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Ferromagnetic instability of ^{3}He layers adsorbed on Grafoil

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The magnetization of 0.9, 1.74, 2.57, 3.57, and 7.04 layers of ^{3}He adsorbed on Grafoil has been measured down to 3 mK. A large ferromagnetic tendency was observed at 2.57 layers, with a corresponding Curie temperature of 1.8 mK. The effect is interpreted as a ferromagnetic instability of liquid ^{3}He when its thickness is of the order of the Fermi wavelength.

The theoretical prediction of a ferromagnetic instability in confined liquid ^{3}He (Ref. 1) and the experimental observation[2] of an excess susceptibility in liquid ^{3}He confined in Mylar foils and carbon black has stimulated intensive research in this field.

The total susceptibility has been found to be described by the expression $\chi = A/(T-\theta) + B$, where B is the constant susceptibility of the bulk Fermi liquid, $\theta \sim 0.5$ mK and is positive, and the magnitude of A would correspond to ~ 5 "Curie-Weiss" ^{3}He layers located on the surface of the Mylar or carbon-black substrate.

Similar studies performed in our laboratory[3] and at the University of Southern California[4] on Grafoil showed a Curie behavior which could reasonably be attributed to a single paramagnetic solid layer. It was deduced that the second layer of ^{3}He on Grafoil is liquid and this was verified by neutron diffraction experiments.[5] This simple model with one layer of solid and bulk liquid is intuitively reasonable except that the origins of the nonzero θ and the discrepancy between the two sets of experiments remain unexplained.

One difference between the experiments described is the nature of the substrate. Grafoil is homogeneous from the point of view of physical adsorption;[6] it can be easily cleaned; the structure of the adsorbed layers is known and is well defined.[5,7,8] Mylar and carbon black, on the other hand, are extremely heterogeneous and degassing *in situ* is very difficult.

The particle size is also an important variable: 400-Å alumina powder (heterogeneous) gave the same results as Grafoil[3]—one Curie-Weiss layer. The size of the cavities in carbon black and Mylar is much smaller; the large "excess susceptibility" could be either attributed to a larger number of solid "layers" (which are not well defined on heterogeneous substrates) because of capillary condensation or to a boundary magnetism effect.

In order to clarify the experimental situation, we have performed susceptibility measurements at millikelvin temperatures on ^{3}He adsorbed on Grafoil as a function of temperature and coverage. This preserves the advantages of a well characterized substrate (Grafoil) and moreover allows us to vary systematically the thickness of the liquid layers.

A plastic (araldite) cell of volume 21.9 cm^3 was located inside the mixing chamber. In its center it contained an NMR coil with 38 Grafoil sheets (mass $=3.26$ g) sintered on 19 copper foils (thickness 25 μm), cut to fill the coil form (diameter 18 mm, length 20 mm), and degassed at 800 °C for 4 h. Kapton foils were used for electrical insulation between the Grafoil sheets. The copper foils had ribbon-shaped tails which were screwed (out of the high rf field region) to a copper post. This post passed through a Stycast seal in the cell wall into the mixing chamber where a sintered silver disk ensured thermal contact. A carbon resistance thermometer (Speer 100 Ω) and a heater were also attached to the copper post. The mixing-chamber temperature was given by carbon resistors and by a CMN mutual inductance thermometer calibrated at zero field and at 273 G. The magnetic field was provided by a superconducting coil wound on the thermal shield at 0.7 K.

cw NMR measurements were performed at 886 kHz (273 G) with a low-level Q meter, while the magnetic field was swept. The magnetization was obtained by integration of the absorption signal by computer. The signal amplitude was normalized to a calibrated absorption signal to eliminate the problem of drift in amplifier gain.

The cell, which was connected to the gas-handling system by Cu-Ni tubes, was evacuated, flushed with ^{3}He and then pumped for several days with a nitrogen-trapped diffusion pump. Adsorption isotherms at 4.2 K were performed with a standard gas handling system including calibrated volumes, a 100-torr Baratron MKS 77 gauge, a charcoal dipstick, and a diffusion pump.

The amount of gas corresponding to monolayer coverage was determined to be 22.0 cm^3 at standard temperature and pressure (STP), the amount adsorbed at 4.2 K at 1 torr pressure. From the densities of the different layers determined by neutron scattering[9] and specific heat measurements,[7] the gas volume corresponding to completion of the second layer would be 14.7 cm^3, and for the third and successive layers, 12.7 cm^3. Accordingly, our 19.8-, 33.0-, 43.9-, 56.6-, and 100.8-cm^3 STP samples corresponded to coverages of 0.90, 1.74, 2.57, 3.57, and 7.04 layers, respectively.

The ^{3}He gas was always introduced at 4.2 K, since we observed that when the gas was introduced at low temperatures it was not adsorbed, but condensed as liquid droplets at the bottom of the cell. An anneal at 10 K was performed for the lowest coverages. The system was cooled down to 1.5 K in several hours, and to 3 mK in ~ 10 h. At low temperatures, all the gas was condensed on the Grafoil, and the vapor pressure was negligible.

