

Ferromagnetism of ^3He Films in the Low-Field Limit

H. M. Bozler, Yuan Gu, Jinshan Zhang, K. S. White, and C. M. Gould

Department of Physics and Astronomy, University of Southern California, Los Angeles, California 90089-0484

(Received 9 July 2001; published 25 January 2002)

We provide evidence for a finite-temperature ferromagnetic transition in two dimensions as $H \rightarrow 0$ in thin films of ^3He on graphite, a model system for the study of two-dimensional magnetism. We perform pulsed and cw NMR experiments at fields of 0.03–0.48 mT on ^3He at areal densities of 20.5–24.2 atoms/nm². At these densities, the second layer of ^3He has a strongly ferromagnetic tendency. With decreasing temperature, we find a rapid onset of magnetization that becomes independent of the applied field at temperatures in the vicinity of 1 mK. Both the dipolar field and the NMR linewidth grow rapidly as well, which is consistent with a large (order unity) polarization of the ^3He spins.

DOI: 10.1103/PhysRevLett.88.065302

PACS numbers: 67.70.+n, 76.60.–k

Two-dimensional magnetic systems order only at zero temperature when their interactions have continuous symmetry [1]. However, the ordering temperature of these systems may become finite when they have a weak anisotropy [2,3], especially when the weak anisotropy is long ranged. This is the case for two-dimensional films of ^3He on graphite [4,5]. These films have large exchange rates which can be described by the multiple spin exchange model over a wide range of coverages [6]. Much of the interest in these films comes from the dramatic changes in their structural and magnetic properties caused by small changes in density, including a change in the sign of the effective exchange [4,7]. In addition to exchange energy, these films have nuclear dipolar couplings which are more than 3 orders of magnitude smaller than the leading order exchange. Thus ^3He films provide a useful test system to observe the behavior of real two-dimensional magnets in the presence of weak anisotropy.

In our experiments we have evaporated ^3He films onto a graphite substrate. We report here the results of coverages between 20.5 and 24.2 atoms/nm². The most striking feature of this range of coverages is the rapidly increasing ferromagnetic tendency associated with the second layer [5]. In this range, the first monolayer is solid with a very low exchange rate [8], while the second solid layer is incommensurate with the first monolayer and has a much higher exchange rate. Third layer promotion occurs near 18 atoms/nm², and therefore a partial fluid layer lies above the magnetic layer throughout our coverage region. The Grenoble group has analyzed in detail NMR and heat capacity data above 4 mK using the multiple spin exchange (MSE) model [7,9] assuming a single incommensurate phase for the second layer, but the possibility of a mixed solid phase (part incommensurate ferromagnetic, part low magnetism) has been suggested by other experiments [5] and has not been excluded in the detailed analysis of multiple spin exchange [7]. In fact, earlier work by Schiffer *et al.* [10] extending to temperatures far below 1 mK reported results that were more consistent with a mixed phase.

Previous experiments have demonstrated that a single effective exchange rate is insufficient to describe both NMR and heat capacity experiments throughout this region; however, as coverages approach 24 atoms/nm² the nearest neighbor Heisenberg model works well [11]. Fitting data to a high-temperature spin exchange model with a single effective exchange constant J remains a convenient (if not precise) method for characterizing the high-temperature data with J ranging from below 0 (below 20 atoms/nm²) to close to 2 mK at ~ 24 atoms/nm².

With the exception of the studies by Friedman *et al.* [3,12] on ^3He which used a SQUID NMR technique [13] with filled pores, all of the previous NMR studies used conventional techniques with applied fields greater than 6 mT. The Larmor frequencies in those (conventional NMR) experiments were much smaller than the exchange rates, but nevertheless the applied fields were large enough to substantially enhance the magnetization in the vicinity of $T \sim J$, effectively masking any spontaneous magnetization—if it should occur. On the other hand, the very low-field studies by Friedman *et al.* indicated the possibility of spontaneous magnetization although their results in filled pores are difficult to compare directly with the numerous experiments using monolayer films.

Our new NMR measurements were performed in applied fields below 0.5 mT using a SQUID technique similar to the one used by Friedman *et al.*, except that an indirect cooling method was employed [14]. Grafoil (grade GTY) substrates [15] were diffusion bonded to silver foils for thermal contact. We lithographically divided the foils into narrow wires prior to the diffusion bonding process to reduce eddy currents in order to obtain the short pulse recovery times required for SQUID NMR and to ensure complete penetration of the time dependent (H_1) field. In order to minimize eddy current effects, we oriented the static magnetic field perpendicular to the nominal plane of the Grafoil. In this orientation, the magnetization experiences a competition between the static field and the dipolar field and can eventually flop into the Grafoil planes at sufficiently high polarizations and low magnetic fields. In

