Two-Dimensional Antiferromagnetism of Solid ^{3}He Adsorbed on Plated Graphite

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We report measurements of the nuclear magnetic susceptibility of the monolayer two-dimensional solid formed by ^{3}He adsorbed on graphite plated with two atomic layers of HD. A low-density solid is observed, consistent with a $\sqrt{7}\times\sqrt{7}$ registered structure, which exhibits large antiferromagnetic exchange. A distinct cusp in the susceptibility is observed near 1 mK, suggestive of a magnetic phase transition. The evolution of this and other features with surface density is discussed.

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Solid films of ^{3}He adsorbed on the surface of graphite provide a model system for the study of purely two-dimensional spin $\frac{1}{2}$ systems with isotropic Heisenberg exchange. Helium films on graphite exhibit strong atomic layering effects and measurements of the nuclear magnetic susceptibility [1] and heat capacity [2] on pure ^{3}He films have revealed a number of novel features. In particular a film of somewhat greater than two atomic layers exhibits a strong anomaly in ferromagnetic exchange. At this coverage the first layer forms a compressed triangular lattice, the second layer is a solid of unknown structure, and there is a fluid overlayer. It is now clear that this anomaly is associated with exchange of atoms in the second solid layer; this has been confirmed by experiments in which the first ^{3}He compressed solid layer is replaced by ^{4}He [3]. The ferromagnetic anomaly is a rather sharp function of the second layer density [2,3]. According to [3] the peak occurs when the relative densities of the second and first layers is $\rho_2/\rho_1 \approx \frac{2}{3}$, suggesting a registered structure stabilized by the periodic potential due to the first layer atoms.

However, the main purpose of this Letter is to provide new experimental information on 2D ^{3}He as a model *antiferromagnet*. It has been shown for ^{3}He films on graphite that prior to the formation of a third layer the second layer solidifies [4] into a structure with antiferromagnetic exchange [3,5]. This solid is first completely established at a second layer surface density of 0.064 Å^{-2} for pure ^{3}He films. It has been proposed [6] that this corresponds to a $\sqrt{7}\times\sqrt{7}$ structure in registry with the first layer triangular lattice. The ratio of the surface densities of the first and second layers for this structure is $\frac{4}{7}$, consistent with experiment, and $\frac{3}{4}$ of the spins form a *kagomé* net. This structure was invoked in part to explain the heat capacity, c, data [4], which peak near 2.5 mK with $c \propto T^{-1}$ above the peak and behavior consistent with $c \propto T$ just below the peak [7]. The nuclear magnetic susceptibility of the second solid layer has been studied in measurements on pure ^{3}He films and with the first layer replaced by ^{4}He. At this coverage a Curie-Weiss θ of ≈ -5 mK has been reported for pure ^{3}He films [5], while with ^{4}He preplating a value of -1.7 mK [3] has been found. Formation of a third fluid layer occurs at 0.069 Å^{-2}. With increasing coverage fluid layers develop and the second solid layer is compressed.

In this Letter we report a study of a single atomic layer of ^{3}He adsorbed on graphite preplated with two atomic layers of HD. The preplating serves to adjust the average surface binding potential, but also alters the periodicity of this potential significantly from that of a compressed helium layer. We find that this appears to stabilize a 2D solid of the lowest surface density yet observed and this solid exhibits large antiferromagnetic exchange. A further remarkable feature is a cusp in the susceptibility near 1 mK, suggestive of a magnetic phase transition. As the surface density of the monolayer solid is increased its properties evolve in a rich and complex way. A significant range of surface density can be explored before the intervention of promotion to a subsequent fluid layer.

The graphite substrate used in this work was Grafoil [8]. The sample consisted of a stack of sheets 20 mm×20 mm×125 μm. Two sheets were diffusion bonded on either side of a 25 μm thick copper foil; these were then stacked, separated by Kapton sheets for insulation, and inserted inside the NMR coil. The static field for NMR was applied in the plane of the sheets. The nuclear magnetic susceptibility was measured by field swept continuous wave NMR using a Robinson oscillator operating at 920 kHz. A small frequency modulation at 21 Hz was applied to enhance the signal to noise. The sample was cooled by a copper nuclear demagnetization stage and the temperature measured by pulsed NMR on platinum wires, calibrated by a ^{3}He melting curve thermometer.

