Experimental Observation of a Two-Dimensional Heisenberg Nuclear Ferromagnet

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We have measured the magnetization of 2.5 atomic layers of ^{3}He adsorbed on graphite to 0.7 mK. The magnetization is well described by the exact high-temperature 2D Heisenberg expansion for $T > J = 2.1$ mK, the highest direct nuclear exchange constant ever reported. At lower temperatures, an exponential increase in susceptibility followed by a linear approach to saturation is seen, consistent with present theories for a 2D Heisenberg ferromagnet. The absence of a phase transition is confirmed for temperatures as low as $J/3$.

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We report the first measurements of the magnetization of ^{3}He films adsorbed on exfoliated graphite for temperatures much less than the exchange interactions between the ^{3}He nuclei ($T \ll J$).[1] This is a particularly interesting 2D system because the very low anisotropy of the effective spin interactions and the expected dominance of Ruderman-Kittel-Kasuya-Yosida or three-particle ring exchange processes make it a nearly pure Heisenberg system. A film thickness of 2.5 atomic layers was used following the work of Franco and co-workers[2] so as to maximize the exchange interactions. Those authors interpreted a peak in the magnetization versus coverage at this point as resulting from the completion of solidification of the second layer as the third is added, with a total magnetization at 2.8 mK well above the free-spin value, indicating ferromagnetic interactions. Recent neutron-scattering experiments[3] show that in such a film there are two incommensurate triangular solid layers covered by a half-filled liquid layer. Our magnetization measurements, combined with simultaneous measurements of the NMR frequency shifts due to the demagnetizing effects in the highly polarized medium, suggest 2D ferromagnetic behavior in one of the two solid layers, presumed to be the second. The details of the behavior we observe are consistent with that expected for a 2D Heisenberg ferromagnet,[4,5] and we see no transition above a limiting temperature of 0.7 mK ($=J/3$), as expected for such a system.

Our substrate consisted of sheets of Grafoil[6] bonded to thin copper foils in good thermal contact with a platinum NMR thermometer and a copper nuclear demagnetization stage. Grafoil is an exfoliated graphite with atomically smooth platelets $\simeq 100$ Å wide which are typically oriented within 15° of the sheet surfaces.[3] The total surface area was determined by adsorption isotherm measurements at 4.2 K to be 50.1 m^2 $\pm 3\%$. The sheets were oriented in the plane defined by the static field B_0 and the radio-frequency field h_1, and were contained within an epoxy cell. The epoxy cell and thermometer were coupled to the demagnetization stage by a 1-$\mu\Omega$ conducting link so that their temperature could be varied independently with use of a heater.

A bridge cw lock-in NMR spectrometer[7] was used to measure the ^{3}He absorption lines at 461 kHz ($B_0 = 14.21$ mT) and 209 kHz ($B_0 = 6.44$ mT); and the magnetic field was swept over 0.95 or 1.9 mT depending upon linewidth. This allowed an integration of the NMR line with an accuracy of 3%. The spectrometer was calibrated to provide absolute measurements of the total magnetic moment M with use of a submonolayer ^{3}He coverage (0.844 layer) above 3 mK where one can safely assume Curie-law behavior.[7,8] The absolute calibration is believed good to within 5%. In this calibration at the lowest temperatures a deviation from the Curie law behavior was observed, presumably caused by a residual heat leak ($\simeq 20$ pW) caused by the Grafoil vibrating in the magnetic field and its poor thermal conductivity.[9] The minimum platinum temperature was 0.5 mK while the minimum ^{3}He temperature was 0.7 mK. A temperature-dependent correction, only important at the lowest temperatures, was applied to the data.

The platinum Curie-law susceptibility was measured with a pulsed spectrometer operating at 125 kHz with an uncertainty $< 1\%$ below 10 mK. The Curie constant was determined by the assumption of a Korringa constant $T_1 T = 30$ sec mK above 30 mK.

The ^{3}He coverage was determined by our scaling the coverage at the ferromagnetic peak found in the earlier studies[2] to the surface areas as determined from the adsorption isotherm measurements. The total adsorbed ^{3}He amounted to 43.40 cm^3 STP, for a total density of 0.233 atom/Å^2. From the neutron-scattering results[3] we determine the areal densities for the three layers to be 0.112/Å^2, 0.081/Å^2, and 0.040/Å^2. The density of a complete third layer is 0.070/Å^2.