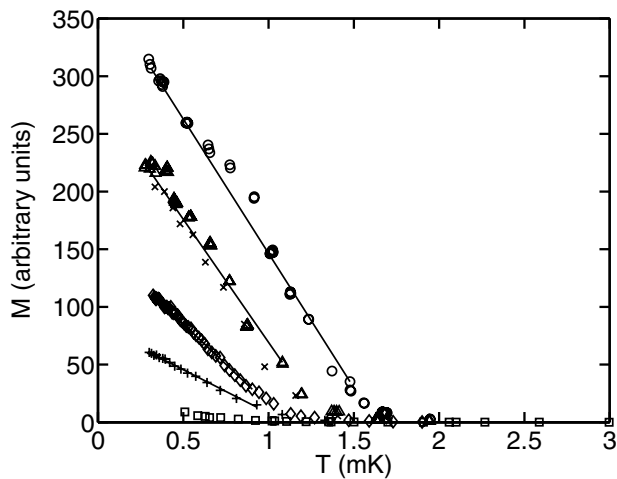


FIG. 1. Magnetization as a function of temperature for a variety of coverages. (\circ) 24.2, (\times , \triangle) 22.5, (\diamond) 21.8, ($+$) 21.3, (\square) 20.5 atoms/nm². All data were taken at 0.35 mT, except for the lowest coverage data which were taken at 0.48 mT.

the data shown in this paper, the field and/or temperature is always sufficient to hold the magnetization approximately perpendicular to the Grafoil sheets. At higher temperatures we used pulsed NMR, but swept frequency continuous wave measurements were needed wherever the effective free induction decay time fell below 300 μ s (typically below \sim 2 mK). Our silver foil contact system allowed us to reach temperatures as low as 0.3 mK with the lowest temperature being limited by long equilibrium times.

³He coverages were measured using ³He isotherms with an *in situ* pressure gauge located near the sample cell on the nuclear demagnetization cryostat. Comparing results with other work can be accomplished by anchoring the coverage scale to the point where the effective exchange rate crosses zero. This point ranges from approximately 20 atoms/nm² in Refs. [5] (Fig. 45) and [10] down to 19.8 atoms/nm² in Ref. [16] and 18.9 atoms/nm² in Ref. [17]. Our coverages give the onset of ferromagnetism at 19.6 atoms/nm² \pm 2%.

Our experimental results provide evidence for spontaneous magnetization by (1) the temperature dependence of the magnetization, (2) the rapid increase in the NMR linewidth at low T with a concomitant shift in the NMR frequency, and (3) the large magnetization observed at finite temperature even in the low-field limit.

Figure 1 shows the magnetization vs temperature for several coverages ranging from densities where the second layer barely shows a ferromagnetic tendency, to the coverage where the maximum effective exchange has been observed [5]. Above 2 mK the magnetization appears quite small. We see that the rapid increase in magnetization is initiated over a relatively narrow range of temperature. In addition, the magnetization appears to increase approximately linearly at temperatures below an “onset” temperature near 1 mK. The onset of magnetization remains

rounded even at our lowest applied fields; however, we believe that the rounding is a result of the considerable heterogeneity of Grafoil samples. The existence of a finite magnetization is well supported by the frequency shift and linewidth data shown below.

Another feature of the magnetization data in Fig. 1 is the apparent increase with increasing coverage in the number of spins participating in the magnetization—even at our lowest temperatures. Since the second layer density increases only by a few percent [4], we interpret this increasing magnetization as evidence that, for coverages below 24 atoms/nm², we have a mixed phase as suggested by Schiffer *et al.* [10]

Below the onset temperature, we observe a rapid increase in the NMR linewidth shown in Fig. 2. This increase is readily observed as a decrease in the effective T_2 of the free induction decay signals from pulsed measurements, and then in direct measurements of the linewidth in the cw measurements at lower temperatures. Typical NMR lines are shown in the inset of Figure 2. Along with the increasing amplitude and linewidth we see a frequency shift caused by the increasing dipolar field from the aligned ³He spins and the appearance of a characteristic distortion in the line shape at temperatures slightly below 1 mK with the central line shifting towards lower frequencies while portions of the NMR line extend to frequencies above the Larmor frequency. These extremely broad NMR lines were also observed in Ref. [10].