The coverage scale was determined using the measured minimum in T_2 as a function of coverage for a submonolayer ^{3}He film as a fiducial point [9]. This is a sharp feature and a precise signature of promotion to the second layer. The density at this point is taken to be 0.106 Å^{-2} from neutron scattering measurements [10]. The HD, of purity 98.6%, was added at 15 K to a total surface coverage of 0.183 Å^{-2}. This coverage was estimated from published neutron scattering measurements [11] of the surface density of the first and second adsorbed layers of H_2 and D_2 to be the coverage required for two completed atomic layers. The HD was annealed at 15 K and slowly cooled to 4.2 K. The ^{3}He samples

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were added below 1 K and annealed for several hours at a vapor pressure of 1 mbar. We note that HD is preferred over the pure isotopes of hydrogen in order to avoid significant heating from ortho-para conversion [12].

The first ^{3}He coverage investigated was 0.04 Å^{-2} and here the susceptibility was consistent with that of a 2D Fermi fluid with a small proportion of the spins, corresponding to 0.003 Å^{-2}, giving a contribution to the susceptibility close to Curie law. We here attribute the Curie contribution to ^{3}He atoms localized in the first two atomic layers of HD as a result of the chosen preplating coverage being slightly too small. Earlier experiments [13] on the 2D ^{3}He fluid adsorbed on graphite plated with a single atomic layer of ^{4}He also showed such an effect; in that case it was a simple matter to increase the ^{4}He coverage until the localized spins were promoted. As far as the inferred fluid contribution to the susceptibility is concerned, both systems, with ^{4}He and HD preplating, have similar behavior. Thus at a coverage 0.04 Å^{-2} the measured enhancement of the fluid susceptibility over the ideal gas value was the same for both systems, suggesting that the quasiparticle interactions in the 2D Fermi system are similar. In the following we subtract 0.003 Å^{-2} from the ^{3}He coverage to infer the surface density in the ^{3}He monolayer.

Over the corrected coverage range from approximately 0.047 to 0.052 Å^{-2} the film goes through what for a classical system might be called a coexistence region. At the surface density 0.052 Å^{-2} the film forms a 2D quantum solid with remarkable properties. Over the entire temperature range 200 to 1 mK the nuclear susceptibility χ closely follows a Curie-Weiss law $\chi \sim (T-\theta)^{-1}$ with a Curie-Weiss constant $\theta = -11.2$ mK. This is the largest θ yet observed for ^{3}He in both 2D and 3D (e.g., at the ferromagnetic anomaly $\theta \approx 6.3$ mK). The extremely low density of this solid undoubtedly contributes to the magnitude of θ. By contrast in our earlier experiments with a single atomic layer of ^{4}He preplating we determined $\theta = -1.7$ mK at a solid surface density of 0.066 Å^{-2} when the antiferromagnet is first completely established.

In fact this is the lowest density 2D quantum solid yet identified and its formation confirms the importance of superlattice structures in registry with the periodic potential of the underlayer for such solid helium films. This underlayer, of HD or ^{3}He or ^{4}He in the various experiments, forms a triangular lattice and hence a honeycomb of adsorption sites. The density of these sites is significantly smaller for HD (for which the triangular lattice has an estimated $\rho = 0.091$ Å^{-2}) than for ^{4}He ($\rho = 0.114$ Å^{-2}). In the present case of HD the solid first forms at ρ_3/ρ_{HD} near $\frac{4}{7}$. This is the same relative value of ^{3}He density to underlayer density found for the second layer of ^{3}He on the first layer of ^{3}He [4], ^{3}He on a preplating solid layer of ^{4}He [3] and most recently the second layer of ^{4}He on the first layer of ^{4}He [14]. This suggests that in all these cases the second helium layer first solidifies into the same structure, possibly the $\sqrt{7}\times\sqrt{7}$ structure

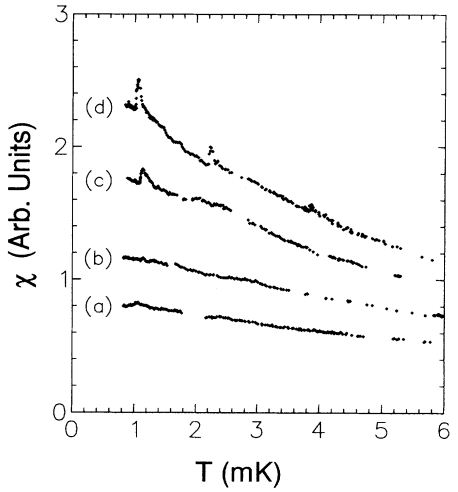

FIG. 1. Temperature dependence of nuclear magnetic susceptibility in 2D solid ^{3}He. Surface densities (a) 0.052, (b) 0.0545, (c) 0.057, (d) 0.062 Å^{-2}.

proposed by Elser [6].

