Magnetization of ^{3}He on Grafoil in the Low-Temperature Limit

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(Received 11 June 1993)

We have measured the magnetization of the ferromagnetic second layer of pure ^{3}He films adsorbed on Grafoil down to 0.075 mK. The zero-temperature magnetization is linear in second layer density (ρ_2) and rises by a factor of ~ 7 with a 6% increase in ρ_2 and less than a factor of 2 increase in the apparent exchange energy, indicating that the ferromagnetic phase coexists with a lower density phase. This is the first experiment to measure the low-temperature limiting magnetization of a 2D Heisenberg ferromagnet. Our data may also support Elser's model of a registered antiferromagnetic phase at lower ρ_2.

PACS numbers: 67.70.+n

Nuclear magnetization measurements [1–3] on thin films of ^{3}He atoms ($S = \frac{1}{2}$) show that while atoms in the completed first atomic layer is a paramagnetic solid with a vanishingly small exchange frequency, the second layer upon solidification displays both antiferromagnetic (AFM) and ferromagnetic (FM) tendencies depending on layer density. Similar results were obtained from ^{3}He films adsorbed on surfaces preplated with ^{4}He [4]. Recent specific heat measurements [5] suggest that the second layer solid has a rich phase diagram and that higher layers are liquid. In this Letter, we report new magnetization data on pure ^{3}He films at a variety of coverages extending to temperatures an order of magnitude lower than previous measurements. Our data suggest two-phase coexistence in the FM coverages and provide a variety of new information about the phases. We also find support for a registered AFM phase proposed by Elser [6] which is based on a *kagomé* net of AFM atoms.

The construction of the sample cell used in these experiments has been described in detail elsewhere [7]. The film substrate consisted of a stack of 37 "sandwiches" of two Grafoil [8] sheets fused to a single silver foil. Grafoil is exfoliated graphite whose surface consists of atomically smooth platelets ~ 100 Å in size. Tabs extending from the silver foils were then thermally linked to our copper demagnetization refrigerator. The ^{3}He magnetization was measured with an NMR coil around the stack and incorporated within a conventional cw NMR spectrometer. The rf from our coil seemed to be somewhat shielded by the stack, preventing us from making reliable measurements at high temperatures ($T > 5$ mK) where the magnetization was relatively small. A Pt wire NMR thermometer attached to the silver rod was calibrated from the ^{3}He melting curve.

Magnetization measurements at 464 kHz ($H \sim 14.3$ mT) were made at total coverages (ρ) of 0.133, 0.182, 0.190, 0.194, 0.204, 0.215, 0.227, 0.248, and 0.259 atoms/Å^2 by sweeping the magnetic field. Samples were prepared by admitting ^{3}He gas to the cell at $T \gtrsim 2$ K, after which they were annealed at higher temperatures. The surface area of the Grafoil was calibrated by comparing a magnetization isotherm at 3.24 mK with one at 3 mK by Franco, Rapp, and Godfrin [3] (the temperature offset is based on a later revision of their temperature scale [9]). The steep nature of this isotherm allowed calibration to within 1% of Franco's scale although a recent reanalysis of that data may lead to a slightly larger uncertainty [10]. The total surface area of 33.3 m^2 for the 2.55 g of Grafoil used is consistent with Greywall's 198 m^2 for 14.5 g [8].

At our lowest coverage, the first layer is solid and at its maximum density, and the second layer is a degenerate Fermi liquid with negligible magnetization at low temperatures. Thus the total magnetization of this coverage (at low temperatures) is equivalent to the first layer magnetization. This was found to obey the Curie law ($M = C/T$) to within the accuracy of our measurements ($|\Theta_{\text{Weiss}}| < 10$ μK), placing a new limit on the exchange energies in the completed first layer and verifying adequate thermal contact between the ^{3}He and the Pt wire thermometer to our lowest temperatures. Using the Curie constant (C) and the known density of the first layer (0.114 atoms/Å^2) [5], we could calibrate our magnetization in absolute units. Furthermore, the known first layer magnetization could be subtracted from the total magnetization at higher coverages to obtain the second layer magnetization (M_2) alone.

Data taken at higher coverages where the second layer is a solid fall into two groups: those taken at total coverages below and above ~ 0.200 atoms/Å^2. Previous data [1,3,4] show the second layer to be AFM at the lower coverages and FM at the higher coverages. We also observe different behavior in these two regimes and thus discuss them separately.

