Growth of a Third Ferromagnetic Solid ^{3}He Layer on Graphite

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We have measured the nuclear susceptibility of ^{3}He in Grafoil filled with pure liquid ^{3}He over the pressure region between 0.6 and 31.38 bars and at temperatures down to 0.5 mK with a cw NMR method. The nuclear magnetization corresponding to the adsorbed ^{3}He layers on the Grafoil surface shows a strong ferromagnetic tendency with a periodic behavior as a function of liquid pressure. This observation is attributable to the growth of third and fourth solid ^{3}He layers with the liquid pressure increase. The pressure dependence of the Weiss temperature indicates the third layer is completed at 19 bars and the fourth probably at 28 bars. The number of localized spins estimated from the solid magnetization is almost doubled from 0 to 28 bars, being consistent with this scenario.

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Adsorbed ^{3}He ($S = 1/2$) on a well defined surface is a fruitful system to study two-dimensional magnetism [1]. In particular, ^{3}He film adsorbed on graphite has extensively been studied owing to the atomically flat surface and large surface area of graphite. According to recent studies on the film, a layer by layer growth is confirmed up to seven layers at least, and the first and the second atomic layers solidify on the graphite surface [2], while the upper layers above the second one exist as a liquid. The motion of ^{3}He atoms in the solid layer is considered to be restricted to a two-dimensional plane, since the binding energy is evaluated to be about 135 K and 65 K for the first and the second layer [3], respectively, much larger compared to the temperature for measurements. Therefore, the magnetism for the first or the second layer is understood as a consequence of an ideal two-dimensional magnet with multiple spin exchange interactions. The exchange of even number of particles is antiferromagnetic, while that of an odd number is ferromagnetic. The competition between them causes an evolution from antiferromagnetism to ferromagnetism as a function of the areal density. In addition to the intra layer interactions, there is a long-standing controversy on a possible indirect interaction mediated by ^{3}He quasiparticle in the over-layer liquid. Theoretically various models are proposed based on the second order perturbation theory, for example, a quasi-localized-Fermion theory, etc, [4–8]. Most of the models predict the existence of RKKY type indirect exchange interaction. Experimentally, the recent experiments on the adsorbed film [9] suggest a rather small contribution of this type of interaction. However on the boundary surface dipped in the bulk liquid ^{3}He [10–12], there is no clear evidence so far. In this Letter, we present the first systematic study with a cw NMR technique for the solid ^{3}He layer on graphite covered with bulk liquid ^{3}He as a function of liquid pressure.

The experimental setup is described in our previous work [13]. The substrate used is an exfoliated graphite (Grafoil GTY grade $d = 76$ μm) [14], which consists of partially aligned micro crystals of graphite. It was degassed at 950 °C in vacuum for four hours. 60 Grafoil sheets (8 mm × 8 mm) are stacked in a sample chamber made of epoxy (Stycast 1266). They are dipped into pure liquid ^{3}He which is cooled through a sintered powder heat exchanger, the mixture of Ag and Pt powder with a surface area of 24 m^2. It has a good thermal contact with the copper nuclear stage. The temperature was measured with a ^{3}He melting curve thermometer and a Pt NMR thermometer both of which are mounted on the nuclear stage. NMR measurements were made with a continuous wave method at a frequency of 698 kHz. A static field of 21.5 mT was applied in parallel to the Grafoil sheets and swept to cover the whole NMR line. A saddle type rf coil wound around the sample chamber (145 μH) produces an rf field parallel to the Grafoil sheet. The rf field was small enough to avoid the saturation of ^{3}He spin system. Magnetization (M) was obtained from a numerical integration of the whole absorption line, which exhibits a positive frequency shift and a line broadening arising from local field due to the large sample polarization [15]. The uncertainty of M is the order of 5% at 5 mK, which is mainly due to the uncertainty of subtraction of the base line.

