Low-Temperature Magnetization of Submonolayer ^{3}He Adsorbed on HD Preplated Graphite

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NMR studies of submonolayer ^{3}He adsorbed on a bilayer of HD preplated graphite have been made down to 100 μK, which is more than 1 order of magnitude smaller than the exchange energy (J). In the highly frustrated antiferromagnetic solid region, the magnetization obeys a Curie-Weiss law even at temperatures around J and then increases gradually down to 100 μK. Nevertheless, it does not show any anomalous behavior corresponding to a spin gap. The normalized magnetization versus the reduced temperature (T/J) is independent of the density just after solidification. This is consistent with the result in the high-temperature region, that the main multiple-spin exchanges have a similar density dependence.

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The frustrated quantum magnets have drawn much attention because the quantum fluctuations may destroy the long range order which is generally present in the classical ground state. Among many materials, a low density solid ^{3}He film adsorbed on graphite is proved to be one of the most ideal two dimensional $S = 1/2$ antiferromagnets on a triangular lattice [1]. Because of the hard-core potential between ^{3}He atoms, the higher order multiple spin exchange (MSE) processes as well as two-particle exchange play important roles [2,3]. The exchange of an even number of particles is antiferromagnetic (AFM), whereas that of odd ones is ferromagnetic (FM). The competition between them, in addition to the geometrical frustration, makes this system strongly frustrated.

In the second layer AFM solid, Ishida *et al.* observed a double peak structure in the heat capacity, which is characteristic of a highly frustrated system [4]. They pointed out the possibility of the quantum spin liquid (QSL) state as a ground state. Recent theoretical studies in the classical and the quantum limit give us various distinct ground states depending on the strength of the frustration [5,6]. The calculation of the exact diagonalization for a finite system showed that the strong competition between the three- and the four-particle exchange favors the QSL state with an excitation gap [5]. But the spin gap has not been observed experimentally either in the heat capacity [4] or in the magnetization [7], and hence the ground state is still an open question.

On graphite plated with a bilayer of HD, ^{3}He atoms solidify into a commensurate phase, whose density is 4/7 of the second layer HD. Because of the low density of the second layer HD (9.2 nm^{-2}), the 4/7 phase has a density of 5.2 nm^{-2}, the lowest ever achieved as a quantum solid. The low density significantly enhances the exchange interaction up to several mK [8,9], leading to the possibility of investigating the properties of the above mentioned frustrated system at much lower temperatures compared with the exchange interaction. However, the lowest temperature obtained so far is a little bit less than 1 mK [8]. In the present work, the temperature region is extended to the lower submillikelvin region to pursue a possible ground state.

CW (continuous wave) NMR measurements of ^{3}He film adsorbed on a bilayer of HD preplated graphite were made over the wide temperature range of 0.1–200 mK not only for the solid but also for the liquid with the same experimental cell as in our previous work [10]. The used substrate was exfoliated graphite (Grafoil GTY grade). A commercially available HD gas (purity 97.6%) contains pure isotopes of hydrogen (H_2 and D_2), which dissipate large heat from ortho-para conversion. To suppress the heat, it was forced to keep in touch with a charcoal catalyzer at 25 K overnight. Then it was carefully admitted through a heated capillary, while monitoring the pressure at the sample cell with a cryogenic pressure sensor. A step observed in a vapor pressure isotherm at 13.5 K determines the amount of HD gas necessary for a bilayer of HD preplating (18.3 nm^{-2}) and therefore the surface area. To prevent ^{3}He from going into the underlying second layer, HD was introduced up to 2.03 layers, slightly higher than a bilayer. After being annealed overnight, it was cooled down slowly to 2 K, and then ^{3}He gas was introduced and annealed. The ^{3}He areal density (ρ) is estimated from the available surface area and the amount of introduced ^{3}He gas. The samples were cooled by our powerful copper nuclear demagnetization stage. The temperature (T) was determined by a platinum NMR thermometer and a carbon resistor calibrated against a ^{3}He melting curve. NMR measurements were mostly made at a frequency of 618 kHz in a static field (191 G) parallel to the graphite sheet. Magnetization (M) was obtained from a numerical integration of the absorption line. No serious heating was observed even at 0.15 mK, judging from an NMR absorption signal of deuterium, which is expected to follow a Curie law.