Our five highest coverages displayed FM behavior, with M_2 rising above the Curie law at low temperatures. Following earlier workers [1–5], we describe our data in terms of a 2D Heisenberg system, with the Hamiltonian

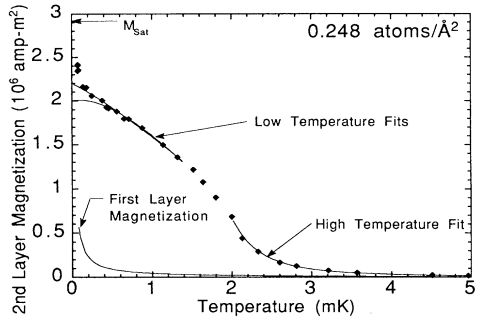

FIG. 1. Magnetization of second layer at total coverage of 0.248 atoms/Å^2. The solid lines are fits to the data as described in the text. The lower low-temperature fit is of the form in Ref. [2] and does not consider $k=0$ magnons.

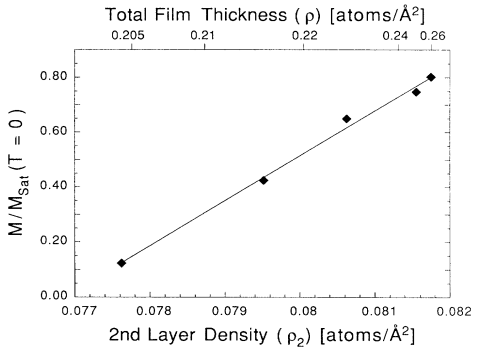

FIG. 2. Zero-temperature magnetization of the second layer as determined by our fits. That M_0 is linear in ρ_2 is evidence for two-phase coexistence.

$H=-Jk_B/2\sum_{i>j}\sigma_i\cdot\sigma_j$ (where $|\sigma^2|=1$, the sum is over nearest neighbors in the triangular lattice, and the exchange energy J is in units of temperature). From this, an expression for the magnetization can be derived as a function of temperature for $T\gtrsim J$ and for $T\to 0$. For $T\gtrsim J$ the data can be fitted to a high-temperature series expansion [11] involving only two free parameters, J and the number of spins. For $T\to 0$, in a magnetic field B, Kopietz *et al.* [12] calculated the magnetization for a finite 2D system by integrating the number of thermally excited magnons with $k\neq 0$:

$$M(T)=M_0\left[1-\frac{T^*/J}{2\pi\sqrt{3}}\left(1-\frac{T}{T^*}\ln[e^{T^*/T}-1]\right)-\frac{2/N}{[e^{T_B/T}-1]}\right].$$

In this model N is the number of FM spins in the 2D system, M_0 is the $T=0$ magnetization, $T_B=2B\mu/k_B$ where μ is the magnetic moment of the ^{3}He nucleus, and $T^*=T_B=8\pi^2J/L^2$ where L is in units of the lattice spacing (dimensionless), and the minimum nonzero magnon k vector is $4\pi/(\sqrt{3}L)$ in an ideal (rhomboidal) finite 2D system ($T^*\sim 20T_B$ in these experiments). This form differs from that in Ref. [2] by including the last term which accounts for the contribution of $k=0$ magnons. In an ideal 2D system, L also corresponds to the size of the system and $L^2=N$. We thus treat L^2 and N as equivalent, and, knowing the total number of spins in our samples, we obtain M_0, J, and N from fits to our data.

To obtain the above parameters, we first fit our high temperature ($\sim$2–5 mK) data to get preliminary values of J leaving the number of atoms as a free parameter. Using these values of J, we then fit the low-temperature data ($T\lesssim 0.9J$) for M_0 and N. We found that M_0 was significantly less than the saturation magnetization for the second layer (M_{sat}), so we refit the high-temperature data for J assuming that only a fraction (M_0/M_{sat}) of the spins was contributing to the FM state. A reiteration of this procedure did not significantly alter the values of the fit parameters. Typical fits to the data are shown in Fig. 1 (a low-temperature fit of the form given in Ref. [2] is shown for comparison), and values of M_0, J, and N are shown in Figs. 2, 3(a), and 3(b), respectively.

