Evidence for Ferromagnetic Ordering of ^{3}He Films on Graphite

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Abstract

Recent experiments at USC have shown that two-dimensional films of ^{3}He on graphite order ferromagnetically at finite temperatures ($T > 0$) for densities above 20 atoms/nm^2. These results, obtained by measuring NMR on these films in the zero field limit, appear to contradict several studies which concluded that there is no spontaneous order by measuring magnetic susceptibility through conventional NMR in finite magnetic fields, and heat capacity in zero field. The low field limit is important in understanding these two-dimensional magnetic systems. At higher temperatures and/or fields, these films have been described in terms of the multiple spin exchange model. However, two-dimensional systems are extremely sensitive to anisotropies such as nuclear dipole interactions, even when they are several orders of magnitude less than the dominant exchange mechanism.

Key words: NMR; grafoil; ^{3}He; heat capacity

^{3}He films on graphite offer an excellent opportunity to study magnetism in two-dimensions. Rapid physical exchange between the ^{3}He atoms dominates the thermal behavior of these films. The thermodynamic quantities magnetization and heat capacity have been measured extensively at millidegree and submillidegree temperatures. By varying the density of these films, a wide range of effective exchange rates can be observed with the sign of the effective exchange rate switching from antiferromagnetic to ferromagnetic as the density is increased.[1]

The dominant exchange process has been modeled by a nearest neighbor Heisenberg exchange process,[2] but the multiple spin exchange model provides a more realistic picture of the thermal properties across the full range of densities.[3,4,6] Because these models have continuous symmetry, the expectation is that there will be no phase transition at finite temperatures due to the Mermin-Wagner theorem.[5] In fact experimentalists studying both magnetic susceptibility using conventional NMR techniques [7,8,6] and heat capacity experiments [9,10] have concluded that there is no finite temperature phase transition in these films.

Nevertheless, NMR experiments performed in the zero field limit (using a SQUID technique) have resulted in the opposite conclusion. Those experiments performed first by Friedman *et al.* [11,12] in filled pores of Grafoil and more recently by Bozler *et al.* [13] on ^{3}He films with second layer densities in the ferromagnetic region, have come to the conclusion that finite order occurs in these films in the zero field limit at finite temperatures. These different conclusions lead to the question of why those results should be different? And then, what is the physical significance of this result?

We contend that both the conventional NMR and heat capacity experiments were not sensitive to ordering for several different reasons. They include the dramatic effect of a finite magnetic field, the inherent heterogeneity of the Grafoil substrate, and the weak critical behavior that is characteristic of magnetic ordering.

For NMR experiments, this situation can be best illustrated by comparing recent data from USC with the earlier results from Schiffer *et al.* In Fig. 1, we show data from Schiffer *et al.* [8] for the magnetization with the coverage 24.8 atoms/cm^2 and with their applied

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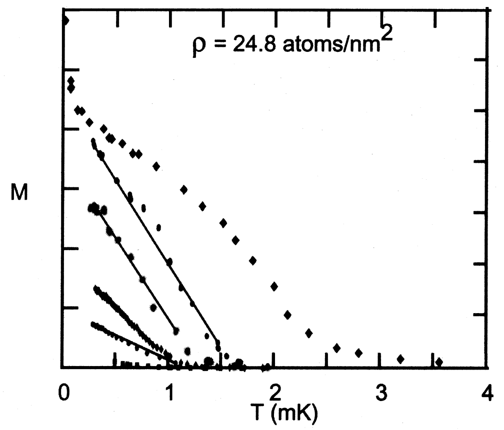

Fig. 1. Comparison of NMR magnetizations measured by SQUID and higher field conventional techniques. Top (◇) from Schiffer *et al.* with total coverage 22.8 atoms/nm^2 and an applied field of 14.3 mT. The lower sets of data are from the USC experiments with (○) 24.2, (x, △) 22.5, (◇) 21.8, (+) 21.3, (□) 20.5 atoms/nm^2. All the USC data were taken at 0.35 mT, except for the lowest coverage data which was taken at 0.48 mT.

field of 14.3 mT. Below those points, we place our very low field data.[13] As indicated in the figure caption, the most comparable coverage is represented by the top line. (The data are arbitrarily scaled so that they agree at zero temperature ignoring the last two points in the Schiffer *et al.* data that are attributed to polarization of the first monolayer.) In essence, the effect of the applied field is to greatly increase the polarization of the ^{3}He spins in the intermediate temperature region where $T \approx J$, obscuring the transition region close to 1 mK.

Fig. 1 also illustrates two other aspects of this system. First, there is likely a two-phase region (one low magnetization, the other ferromagnetic) between total coverages of approximately 20 to 24 atoms/nm^2. Our conclusions regarding this point coincide with those of Schiffer *et al.* Second, there is a significant rounding of this transition, even at the lowest fields. We think that this rounding is a spreading of transition temperature due to the heterogeneity of the grafoil substrate and also because of finite size effects. In fact, we cannot entirely rule out finite size effects as the cause of the magnetic ordering, but we think that it is much more likely that weak long-ranged interactions are responsible.

Even a modestly spread out transition greatly reduces the likelihood of observing an anomaly in heat capacity since the critical exponent will be small. (A 2D Ising transition would result in a logarithmic divergence.)

The influence of dipole-dipole interactions and weak anisotropies on Heisenberg model systems has been studied in quasi-two-dimensional solids and thin magnetic films.[14,15] Dipolar interactions or weak anisotropies result in a modification of the spin wave spectrum and stabilize a magnetic phase. The importance of weak, but long ranged effects, is that they become greatly amplified so that the reduction in the transition temperature is only logarithmic in the ratio of the dominant exchange and anisotropy energies. Ignoring these weak effects leads to incorrect conclusions about the nature of the low temperature state of these interesting 2D systems.

Acknowledgements

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