But perhaps the most interesting feature observed in the present measurements at this coverage is the cusplike maximum in susceptibility near 1 mK. This is strongly suggestive of a magnetic phase transition. This occurs at $T \ll \theta$, indicative of a strongly frustrated system such as the *kagomé* net. It is of interest to compare this behavior with that of $SrCr_8Ga_4O_{19}$, which has been discussed as a quasi-2D magnet and for which the $S = \frac{3}{2}$ Cr^{3+} ions form a *kagomé* lattice. In that case [15] the Curie-Weiss law is followed to $T/\theta \sim 0.3$ ($\theta = 515$ K), i.e., to tempera-

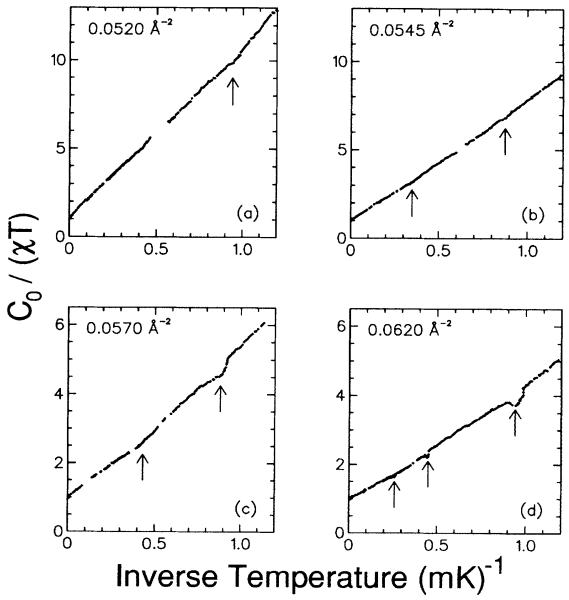

FIG. 2. $(\chi T)^{-1}$ normalized by the Curie constant at each coverage. Slope changes and susceptibility peaks are highlighted by arrows.

tures much less than θ, similar to our observations. However, the ordering transition found at 3.5 K is attributed to 3D interactions. This provides a contrast with the monolayer ^{3}He film which is a perfect 2D magnet.

With increasing coverage the behavior of the film evolves in a very interesting way. For all the data discussed there is no signature of promotion to a subsequent fluid layer. Therefore we are dealing with a monolayer 2D solid whose structure evolves in an unknown way, with average surface density in the range 0.052 to 0.062 Å^{-2}. The data are plotted in two ways in Figs. 1 and 2 to emphasize different systematic features. Data above 10 mK were taken while regulating the temperature using the melting curve thermometer; the data shown at lower temperatures are from a single continuous warmup after demagnetization. The main gross effect is that with increasing density the Curie-Weiss constant decreases. Values of θ inferred from a linear fit of $(\chi T)^{-1}$ as a function of T^{-1} for data $T > 10$ mK are shown in Fig. 3. For reference we also show earlier data for ^{3}He on ^{4}He plated graphite [3]; the two sets of data appear to join reasonably smoothly [16].

Figure 4 shows in more detail the evolution of the peak near 1 mK with increasing density. At first the peak structure becomes rather indistinct; thus at 0.0545 Å^{-2} there is an indication of what may be two rather weak anomalies, just distinguishable above the scatter in the data. However, on increasing the coverage to 0.057 Å^{-2} a pronounced peak reappears with a somewhat different shape from the cusplike feature seen at 0.052 Å^{-2}. The "1 mK peak" is now asymmetric, with almost a discontinuity in susceptibility on the low temperature side. There is also perhaps a broad feature between 2 and 3 mK. At 0.062 Å^{-2} the 1 mK peak is stronger and narrower, here too the susceptibility decreases sharply on the low temperature side. The remarkable observation at this density, seen in Fig. 1, is that there are three quite sharp and distinct peaks in susceptibility at 1.0, 2.2, and 3.9 mK [17].

We note that this last surface density is near to $\frac{2}{3}$ of that of the second layer of the HD preplating and is a natural candidate for a registered structure. The simplest of these is the $\sqrt{3}\times\sqrt{3}R30°$ honeycomb structure for which $\frac{1}{3}$ of the honeycomb of adsorption sites are occupied. Our earlier measurements [3] suggest that in the case of ^{3}He on graphite plated with ^{4}He the ferromagnetic anomaly occurs at or very close to this site occupancy; alternative structures are discussed elsewhere [18]. The main suggestion we wish to make here is that the $\frac{4}{7}$ and $\frac{2}{3}$ structures appear to be in some sense generic for adsorbed helium systems explored to date.