In the simplest case where the static field and polarization of two-dimensional ³He spins are perpendicular to the graphite planes, a dipolar field (λM_0) results in a negative frequency shift. Much of the characteristic line shape that we observe at low temperatures can be attributed to the distribution of plate angles for surfaces in Grafoil. These

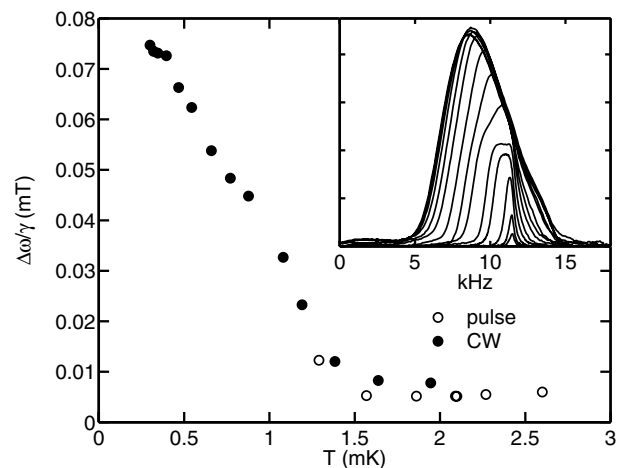


FIG. 2. Linewidth as a function of temperature for a coverage of 22.5 atoms/nm² at 0.35 mT based on the fitting procedure described in the text. γ is the gyromagnetic ratio for ³He. The inset shows typical absorption lines taken from cw NMR ranging from 2 mK (smallest) down to 0.3 mK (largest).

angular distributions have been studied by neutron scattering [4,18,19] and x-ray scattering [20] with the probability distribution shown to be a Gaussian centered around the normal ($\theta = 0$) to the Grafoil plus a randomly distributed portion so that $P(\theta)/\sin(\theta) = a \exp(-\theta/0.3145)^2 + b$, where a and b are normalized to a fraction of spins in the central distribution and at random angles. Reported values of the random fraction range from 50% [4,18] down to 33% [20]. In order to calculate the distribution of frequencies that simulate our NMR lines we need to calculate the angle of the magnetization (which is not exactly the angle of the applied field at low fields) and then the resulting NMR frequency, assuming (i) a temperature dependent average dipolar field (λM_0), (ii) the equations for spin dynamics from Friedman *et al.* [12], and (iii) a temperature dependent intrinsic linewidth. We can approximately fit our NMR absorption lines to this model; however, this process works best when we assume that only 20%–25% of the spins are on the randomly distributed plates. We find that fits to the data work best when the intrinsic line shape is described by a Gaussian rather than a Lorentzian (especially at temperatures below 1 mK). This fit results in estimates for the Gaussian linewidth and dipolar field produced by the ^3He spins.

Figure 2 shows the effective linewidth resulting from fits to the NMR lines. We can see that the NMR linewidth grows rapidly below the onset temperature. We show data at 22.5 atoms/nm² but the linewidths at other coverages are very similar in both magnitude and temperature dependence. A further contribution to the linewidth (and shape) is the variation of the local dipolar field across a finite sheet and at edges.

The negative shift of the maximum in the absorption peak is a measure of the dipolar field. In Fig. 3 we plot the dipolar field λM_0 based on our fits to the NMR lines.

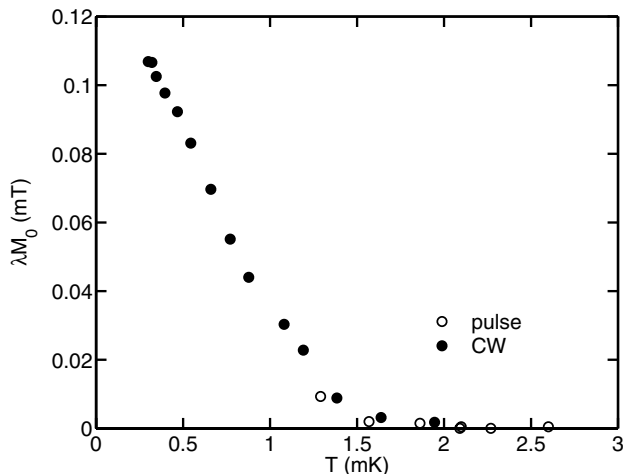


FIG. 3. Average dipole field (NMR resonance shift) λM_0 as a function of temperature for a coverage of 22.5 atoms/nm² at 0.35 mT.

We see that the dipolar field increases rapidly below the onset temperature. We also see that the dipolar field as well as the linewidth are proportional to the magnetization in Fig. 1 which was obtained by directly integrating the NMR lines. The magnitude of the dipolar field extrapolates to close to 1.5 mT at zero temperature which is somewhat less than the dipolar field we would expect for a fully polarized sheet of spins at the second monolayer density but comparable to the shifts observed by Schiffer *et al.* [10] at similar coverages with much higher applied fields. The dipolar field from the higher (24.2 atoms/nm²) coverage is approximately 10% greater.