The total ^{3}He magnetization measured as a function of temperature for these samples is shown in Fig. 1. This is plotted as the product of magnetization and temperature to emphasize deviations from a Curie law. With 0.90 ^{3}He layers the behavior was essentially paramagnetic as expected for a solid layer at this density.[7] The results at this coverage show that the ^{3}He has been effectively cooled down to millikelvin temperatures. With 1.74 layers the Curie law is still

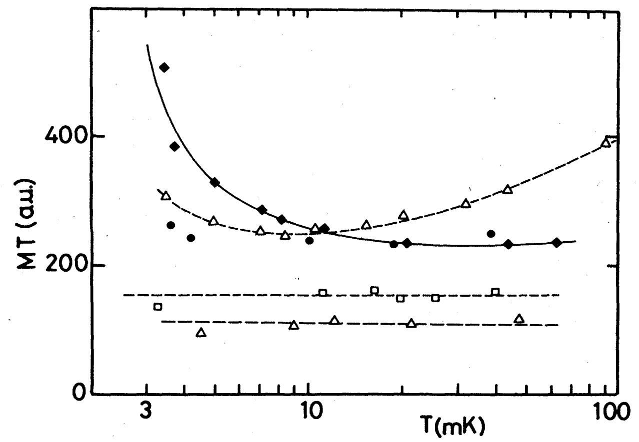

FIG. 1. The product of the magnetization and the temperature (arbitrary units) as a function of temperature for different coverages. ▲ 0.90 layers; ■ 1.74; ◆ 2.57; ● 3.57; Δ 7.04 layers.

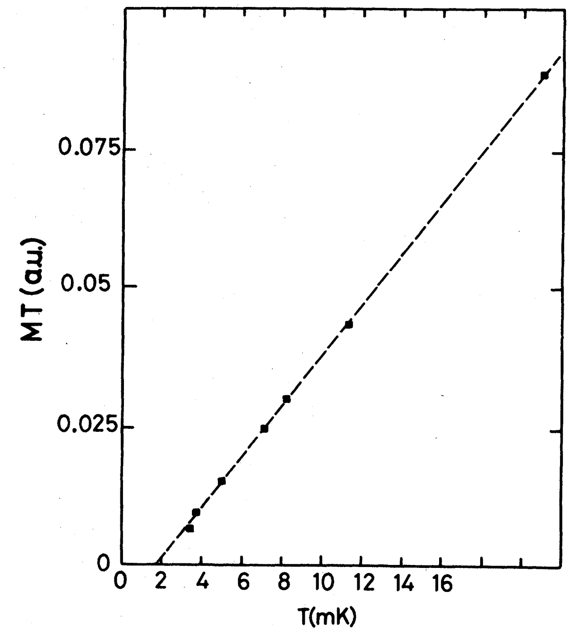

FIG. 2. The inverse of the total magnetization is plotted as a function of temperature for a coverage of 43.9 cm^3 (2.57 layers). The Curie-Weiss temperature is 1.8 mK.

followed and the magnetization has increased proportionally to the number of adsorbed atoms. This is a new effect, since the incomplete second layer is known to be liquid,[7] but nevertheless, no Fermi degeneracy is observed.

With 2.57 layers, the magnetization at high temperatures ($T \gtrsim 10$mK) has again increased linearly with the adsorbed amount, still following a Curie law. At low temperatures a new effect is observed. The product MT diverges with a Weiss temperature $\theta = 1.8$ mK, as deduced from the plot of M^{-1} vs T (Fig. 2). This behavior was modified by the addition of one further layer (3.57 layers). In this case, the Curie contribution remained unchanged, but the Weiss temperature was reduced to $\theta \sim 0.5$ mK.

For the same coverage a small increase of MT was also observed at the highest temperatures. This effect was better seen for our highest coverage (7.04 layers) where a linear increase of MT above 10 mK indicated the existence of a Fermi liquid with a constant magnetization. However, it appears that the degeneracy temperature was lower than in bulk liquid ^{3}He. The ferromagnetic tendency at low temperatures gave a small θ (~ 0.5 mK), as observed with Grafoil in bulk liquid.[3,4]

This experiment clearly shows that two-dimensional liquid ^{3}He has a much larger magnetization than the bulk, since the Curie behavior persists up to ~ 2.6 layers, even though there is only one solid layer. It has been suggested that the second layer might solidify.[7] The disagreement with the susceptibility measurements[3,4] could, in principle, be understood if the "bulk" liquid susceptibility in the Grafoil voids is also enhanced by a factor ~ 2, modifying the normalization procedure that was used. However, the neutron experiment[5] did not detect the existence of a solid second layer. Even with the hypothesis of two solid layers at high coverages, the Curie susceptibility for 1.74 layers and 2.57 layers indicates that thin liquid ^{3}He films have a Curie susceptibility.

We interpret the ferromagnetic instability observed at 2.57 layers to be an effect in the liquid. This effect appears to be very sensitive to the thickness of the liquid which is of the order of the Fermi wavelength $\lambda = h/p_F$. Furthermore, such behavior and a Curie-Weiss law may be inferred from the theory.[1]

If the ferromagnetic instability were to be ascribed to the adsorbed solid, it would be difficult to understand why 2.57 layers are necessary to observe this effect, and why it would be reduced at larger coverages. Suppose two layers of ^{3}He solidify, as was discussed earlier, then there is an alternative explanation of our data, in terms of solid ferromagnetism. Mullin and Landesman[10] have proposed a reduction of the interlayer exchange as a function of the liquid-layer kinetic energy; this provides a coverage dependence of the ferromagnetic indirect exchange-interaction theory.[11]

From our experiments, it appears that thin ^{3}He liquid layers are paramagnetic, that there is a ferromagnetic instability between two and three layers. For larger coverages, the Fermi temperature is reduced compared to that of the bulk.

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