Over a narrow range in second layer density (ρ_2) (where ρ_2 is determined from a relation deduced by Greywall [5]) we find that M_0 rises linearly from about 12% to about 80% of its full saturation value, even though J does not change by even a factor of 2. This strongly suggests a "two-phase model" of the second layer in which the FM phase coexists with an AFM phase, with the FM phase being higher in density and probably incommensurate with the first layer. In this model, as the number of atoms in the second layer increases, more of its area converts to the higher density FM phase. This model is consistent with Greywall's [5] suggested phase diagram, taking his $R2b$ phase to be AFM, and his S phase

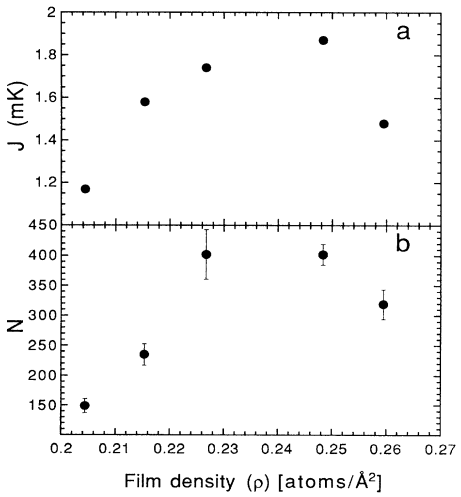

FIG. 3. (a) The exchange energy, J, and (b) the parameter N as a function of the total film thickenss.

to be FM. The maximum M_0/M_{sat} of ~ 0.8 is consistent with Godfrin, Ruel, and Osheroff [2], and suggests that $\sim 20\%$ of the spins do not contribute to the FM phase. One possible explanation is that the spins at the edges of the Grafoil platelets do not participate fully in FM exchanges. Assuming a typical 100 Å platelet holds ~ 1000 atoms, as many as $\sim 15\%$–25% could be on an edge. Alternatively, these missing spins may reside on platelets or particles which are too small to allow FM exchange. Scattering experiments [13] suggest that as much as $\frac{1}{3}$ of the Grafoil surface area is not in the form of large aligned platelets, but isotropically oriented smaller surfaces which may not support FM exchange.

A deviation of the magnetization data above the functional fit for $T \lesssim 0.2$ mK was present at each FM coverage and increased in magnitude with coverage. The number of additional *free* spins necessary to account for this deviation is on the order of one-third of all the atoms in the sample, and no similar deviation was observed in the AFM regime, ruling out faulty thermometry and isolated spins as causes. Perhaps this effect is due to density dependent FM exchange in the edge atoms discussed above.

Our values of J agree with earlier measurements [4, 5,10], but at our lowest two FM coverages we obtain somewhat larger values. Since we analyzed M_2 at lower temperatures, however, we would expect the FM islands' magnetization to more completely dominate the AFM contributions. Our counting only the FM fraction (M_0/M_{sat}) of ρ_2 also necessarily changes the fitted values of J.

The increase of J with ρ_2 (for $\rho \lesssim 0.24$ atoms/Å^2) can be explained within the two-phase model by postulating that the boundaries of the FM islands fluctuate due to the low expected interface energy in two dimensions. These boundary fluctuations and the reduced number of FM sites at the boundaries with which exchange is possible, will inhibit FM exchanges for atoms at the edges of the islands. Since at least 30% of the second layer atoms at our lowest FM coverage will be on an island edge, this would reduce the effective J for the entire island.

The decrease in J at our highest coverage was observed by previous workers [3–5,10] for $\rho \gtrsim 0.24$ atoms/Å^2 and is not easily attributed to simple compression of the FM phase [5] since J decreases by $\sim 21\%$ while ρ_2 increases by only $\sim 0.25\%$, requiring an unreasonably high Grüneisen constant ($\Gamma > 50$). Since similar FM behavior has been observed at similar densities in monolayer samples without a fluid overlayer [14], the exchange process probably only involves second layer atoms. One possible explanation is that FM exchange depends on vacancies or strong density fluctuations within the FM islands. As ρ_2 is increased to its maximum value and the fluctuating boundaries with the lower density AFM phase are eliminated, the number of such density fluctuations and thus the exchange frequency would then be reduced. This model is consistent with Greywall's [5] measurements which showed that while J decreased rapidly with increasing ρ for $\rho \gtrsim 0.24$ atoms/Å^2 there was a kink at $\rho \sim 0.26$ atoms/Å^2 above which J decreases less rapidly, presumably due to completion and subsequent compression of the FM layer.

The final parameter obtained from the fits to our data, N, might be associated with the average number of spins per FM island. This association is not, however, supported by the data, since N increases by only a factor of ~ 2.7 between $\rho = 0.205$ and 0.248 atoms/Å^2 while M_0 increases by a factor of 6.5 in the same range. Furthermore, we find that N decreases significantly at our highest coverage while M_0 continues to rise. This apparent contradiction might result from our assumption in fitting the data that $L^2 = N$. If the islands are elongated or their boundaries fluctuate significantly, L^2 and N would not be equivalent. Unfortunately the dependence of M_2 on both L and N is too weak to allow an accurate determination of them separately from the data.

