RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 27, NUMBER 11 1 JUNE 1983

Evidence for ferromagnetic order in the boundary layer of ^{3}He on graphite

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(Received 2 March 1983)

Recent measurements of the surface magnetism of ^{3}He on graphite show that the surface susceptibility χ_s in a magnetic field would increase rapidly at temperatures below the temperature estimated for a ferromagnetic transition if the applied magnetic field were zero. An anisotropic frequency shift develops at low temperatures and is consistent with the magnetization coming from a two-dimensional sheet of spins. A low-temperature decrease in the spin-spin relaxation time gives another indication of the two-dimensional character of this system.

The boundary layer of ^{3}He provides a unique system for the study of two-dimensional magnetism. The surface layer of ^{3}He has been shown to have a ferromagnetic tendency on several substrates.[1–3] On graphite, the surface layer of ^{3}He is composed of a single monolayer of highly compressed solid,[4] while, because of the epitaxial mismatch, the second monolayer is probably liquid, at least at low pressures.[5] Above the surface, there is bulk liquid behaving as a degenerate Fermi liquid, so that the localized spins at the surface form a separate system distinct from the bulk. The degree to which this system behaves like a two-dimensional magnetic system depends on the type and range of interactions which the solidlike spins have with the bulk liquid as well as with the substrate.

Recent studies[6] of the heat conduction across boundaries between Pt particles and ^{3}He show that magnetic coupling across the boundary may account for most of the conductance. Another experiment[7] has shown that an anomalously high degree of coupling exists between ^{19}F spins in tiny Teflon spheres and ^{3}He spins. Even though the surfaces in these experiments are far more complex, the surface layer of ^{3}He must play a vital role in the magnetic coupling observed.

In this paper we will present results of the measurement of NMR free-induction-decay (FID) signals of ^{3}He on Grafoil. The Grafoil is completely filled with liquid, but the contribution of the degenerate Fermi liquid to the signal is subtracted out. All of the data presented here result from tipping the magnetization less than 10°. At temperatures $T > 1.6$ mK, the surface susceptibility χ_s follows $1/(T-\theta)$ very well (with $\theta = 0.80$ mK). The results reported here differ from previous work[2] in that we cool below the temperature θ where χ_s would diverge according to an extrapolation of the Curie-Weiss law from higher temperatures. This value of θ appears to vary between different batches of Grafoil (and to a lesser degree with the technique used for analysis), but is fairly consistent within a given batch. We have utilized three samples from the current batch of Grafoil,[8] and they all have given approximately the same results. We have also measured a frequency shift consistent with two-dimensional polarized spins.

Figure 1 shows the temperature dependence of the susceptibility of the solid layer χ_s, where χ_s is normalized to follow $\chi_s = 1/T$ in the limit of high temperatures where $T >> 1$ mK. For reference we have indicated the extension of the Curie-Weiss law to lower temperatures on the figure. As for the expected transition we must emphasize that the applied magnetic field is high compared with any dipole field. We are not close enough to the phase transition

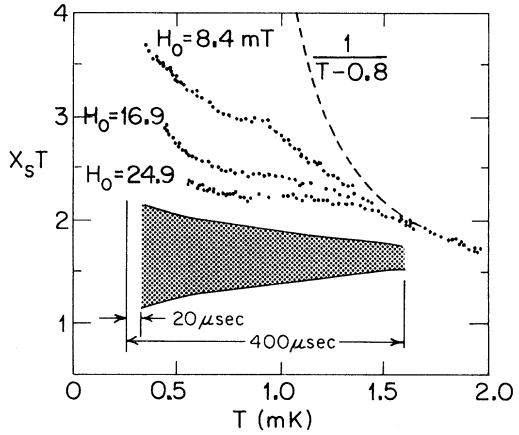

FIG. 1. The product of the normalized value of the surface susceptibility χ_s and temperature T. The static applied fields for the three sets of points are indicated. The pressure of the ^{3}He was 0.8 bar. The inset shows the envelope of a typical FID signal at 0.55 mK with $H_0 = 29.9$ mT.

($H \ll 3$ G) to look for critical behavior in the magnetization. Nevertheless, we can see that there is some structure in $\chi_s T$ near $T = 0.8$ mK. The value of the magnetization at 0.8 mK scales roughly as $H^{0.7}$.

Estimates of the susceptibility obtained from free-induction-decay signals depend crucially on the ability to extrapolate the signal back to the time of the rf pulse. The inset in Fig. 1 shows a typical free-induction-decay at a low temperature and the dead time is due mainly to the recovery of the signal coil from the rf pulse. All of our Grafoil samples give a similar line shape and one can see that there is not a simple exponential decay for the FID signal. We have used several models to extract the amplitude information. One model has been to assume that we have two distinct regions (layers) with the same frequency, but greatly different values of ring down time T_2^*. One layer is a very localized monolayer of ^{3}He at the surface with negligible exchange ($T_2 = 40$ μsec), and the second is a ferromagnetic surface layer in the liquid.[9] An analysis of this type has been carried out by Ahonen *et al.*[1] on carbon powder and by us for data on Grafoil.[10] Recent theoretical work[11] suggests, however, that substantial indirect exchange may occur in the first solid layer through the liquid.

An alternative explanation for the free-induction-decay line shape is that there is an inhomogeneous frequency shift in part of the sample. Possible sources of this shift include the anisotropic diamagnetism of graphite or possibly a chemical shift in the surface layer. We have analyzed the data in Fig. 1 assuming (somewhat arbitrarily) that 10–15% of the signal is at a slightly different frequency and has a longer ring down time than the majority, and that each component was exponential. This analysis fits the FID signals very well with a frequency difference of 300 to 670 Hz depending on H_0. However, for a two-dimensional array of spins, an amplitude which obeys the relationship $\exp(-\alpha t \ln \alpha t - \beta t)$ is expected,[12] where α and β are constants. We have tried using this time dependence and we get reasonable fits, but there are too many degrees of freedom to distinguish between these shapes. Analysis of free-induction-decay envelopes alone cannot determine the location of the magnetization.