The Grafoil substrate we used has a density of 1.1 g/cm^3, nearly half of the bulk graphite density (2.1 g/cm^3), indicating the existence of a large amount of inner space within the Grafoil sheet itself. The total volume (V_i) of the inner space in our cell is estimated to be 0.14 cm^3, about 48% of the volume for 60 Grafoil sheets with surface area (S) of 6.74 m^2. Therefore an average inner spacing between the graphite surfaces ($2V_i/S$) is calculated to be about 40 nm . In such a narrow space, liquid ^{3}He is not expected to make a superfluid transition. Additionally, a loose packing of the Grafoil sheets in the sample chamber produces interspaces between the sheets. An average spacing of such interspaces is estimated to be 56 μm, judging from the total length of the packed sheets, 8 mm. Since the spacing is much larger than a superfluid coherence length, liquid ^{3}He there

should become superfluid, whose texture is well controlled by the Grafoil wall.

Figure 1 shows the temperature dependence of ^{3}He magnetization at 3.39, 12.30, and 27.62 bars. The inset gives the expanded view in the high temperature region where the liquid contribution increases with increasing the liquid pressure. The solid, dotted, and dashed curves in the inset are the liquid contribution estimated based on the expression for a three dimensional Fermi gas [16] with the known Fermi temperature (T_F^{**}) [17]. The calculated pressure dependence well reproduces the observed behavior above 70 mK. Moreover, a part of liquid signal shows a frequency shift due to a superfluid transition as shown for 27.62 bars in the previous report [13]. The integrated signal magnitude is about 65% and 51% for A and B phase, respectively, compared with the total liquid contribution obtained from the fitting in the normal state. The liquid magnetization arises from three regions, (a) the innerspaces within the Grafoil (b) the interspaces between the Grafoils (c) the other spaces below and above the Grafoil. The A phase frequency shift comes from (b) and (c), while the B phase shift only from (b). Hence the quantity of (b) compared with (a) + (b) is estimated to be about 59%. It is in good agreement with the value, 60%, estimated from the geometrical dimensions described above. These results confirm the above estimation of liquid magnetization is reasonable.

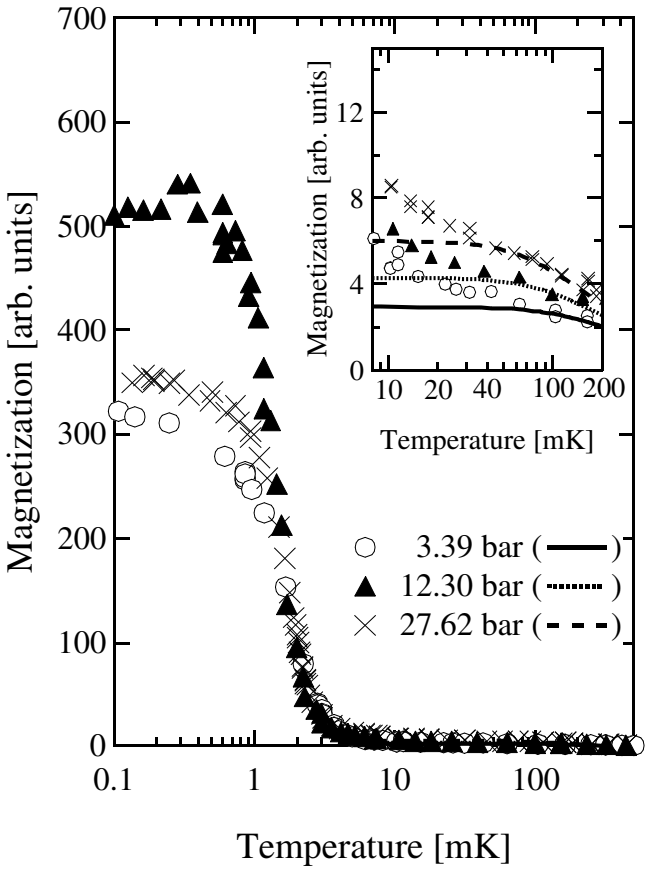

FIG. 1. Temperature dependence of total magnetization under liquid pressure of (∘) 3.39, (▲) 12.30 and (×) 27.62 bars. The curves are the estimated liquid magnetization (see the text.)

