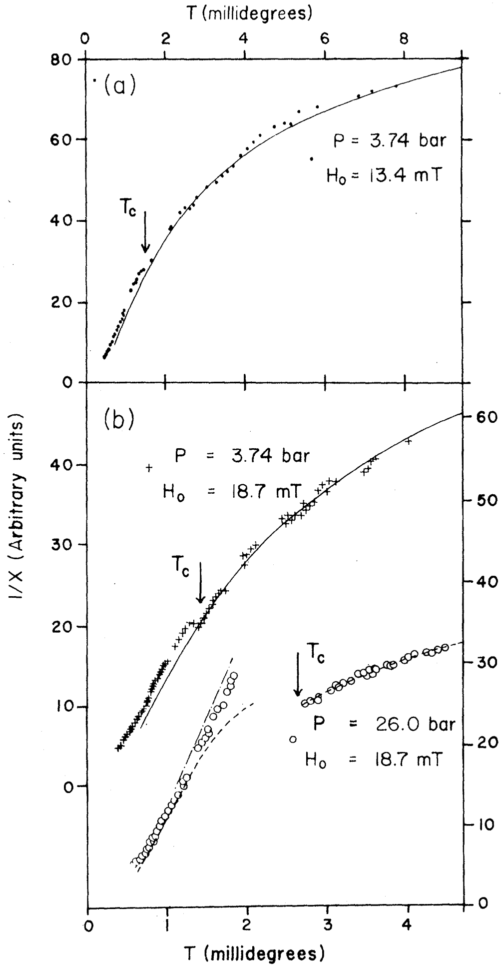

FIG. 1. Inverse susceptibility vs temperature. The solid lines represent the best fit to the data assuming $\theta = 0.38$ mK [see Eq. (1)]. The resulting values of B/A are listed below along with the nominal orientation of the grafoil. (a) $B/A = 3.04$ mK, ⊥ (T indicated on top scale). (b) Crosses, $B/A = 2.96$ mK, || (T on lower scale, χ^{-1} on left); circles, $B/A = 2.14$ mK, || (χ^{-1} on right scale). Dashed line represents the fit with a weak-coupling correction for the effect of superfluidity on the liquid below T_c; dash-dotted line represents the fit with the term $A = 0$ [see Eq. (1)] below T_c.

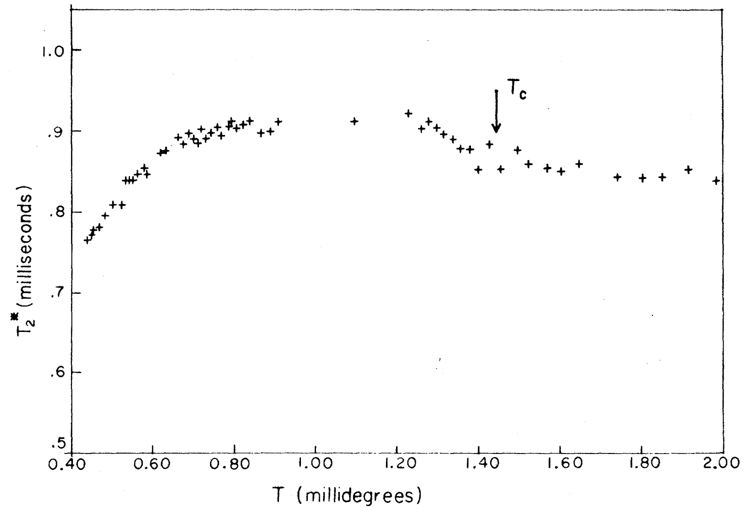

FIG. 2. Observed free-induction decay time, T_2^*, vs T for data in Fig. 1(a).

implies that for this particular pressure 95 times as many atoms have the constant susceptibility as opposed to the Curie-Weiss one.

In a separate series of adsorption isotherms using the point-A method[7] we found that the first monolayer was completed on the Grafoil with an amount 11.8 cm^3(STP)/g. Also we determined the internal volume of the Grafoil so that the amount of liquid both inside and around the Grafoil could be calculated. An important contribution comes from liquid external to the sample within the tube leading down to the main cell. A probe of the distribution of the rf magnetic field at the end of the coil showed that about 40% of the liquid signal came from here. The net result of these calculations is that for this pressure (3.74 bars) 0.89 of the monolayer appears to possess the Curie-Weiss behavior. For pressures 0, 6.23, and 26.0 bars, this number becomes 0.86, 0.95, and 1.19, respectively. Thus we have a small monotonic increase in this effective number of layers with pressure. This measurement does not, or course, distinguish where the spins are. In fact heat capacity measurements in films of ^{3}He at higher temperatures indicate that the two monolayers may solidify.[8]