The results at a field of 14.21 mT are shown in Fig. 1. A large deviation above free-spin behavior is seen throughout the temperature range: There is a clear ferromagnetic tendency with a Weiss temperature of several millikelvins, and the Curie constant is smaller

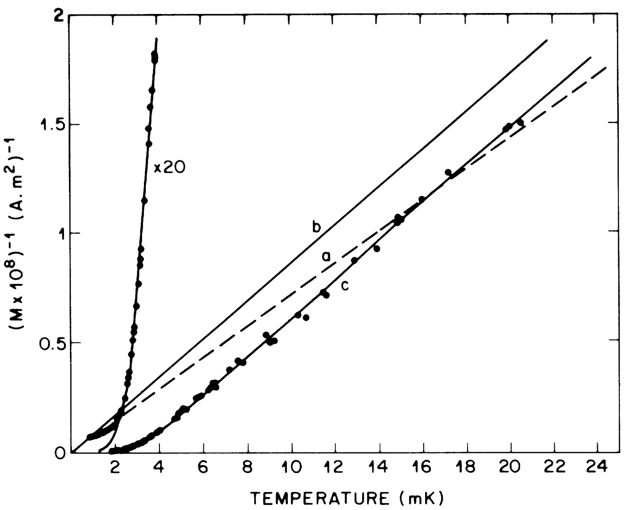

FIG. 1. Inverse magnetic moment vs temperature; *a*, free-spin (Curie) value for all spins; *b*, two-solid-layers Curie law; *c*, calculated with the assumption of a Curie law for the first layer and a 2D Heisenberg ferromagnet ($J=2.1$ mK) for the second layer. Curve marked $\times 20$ is the same as *c* except the vertical scale is expanded twentyfold.

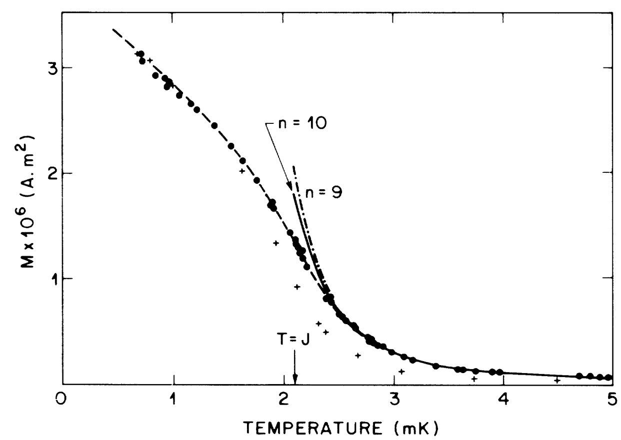

FIG. 2. Total magnetic moment vs temperature at a magnetic field 14.21 mT (dots) and 6.44 mT (crosses). Lines: ten- and nine-term high-temperature series ($B_0=14.21$ mT).

than the free-spin value. The slope $d(M^{-1})/dT$ is well described by that of curve *b*, the contribution (for free spins) of the two solid layers, suggesting that the third fluid layer is degenerate at these low temperatures.

A quantitative analysis of the data can be performed following the conclusion of Refs. 7 and 8 that only the second layer is ferromagnetically active. This conclusion is also supported by frequency-shift data presented below. We presume that it is the second layer which is ferromagnetically active, but for most of our analysis either layer will suffice. The first layer plays the role of a paramagnetic substrate weakly coupled to the active layers: In-plane exchange in the second layer, and exchange between the second and third layer, must be substantially larger than similar processes involving the first layer.