Subtracting the estimated liquid contribution, the magnetization for the localized spins is obtained for various pressures as shown in Fig. 2. Here the change of the liquid magnetization due to a superfluid transition is negligible because of much smaller liquid magnetization itself below the superfluid temperatures than the solid magnetization. The solid magnetization shows a ferromagnetic tendency over the whole pressures range. The magnetization at 0.62 bars shows the weakest temperature dependence. Below 7.7 bars, the data exhibit a rapidly rising magnetization around 3 mK and the temperature corresponding to the rapid rise shifts to the lower temperature with increasing the pressure. This shift continues to exist up to 19.35 bars, where the temperature dependence is weak and close to that of 0.62 bars again. The evolution above 19.35 to 27.62 bars resembles that in the lower pressure region. The pressure dependence above 27.62 bars is much weaker than in the low pressure regions.

What causes such a periodic behavior of the magnetization? It is difficult to explain by RKKY interaction through ^{3}He quasiparticles, because the Fermi momentum is a monotonically and weakly increasing function of the pressure. The first layer solid is known to be paramagnetic and the ferromagnetic tendency of the second layer in liquid ^{3}He is too small to reproduce the observed behavior [1]. The other possibility is solidification of the third and the fourth layers with increasing the liquid pressure, analogous to the liquid pressure dependent epitaxial adsorption of solid ^{4}He up to seven layers [18]. The adsorbed atoms feel the pressure from the graphite substrate given as follows,

$$p(r) = p_L + \frac{n_s}{h}\left(\frac{C_3}{r^3} + \frac{C_4}{r^4}\right). \quad (1)$$

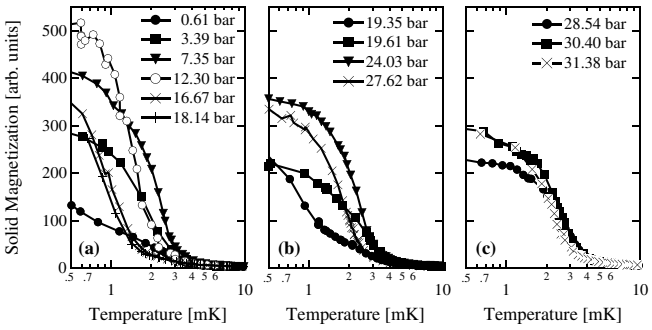

FIG. 2. Temperature dependence of the solid magnetization at (a) low pressures, 0.64 ~ 18.14 bar, (b) middle pressures, 19.35 ~ 27.62 bar and (c) high pressures, 28.54 ~ 31.38 bar. The solid lines are guides to the eye.

Here r is the distance from the graphite surface and p_L is the pressure of bulk liquid. The second term arises from van der Waals potential from the graphite surface where $C_3(= 2.53 \times 10^{-23}\ \mathrm{J\,nm^3})$ and $C_4(= 1.42 \times 10^{-22}\ \mathrm{J\,nm^4})$ are the coefficients of van der Waals potential determined experimentally [19]. h is a monolayer thickness and n_s is an areal density which is assumed constant here. According to Franchetti model [20], the liquid layer solidifies in the region $p(r) > p_s$ where p_s is the melting pressure of bulk liquid ($\sim$34 bar). If we assume the monolayer thickness is 0.3 nm and n_s is equal to a maximum value for the second layer ($=$ 8.7 atoms/nm^2), the solidification at the third and the fourth layer position is expected to be completed at 19 and 28 bars, respectively. Thus the present scenario seems to reproduce the observed behavior, although it is believed from the film experiments that the only two layers exist as a solid. Above 28 bars, the pressure dependence is weak, and it is not clear whether the fifth or more solid layers exist or not.

Based on the above described model, the data are analyzed at high temperatures where the Curie-Weiss approximation is valid . In the low pressure region below 19.35 bars, a three solid layers model is employed:

$$M_{\mathrm{solid}}(T) = C \times \left(\frac{n_{1\mathrm{st}}}{T - \theta_1} + \frac{n_{2\mathrm{nd}}}{T - \theta_2} + \frac{n_{3\mathrm{rd}}}{T - \theta_3} \right), \qquad (2)$$