The NMR line shapes in the FM phase display broadening and frequency shifts at low temperatures, as shown in Fig. 4. These effects were noted by Godfrin, Ruel, and Osheroff [2] and attributed to the angular distribution ($\pm 15°$) of the Grafoil planes [13] and to the demagnetization effects which should shift the line by $\Delta\nu \sim M_2(1 - 3\cos^2\phi)$ [15], where ϕ is the angle between **H** and the normal to the plane, **n** (Godfrin, Ruel, and Osheroff had $\mathbf{H} \perp \mathbf{n}$). By rotating **H** [16], we found that the line shape and the frequency shift (but not the magnetization) depended strongly on ϕ. At high temperatures our lines were narrow and symmetric as shown in Fig. 4. With $\mathbf{H} \perp \mathbf{n}$, at low temperatures, we observed a positive $\Delta\nu$ as well as broadening. For $\mathbf{H} \| \mathbf{n}$ we found a negative $\Delta\nu$ approximately twice that for $\mathbf{H} \perp \mathbf{n}$ (as expected and previously observed by Bozler, Bates, and Thomson [15]) and correspondingly more broadening.

For both field orientations, we found that the shift was

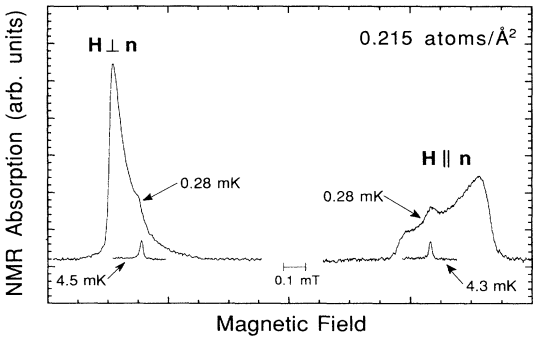

FIG. 4. The NMR line shapes with $\mathbf{H} \perp \mathbf{n}$ and $\mathbf{H} \| \mathbf{n}$ in the FM phase. The signals are measured by sweeping field instead of frequency so the shifts have the opposite sign from $\Delta\nu$ discussed in the text. The relatively unpolarized first layer spins produce a peak near the original resonance position at low temperatures.

approximately proportional to the magnetization at each coverage. Additionally, we saw a small, relatively unshifted peak at low temperatures, corresponding to the first layer spins. This peak is the first direct evidence that the first layer is paramagnetic with $J \sim 0$ and can be treated separately. In further support of the two-phase model, $\Delta\nu(T=0)$ only rise $\sim 15\%$ (attributable to edge effects) between 0.215 and 0.248 atoms/Å^2 while M_2 rises by almost a factor of 2. This indicates that the local magnetization density in the FM phase remains nearly constant with coverage, as expected in a two-phase model.

At the AFM coverages, Godfrin, Ruel, and Osheroff measured the second layer magnetization to show $\Theta_{\text{Weiss}} \sim -5$ mK [1]. Since, in the low-temperature limit, the polarization of AFM spins is small relative to the Curie behavior of the first layer, one would expect that the net magnetization of the film (first and second layer) to approach Curie law behavior for $T < 1$ mK with the same Curie constant as the completed first layer. We do find $M = C/T$ for the AFM coverages at low temperatures, but C is $\sim 15\%$ higher than that for the first layer alone. This difference suggests that $(21 \pm 5)\%$ of the atoms in the second layer display Curie rather than AFM behavior, in quantitative agreement with Elser's [6] model of a registered AFM phase where only $\frac{3}{4}$ of the second layer atoms participate in AFM exchanges. The other 25% are expected to have low exchange frequencies and should exhibit Curie law behavior. It is possible, however, that these additional free spins should be associated with the 20% of the second layer spins discussed above which do not exhibit FM behavior at higher coverages, in which case our data do not support Elser's model.

At no coverage (FM or AFM) did we see anomalous behavior in M_2 at 2.5 mK where Greywall observed a peak in second layer specific heat for $\rho > 0.18$ atoms/Å^2 which had been interpreted as a possible phase transition. Our finding is consistent with theoretical models by Elser [6] and Roger [17], which explain the peak without invoking a phase transition.

This work was supported by NSF Grant No. DMR-9110423, and H.F. was supported in part by the Nishina Memorial Foundation. The authors are grateful for discussions with H. Godfrin, D. S. Greywall, D. S. Fisher, and M. Dykman. We also thank D. S. Greywall for providing the Grafoil used in these experiments.

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