The solid line, *c*, shown in Fig. 1 has been calculated with the assumption of a free-spin susceptibility for the first layer, and a 2D nearest-neighbor ferromagnetic $S=\frac{1}{2}$ Heisenberg Hamiltonian to describe the second-layer magnetism. The high-temperature series expansion for the 2D triangular lattice is known[4] to ten terms as a function of J/T, where the exchange constant J is defined by the expression $H=\frac{1}{2}J\sum_{i<j}\sigma_i\cdot\sigma_j$ ($|\sigma^2|=1$), and the sum is only over nearest neighbors. In adsorbed ^{3}He, three-particle ring exchange processes are believed to be the dominant interactions, and they lead directly to such an effective spin Hamiltonian for spin-$\frac{1}{2}$ particles. If other exchange processes are also important, they will produce a more complex Hamiltonian. Since the number of spins in each layer is known, J is the only adjustable parameter. Our data are well described with $J=2.1$ mK, the largest exchange constant ever found for ^{3}He. The good quality of the fit is not likely to be fortuitous: The choice of J has a large influence at high temperatures (the Weiss temperature of the second layer is $\theta=3J=6.3$ mK), and the temperature dependence at low temperatures is a strong function of J/T as shown by the vertically expanded ($\times 20$) plot in Fig. 1. This value of θ is nearly twice as large as that presented in Ref. 2 for a similar film. In that work θ was estimated by a linear extrapolation of M^{-1} to zero, a procedure which underestimates θ because of the importance of higher-order terms in the series expansion.

Deviations from the high-temperature expansion can be observed (Fig. 2) at temperatures of the order of $J=2.1$ mK, precisely where the validity of the expansion ceases. This is illustrated by the difference between a nine-term and a ten-term series. It should be noted that for $T=J$ the magnetization is 76 times larger than the free-spin value. Also shown in Fig. 2 are the data at the lower field. At high temperatures the magnetic moment M is proportional to the field, and the system can be described by a susceptibility. At the lowest temperatures, the data at the two fields merge and the temperature dependence is weaker. The first-layer contribution $[M_1=(6.65\times 10^{-11}\text{ K A m}^2)/T]$ is relatively small in this temperature range, and M is essentially equal to M_2, the second-layer magnetic moment. The data are shown in Fig. 3 (after subtraction of the first-layer contribution) normalized by the field B_0 and the Curie constant C; this defines a susceptibility equal to $1/T$ at high temperatures. An exponential behavior is clearly observed for $J/T\simeq 1$, in agreement with Heisenberg 2D theoretical calculations[5] which indicate that $\chi=CT^le^{aJ/T}$. The actual values of C, l, and a depend on the model; theoretical curves[5] (recalculated for a triangular lattice when necessary) lie above and below our data (within a factor of 2), with a slope approximately equal to the ex-

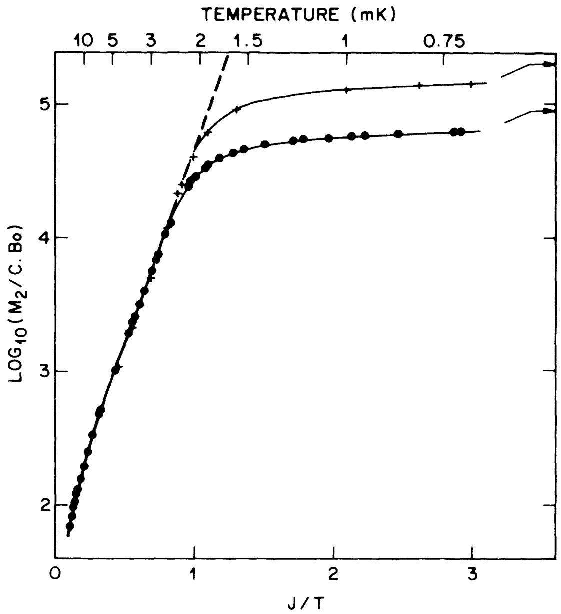

FIG. 3. Second-layer susceptibility vs J/T. Note the exponential behavior for $T \simeq J$. Dots: $B_0 = 14.21$ mT; crosses: $B_0 = 6.44$ mT. Arrows: total polarization values. Thick solid line: high-temperature expansion.

perimental results ($\pm 30\%$). This exponential susceptibility suggests a transition at $T=0$ as expected for a Heisenberg ferromagnet at the lower critical dimension. It is interesting to note in Fig. 3 that the high-temperature behavior ($T > J/K$), which follows the high-temperature expansion, also appears exponential. Our data at the lowest field show that this law can be extended to temperatures $T \lesssim J$. A significant departure from the exponential law is observed at the lowest temperatures. The total magnetic moment becomes independent of the field (see Figs. 2 and 3), suggesting a saturation. The saturation value of M_2 is calculated to be 4.3×10^{-6} A m^2. With use of this value, the maximum nuclear polarization of the second layer, observed at 0.7 mK, was $(69 \pm 5)\%$. The data (Fig. 2) seem to extrapolate to a value lower than the saturation value by $\simeq 20\%$, twice our estimated error bars. This could be due to exchange processes neglected in the present analysis, or to a finite-size–limited correlation length. It is clear that deviations from the exponential susceptibility are due mainly to the large (almost saturation) polarization of the layer in a finite magnetic field.