In Fig. 4 we show the measured magnetization over a wide range of temperatures and at four magnetic fields on a logarithmic scale. An important aspect of the data in Fig. 4 is the enormous (more than 4 orders of magnitude) increase in magnetization. In fact, if we normalize our observed signals to the calculated high-temperature polarization (including contributions from the first and fluid layers), we obtain low-temperature polarizations of the second layer which are very close to 100%. At the lowest applied field (0.03 mT) there is a very high slope near 1.3 mK (magnetization doubling each 10% change in temperature) and near that temperature we see a crossover from the high-temperature region, where the magnetization is proportional to the magnetic field, to the low-temperature region, where the magnetization is independent of the field.

The question arises whether the ordering which we observe is intrinsic to the ^3He /Grafoil system and thus a property of such two-dimensional magnetic systems, or

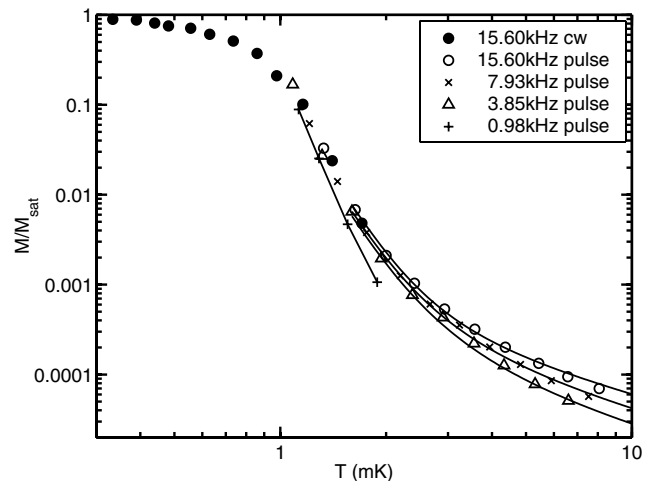


FIG. 4. Total magnetization at 22.5 atoms/nm². The open symbols were from pulsed NMR with applied fields of (\circ) 0.48 mT, (\times) 0.24 mT, (\triangle) 0.12 mT, ($+$) 0.03 mT. The closed symbols (\bullet) were from cw NMR at 0.48 mT. Additional cw data (not shown) from lower fields agree well with the 0.48 mT results. M_{sat} was the estimated low-temperature limit of M . The solid lines, while intended as “guides to the eye,” were the result of a high-temperature Heisenberg model expansion in the case of the three largest fields.

whether it is caused by some artifact of an imperfect substrate. We note that, at coverages below 19.5 atoms/nm², we did not observe any evidence of a low-temperature order. Thus, the ordered system in the low-field limit that we observe coincides in coverage with the change in sign of the effective exchange. If the order were solely due to defects in the substrate, it would seem unlikely that these effects would disappear within this tiny change in coverage. In principle, heat capacity measurements should also indicate the existence of a magnetic ordering. In fact, measurements in this coverage range have been used to argue that no such order exists [21]. However, it is likely that any divergence in the heat capacity would be extremely weak, making it extremely difficult to observe anomalies when samples are heterogeneous.

The dominant interaction between ³He atoms in these films is particle exchange which is well described by the MSE model [6]. By itself, this model predicts phase ordering only at zero temperature, consistent with the Mermin-Wagner theorem [1,22]. However, weak anisotropies modify the spin-wave spectrum, so that long wavelength magnons no longer destroy order [2,23]. Recently there has been a suggestion by Gov *et al.* [24] of a large anisotropic effect due to correlations in zero-point motion; however, this proposal requires a substantial modification of the MSE model and we do not yet have a way to experimentally test this idea.

The reduction of magnetization by spin waves in the pure Heisenberg model has been calculated by Kopietz *et al.* [22], but any anisotropy breaks the Heisenberg symmetry. Yafet *et al.* [2] calculated the reduction of magnetization caused by spin waves in the presence of dipole-dipole interactions. They showed that these interactions introduce a linear term in the spin-wave spectrum that in turn stabilizes the ordered state. Their numerical estimates for the magnetization (in a square lattice) are consistent with our data. In the case of metal films with weak anisotropy this magnetization is described by the relation [23] $\delta M/M_0 \equiv 1 - M/M_0 = 1 - (cT/4\pi J) \ln(T/\delta H)$, where δH is an anisotropic energy favoring perpendicular magnetization (including the applied field) minus the dipolar energy, and c is a constant of order unity. Because of the competition between exchange cycles in the MSE model, the correct value of J to use is uncertain. Nevertheless by putting in our values for $J \approx 2.2$ mK (at our highest coverage) and δH corresponding to our applied field minus our measured dipole field, we get $\delta M/M_0$ to fit our data when $c = 0.5$. Clearly a more complete theory which includes calculations on a triangular lattice is needed to complete a quantitative comparison. In addition, experiments on a more homogeneous substrate could clarify the nature of the ferromagnetic ordered state.

We wish to acknowledge the assistance of Barry Fink and useful discussions with Douglas Mills. This research

is supported by the National Science Foundation Grant No. DMR-9973255.

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