The low-temperature behavior of the susceptibility for $P = 3.74$ bars taken at three different

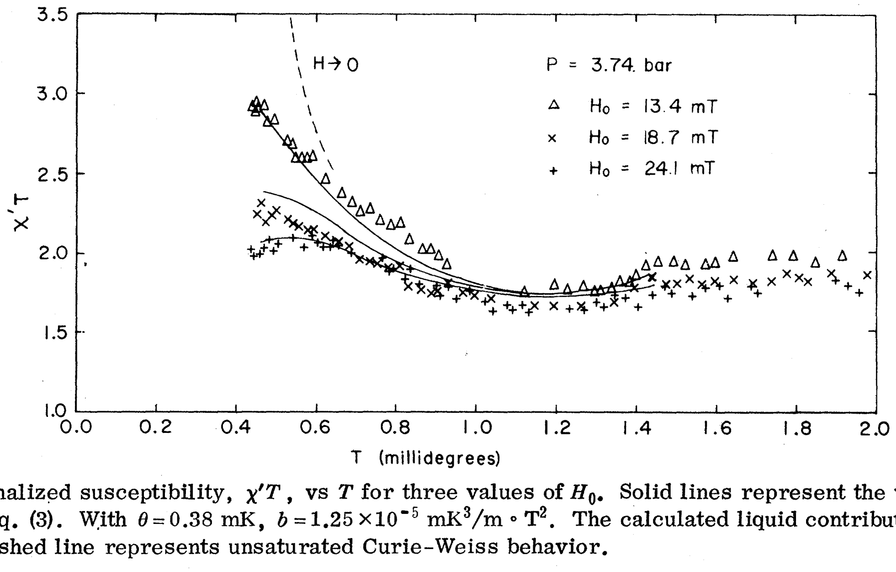

FIG. 3. Normalized susceptibility, $\chi' T$, vs T for three values of H_0. Solid lines represent the values of $\chi' T$ calculated using Eq. (3). With $\theta = 0.38$ mK, $b = 1.25 \times 10^{-5}$ mK^3/m $\circ$ T^2. The calculated liquid contribution has been included. The dashed line represents unsaturated Curie-Weiss behavior.

magnetic fields is displayed in Fig. 3. The values plotted are $\chi' T$ where χ' ($\equiv \chi/B$) is the susceptibility renormalized to the effective Curie constant. The amplitudes of the signals determining the susceptibility were measured to within an accuracy of 2% while the uncertainty in the absolute values of the temperature scale is estimated to be about ± 3%. As can be seen, the higher values of magnetic field depress the values of $\chi' T$ in the region below 0.8 mK compared with the $\chi' T$ values for the lowest field 13.4 mT.

With all fields used χ deviates from Eq. (1) below 0.8 mK. One way of analyzing these data is to consider the Landau-Ginzburg model for a magnetic phase transition which predicts a free energy of the form

$$F = \alpha(T - \theta)M^2 + 2b\alpha^3 M^4 - MH, \tag{2}$$

where $\alpha \equiv 2k_B/\gamma^2\hbar^2$. Then if $M' \equiv 2M\alpha/H$, the free energy is minimized when

$$1 = M'(T - \theta) + bM'^3 H^2, \tag{3}$$

where M' is the magnetization normalized so that $M' \sim T^{-1}$ as $T \to \infty$. In our case M' is the renormalized susceptibility minus the "liquid" part, i.e., $M' = (\chi - A')/B$ where A' is the value of A reduced to its calculated superfluid value. The solid lines in Fig. 3 represent the best fit of Eq. (3) to the data shown below 1 mK. ($\theta = 0.38$ mk, $b = 1.25 \times 10^{-5}$ mK^3/m$\cdot$T^2.) The values for A and B were obtained from the high-temperature fit ($T > T_c$). Only θ and b were variables for the low-temperature fit. The value of the maximum spontaneous magnetization ($T \to 0$) can be estimated by comparing M_0 to $\frac{1}{2}N\gamma\hbar$. This ratio becomes $(\theta/b)^{1/2}\gamma\hbar/2k_B \sim 0.15$.

Béal-Monod and Doniach[9] have suggested that the mechanism for ferromagnetism is closely related to the presence of high density liquid near the surface. This must be reconciled with our measurement of only the amount of magnetization for one "solid" monolayer. Guyer[10] has suggested that vacancies in the "solid" layer may induce ferromagnetism. It should be emphasized that the good fit we obtain with Eq. (3) does not necessarily indicate the onset of a conventional ferromagnetic transition. The possibility of effects due to reduced dimensionality should be considered; however, the applied field should reduce the available degrees of freedom and make the use of a Landau-Ginzburg model justifiable. Studies of this system as $H \to 0$ to look for possible effects of phase fluctuations[11] should be very useful.

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