A simple spin-wave calculation in two dimensions shows that the total magnetic moment should be reduced by

$$\frac{\Delta M}{M_{\text{sat}}} = \frac{2}{\pi\sqrt{3}} \frac{T_0}{J} \left\{ 1 - \left(\frac{T}{T_0} \right) \ln \left[\exp\left(\frac{T_0}{T} \right) - 1 \right] \right\}.$$

Here T_0 is a low-frequency cutoff, and the magnetic field is too low to introduce a significant gap in the spin-wave spectrum. Size effects, on the other hand, give $T_0 = 4\pi^2 J (a/\xi)^2$; a is the interatomic distance $\simeq 3.8$ Å and ξ the finite-size–limited correlation length. The data at the lowest temperatures are consistent with $\xi \simeq 60$ Å ($T_0 = 0.3$ mK), a value comparable to the platelet dimension.[3]

The conclusion that the ordering is occurring in a single layer is supported by simultaneous measurements of large positive frequency shifts ($\Delta\nu \lesssim 3.0$ kHz), similar to those observed in confined ^{3}He by Bozler, Bates, and Thomson.[10] Our maximum shift corresponds[11] to a second-layer polarization $\simeq 65\%$, in good agreement with the direct measurement described above. We have also observed an asymmetric broadening of the line which can be quantitatively described by consideration of the distribution of orientations of the platelets in Grafoil[3] and the angular dependence[10] of the frequency shift.

Long time constants were observed below 2 mK. Using the thermal properties of Grafoil[9] we have estimated the heat capacity of the ^{3}He ferromagnetic layer. A linear dependence at low temperatures, a maximum at $T \simeq J$, and a field-dependent decrease at higher temperatures are found, as expected for a 2D Heisenberg ferromagnet.[4]

We now discuss the origin of the 2D ferromagnetic coupling. A possible ferromagnetic mechanism is in-plane three-particle exchange within the second layer.[12] Recent experiments[7] at submonolayer coverages show that only very small deviations from free-spin behavior occur in the density range of interest. This does not necessarily imply, however, that exchange within the second layer might not be dominant.[12] It has also been suggested, however, that an indirect exchange, involving the fluid layer, may be dominant.[8] A theoretical calculation[13] has demonstrated the ferromagnetic Heisenberg nature of this Ruderman-Kittel-Kasuya-Yosida exchange process. Both mechanisms are consistent with the coverage dependence observed at the ferromagnetic peak in earlier work.[2,8]

Our data, in agreement with what is known of the critical behavior of 2D Heisenberg ferromagnets, do not display any evidence of a phase transition at finite temperatures; furthermore, the exponential susceptibility (in the regime where size effects and large polarizations due to finite fields are not important) suggests a $T=0$ transition. We have not found any evidence of spontaneous magnetization or hysteresis.

Early work by Bozler *et al.*[1] on ^{3}He confined in Grafoil seemed to indicate a phase transition at 0.38 mK. Very recent similar experiments[11] at low ($\simeq$dipolar) fields gave evidence of nuclear ordering, and also of a large anisotropy in the graphite planes. It has been shown[2,8] that the ferromagnetic mechanism discussed here is also responsible for the behavior observed in confined ^{3}He, although strongly attenuated by the compression of the second ^{3}He layer; the results reported here will allow an interpretation of these experiments on a quantitative basis.

More experimental work is certainly needed to understand fully this beautiful 2D system, and eventually its limitations (size effects, anisotropy, competing exchange couplings, etc.). Theories need to be refined to account more quantitatively for the saturation in finite fields, and quantum Monte Carlo calculations may be able to determine directly the magnitudes of possible in-plane and Ruderman-Kittel-Kasuya-Yosida exchange frequencies. A nuclear 2D Heisenberg-ferromagnet experimental model system may now be available for detailed confrontations between theory and experiment.

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