In addition to the peaks discussed so far the data show further features revealed when we plot in Fig. 2 normalized $(\chi T)^{-1}$ vs T^{-1}. This plot shows up distinct discontinuities in slope, in some cases coincident with the susceptibility maximum. We may summarize the results as follows. At 0.052 Å^{-2}, a clear change in slope is seen at 1 mK, at the same temperature as the susceptibility maximum. At 0.0545 Å^{-2} there is a slope change near 3 mK but none at the very weak feature just above 1 mK. At 0.057 Å^{-2} there is a slope change near 2.5 mK but none at the 1 mK peak. And finally at 0.062 Å^{-2} there are two slope changes both at the 1 mK peak and at the 3.9 mK peak [19]. As soon as the film exhibits a ferromagnetic tendency the susceptibility becomes a smooth function of temperature, and no anomalies are observed. This occurs at a corrected coverage of 0.077 Å^{-2} following promotion to a subsequent fluid layer. This confirms the anomalies as feature of the 2D antiferromagnet. The crossover to ferromagnetic exchange will be reported in a separate publication. We observe no large increases in linewidth and shifts in resonance frequency such as observed in pure ^{3}He films at the ferromagnetic peak [20].

These experimental results, in particular the anomalous peaks in susceptibility, strongly suggest the occurrence of magnetic phase transitions in the 2D ^{3}He antiferromagnet at finite temperature. We propose that it is important

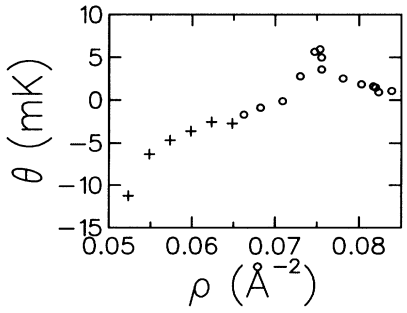

FIG. 3. Curie-Weiss θ as a function of solid density; (+) this work, (○) data from Ref. [3].

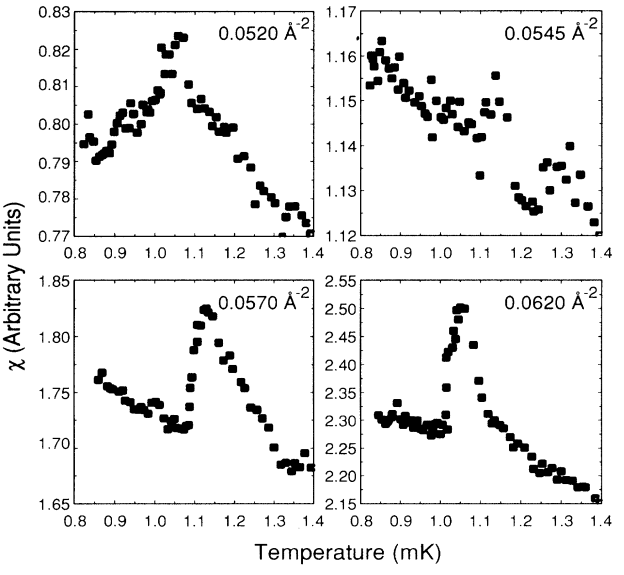

FIG. 4. Expanded view of susceptibility anomalies near 1 mK.

for this system to distinguish between coverages for which the solid has a uniform structure and otherwise. As we have seen the coverage at which the film first solidifies is consistent with a $\sqrt{7}\times\sqrt{7}$ structure; here $\frac{3}{4}$ of the atoms form a *kagomé* net. The *kagomé* lattice is a highly frustrated system; here phase transitions from paramagnetic to chiral spin liquid followed by spin liquid to spin Peierls have been suggested [21]. Finite N calculations [22,23] suggest that spin Peierls order could be rather weak. Other forms of order such as spin nematic are also possible. In this case the role of the $\frac{1}{4}$ spins not part of the *kagomé* lattice but which are highly delocalized is not clear. The experimental result at 0.052 $Å^{-2}$, where the susceptibility at 1 mK is a factor of 10 smaller than the Curie value, tells us that treating these spins as free, as done by Elser [6], is an oversimplification. Explicit calculation of the experimental signatures of these various phase transitions would be very helpful.

In the case of coverages where there is both structural disorder and frustrated spin exchange it is possible that a 2D spin glass transition will occur. Here the observation of sharp peaks in the susceptibility, characteristic of spin glass transitions, is suggestive. This hypothesis may be further investigated by measurements as a function of magnetic field. At 0.062 $Å^{-2}$, the three peaks may characterize a registered superlattice structure or a domain wall solid in which the surface density is regularly modulated.

In conclusion we have demonstrated that the use of graphite preplated with two atomic layers of HD enables the study of more extreme quantum solids than achievable hitherto. This follows from the possibility in 2D of controlling the surface structure through the formation of registered solid layers. Here direct structural determinations would be very useful. This system is of intrinsic interest as a model antiferromagnet and provides to our knowledge the first experimental evidence for a magnetic phase transition in a genuine 2D isotropic Heisenberg antiferromagnet.

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