where C is a Curie constant for each spin, $n_{1\mathrm{st}}$, $n_{2\mathrm{nd}}$, $n_{3\mathrm{rd}}$, and θ_1, θ_2, θ_3 are the amount of ^{3}He atoms and the Weiss temperature for the first, the second and the third solid layer, respectively. According to the film experiment, the first layer is highly compressed [1], being consistent with the calculated pressure from Eq. (1) ($p(r) > 750$ bar) at the first layer position. Therefore, we assume $n_{1\mathrm{st}} =$ 11.4 atoms/nm^2, a maximum density observed in the film experiment, and $\theta_1 = 0$ mK, independent of the liquid pressure. In the same way, the parameters for the second layer at saturated-vapor-pressure conditions are estimated from those of the previous film experiments to be $n_{2\mathrm{nd}} = 8.7$ atoms/nm^2 and $\theta_2 \sim 1.5$ mK [1]. The liquid pressure dependence of these values is unknown, but seems to be negligible. Because the first and the second layer feel strong pressures much higher than 34 bars from the graphite substrate and therefore their densities should not be affected seriously by the liquid pressure. Consequently, $n_{3\mathrm{rd}}$ and θ_3 are two free parameters which should be determined by fitting the data between 0 and 19.35 bars to Eq. (2). Some of the fitted curves are shown in Fig. 3. The magnetization is pretty well fitted to Eq. (2) down to the temperatures close to θ_3 [21]. Pressure dependence of $n_{3\mathrm{rd}}$ is shown in Fig. 4 as a total solid contribution $n_{\mathrm{total}} = n_{1\mathrm{st}} + n_{2\mathrm{nd}} + n_{3\mathrm{rd}}$ and the corresponding θ_3 is shown in Fig. 5.

A relatively rapid increase of n_{total} and a weak dependence of θ_3 with a large value between 0 and 10 bars should be attributable to the growth of the low density third layer, while a weak dependence of n_{total} and a rapid drop of θ_3 between 10 and 18.35 bars is possibly due to the compression of the third layer. This behavior of n_{total} is qualitatively consistent with the pressure dependence of magnetization at 0.5 mK below 12.30 bars, although it is difficult to estimate an exact saturation magnetization

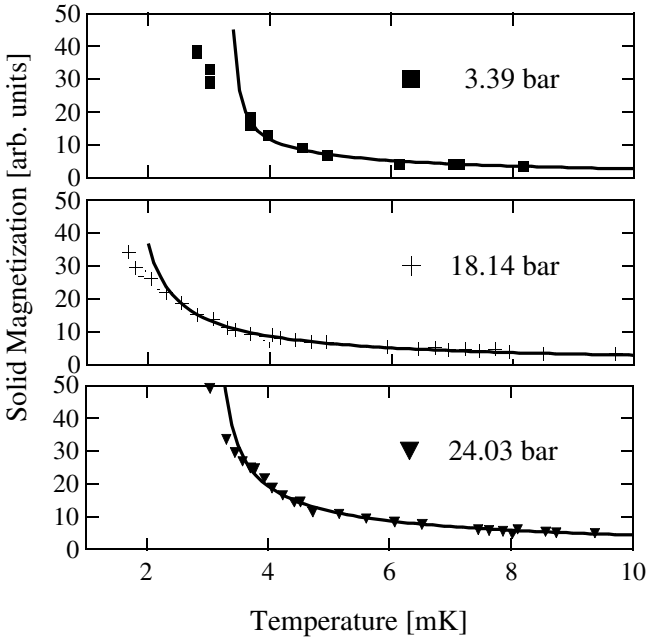

FIG. 3. Solid magnetization for 3.39, 18.14, and 24.03 bars. The solid lines are those fitted to Eq. (2).