Additional information can be gained by examining the frequency of the FID signals and we have observed an anisotropic frequency shift on Grafoil as shown in Fig. 2(a). A polarized two-dimensional sheet of spins produces a frequency shift due to the local field of the oriented neighboring spins. This shift is expected to follow[12]

$$\Delta\nu = \Delta\nu_{\max} P(1 - 3\cos^2\phi) \ , \qquad (1)$$

where P is the degree of spin polarization and ϕ is the angle between H_0 and the normal to the surface. The maximum positive value of the frequency shift

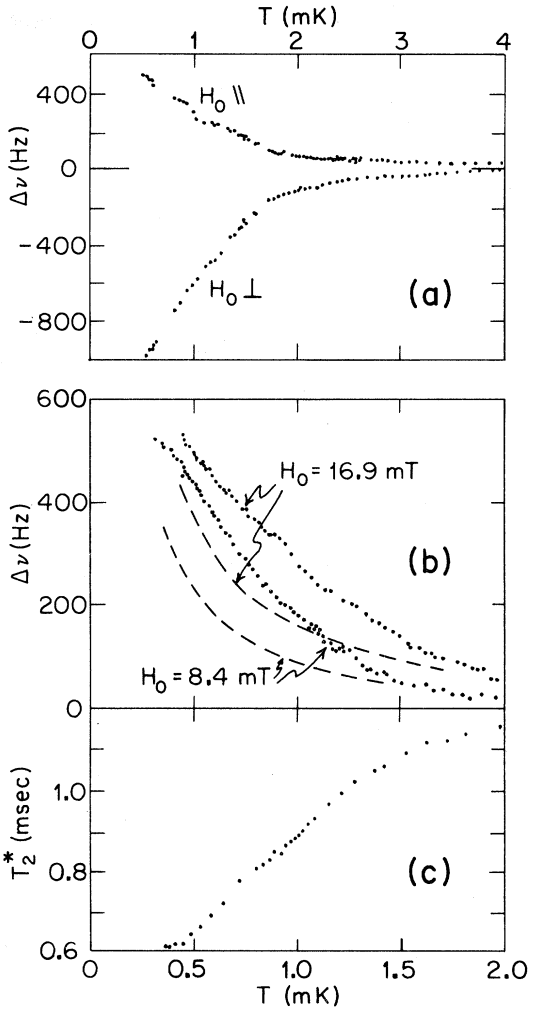

FIG. 2. The measured frequency shift vs temperature (points) at the indicated values of H_0. (a) Identical samples placed parallel and perpendicular to H_0 ($H_0 = 16.9$ mT). (b) Parallel oriented sample in two different magnetic fields. The dashed lines show the values of the frequency shift calculated from the values of $\chi_s T$ in Fig. 1. (c) The measured values of the FID decay time T_2^* vs temperature ($H_0 = 8.4$ mT).

$\Delta\nu_{\max}$ was obtained by calculating the local field of a two-dimensional (2D) triangular dipole lattice with nearest-neighbor spacing of 3.32 Å (corresponding to the density of our solid monolayer). The spin polarization P can be estimated by using the data for $\chi_s T$ which gives us the excess magnetization above the Curie law. Our triangular lattice will give a frequency shift of 5.17 kHz with $\phi = 90°$ when fully polarized. We have compared these estimates with the measured frequency shifts for two values of H_0 in Fig. 2(b). The estimates are qualitatively similar to the observed shift, but run somewhat smaller. For the data with $H_0 = 16.9$ mT and $T = 0.44$ mK, the max-

imum polarization as derived from Fig. 2 is about 0.13, while it is 0.085 as derived from Fig. 1.

Figure 2(c) shows typical behavior of the time T_2^* where the FID signal has decayed to $1/e$ of its initial value. The trend towards decreasing values of T_2^* at lower temperatures is seen consistently on all Grafoil samples, but is opposite to the results on carbon powders,[1] and more recently on 150-Å MgO powders.[13] Normally T_2 for 3D ferromagnets increases as the temperature is lowered towards the transition; however, the reverse is predicted for 2D magnetic systems.[12] Our results that the frequency shift appears to obey Eq. (1) and that T_2^* decreases at low temperatures give strong support to the idea that the surface layer exhibits two-dimensional behavior. It would obviously be advantageous (but experimentally very difficult) to study this system in the limit of extremely low applied fields to see if spontaneous magnetization develops, or whether the system exhibits two-dimensional Heisenberg characteristics with suppressed ordering.[14]

Theoretical models of surface magnetism dealing with exchange in just the localized layer[15] or with the modified liquid at the surface[9] do not quantitatively predict ferromagnetic transition temperatures. The recent theory of Jichu and Kuroda[11] used a more realistic model of ^{3}He at surfaces. They find that a Ruderman-Kittel-Kasuya-Yosida type of indirect interaction with liquid spins modulating the exchange between localized surface spins can give values of θ, the ordering temperature, of the correct size (0.45 mK). This interaction is very sensitive to the degree of localization of the surface layer which may account for the experimental variation in the measured values of θ.

ACKNOWLEDGMENTS

We wish to acknowledge the assistance of T. J. Bartolac and K. Luey in the early phases of this experiment. We also thank C. M. Gould, R. C. Richardson, and S. E. Trullinger for valuable comments and discussion. This work is supported by National Science Foundation Grant No. DMR 82 00661.

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