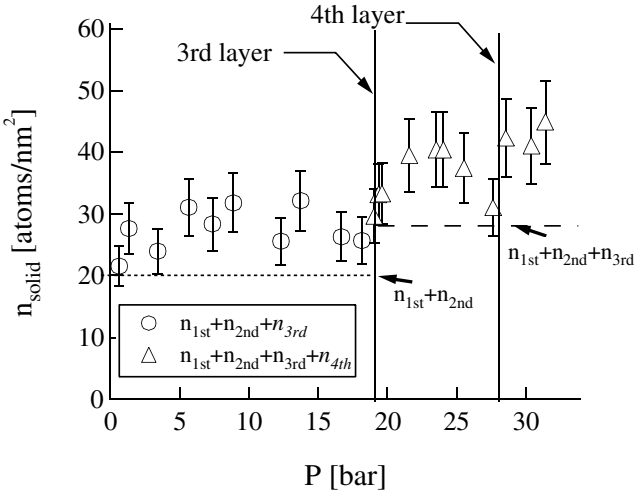

FIG. 4. The total amount of solid ^{3}He layer, ($\circ$) $n_{\mathrm{total}} = n_{1\mathrm{st}} + n_{2\mathrm{nd}} + n_{3\mathrm{rd}}$ and ($\triangle$) $n_{\mathrm{total}} = n_{1\mathrm{st}} + n_{2\mathrm{nd}} + n_{3\mathrm{rd}} + n_{4\mathrm{th}}$. Vertical lines correspond to the third and fourth layer completion. The horizontal dotted and dashed lines indicate the amount of solid ^{3}He corresponding to the second and third layer completion.

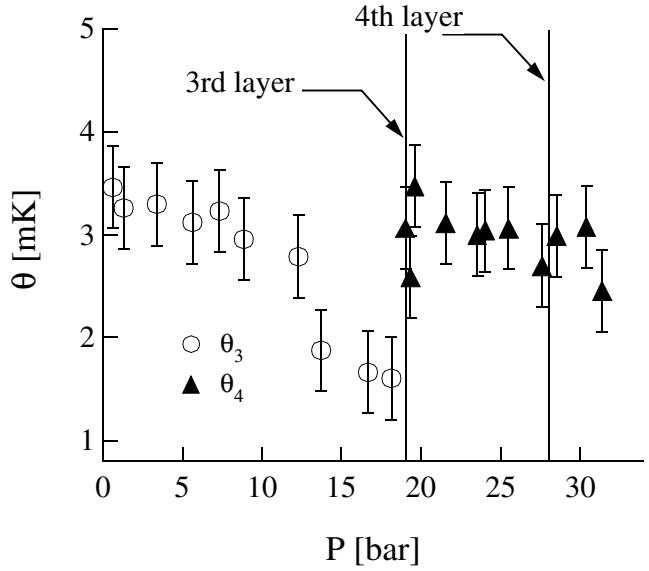

FIG. 5. Pressure dependence of the Weiss temperature, (○) θ_3 for the third layer and (▲) θ_4 for the fourth layer.

from the present data below a few mK. Moreover the observed pressure dependence of θ_3 reminds us of the density dependence of the Weiss temperature in the adsorbed ^{3}He film above 24 atoms/nm^2 [1].

In the higher pressure region than 19.35 bars, a similar analysis is made by assuming the growth of the fourth layer and adding the term $n_{4\text{th}}/(T - \theta_4)$ to Eq. (2). The first and second layers' parameters are assumed to be the same with in the above analysis. As for the third layer's parameters, the values obtained at 19 bars are employed, $n_{3\text{rd}}$ of 8.1 atoms/nm^2 and θ_3 of 1.6 mK. The results for $n_{4\text{th}}$ and θ_4 are shown in Fig. 4 and 5. The increase of n_{total} is similar to the third layer region. The difference of n_{total} between 0 and 28 bars is almost the same with the n_{total} at 0 bars, supporting our assumption that the third and the fourth layers are added to the two original solid layers. θ_4 does not show any particular pressure dependence, being nearly constant, in contrast to a large pressure dependence of θ_3 near the third layer completion. Moreover the value of θ_4 is large and independent of the fourth layer completion. A possible cause of this behavior is a weaker adsorption potential at the fourth layer position than at the third layer. Therefore the normal motion of ^{3}He atoms to the substrate is enhanced between the solid layer and the liquid. This situation is favorable for investigating a liquid mediated RKKY type interaction.

In summary, the third and fourth layers grow as a solid with increasing the liquid pressure. In our knowledge it is the first experimental evidence that more than two ^{3}He layers solidify on graphite surface. The upper solid layers show a strong ferromagnetic tendency that depends on liquid pressure. Further experiments, such as a layer by layer preplating with ^{4}He on the graphite surface, are desired in order to reveal the nature of new solid layers.

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