Figure 1 shows the magnetization M in the liquid region. It exhibits a weaker increase than that for a Curie law as decreasing temperature, although it is expected to be constant in the investigated temperature region because of Fermi degeneracy at around a few hundred mK. The behavior is common to three densities except for a small difference arising from the density difference. The

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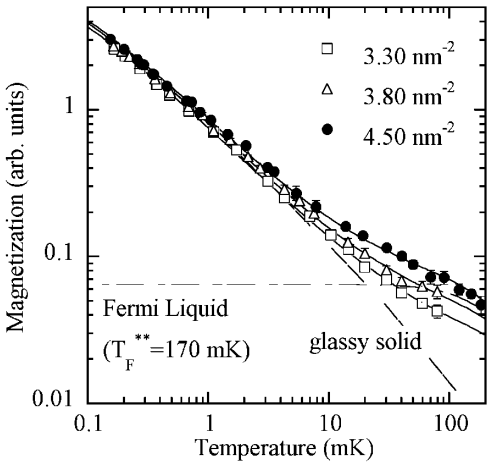

FIG. 1. The temperature dependence of the magnetization in the liquid region. Solid lines represent the fitting curves (see text). Contribution from the glassy solid (dashed line) and that from the Fermi liquid (dash-dotted line) are also shown for 4.50 nm^{-2}.

observed increase of M is attributable to a small amount of glassy solid ^{3}He trapped to the heterogeneities which cover about 10% of Grafoil surface [11]. Its existence has been pointed out to explain the temperature independent heat capacity term observed for the fluid layer adsorbed on pure ^{3}He [11] and also on a bilayer of HD [12]. Preplating with HD does not improve the situation, because HD crystals do not grow uniformly around the heterogeneities. In the glassy solid model [13], the exchange interaction J has a broad distribution, because of variations in ^{3}He density. For simplicity, a uniform distribution of the logarithm of J is assumed, which ranges from a lower energy cutoff J_l to a higher one J_h. Each spin locally orders and loses its entropy $k_B \ln 2$ at $T = J$, giving rise to a temperature independent heat capacity $k_B N_G \ln 2/\ln(J_h/J_l)$ and a total magnetization $c_0 N_G \ln(T/J_l)/[T \ln(J_h/J_l)]$, where N_G is the number of ^{3}He atoms in the glassy solid, c_0 is a Curie constant per spin, and k_B is the Boltzmann constant. The data are fitted to the following expression:

$$M = c_0\left(N_G \frac{1}{T}\frac{\ln(T/J_l)}{\ln(J_h/J_l)} + N_L \frac{1}{\sqrt{T^2 + T_F^{**2}}}\right),$$

where N_L is the number of ^{3}He atoms in the liquid phase and T_F^{**} is the effective Fermi temperature. The fitting is very good as shown by the solid lines in Fig. 1. Here we use $J_h = 30$ mK, the temperature below which the glassy solid contribution appears in the heat capacity of the ^{3}He second layer [14]. 10 μK, the value obtained from the fitting for 4.5 nm^{-2}, was chosen as J_l for all coverages. Such a small magnitude of J_l suggests the renormalization of the exchange interaction [15], because the adsorption potential to HD preplated graphite is too weak to form such a highly closed packing corresponding to 10 μK. The obtained T_F^{**} and N_G are shown in Fig. 2. T_F^{**} is not strongly affected by the choice of J_h and J_l and agrees very well with the results from the other preplated systems. N_G is about 10% of the total ^{3}He atoms, which is consistent with the amount of the heterogeneities in Grafoil. It slowly increases in proportion to the coverage but does not cross the origin, indicating that ^{3}He atoms are initially trapped to strong binding sites of the heterogeneities and then the glassy solids grow slowly. The same behavior was found in the first layer, where the heat capacity of the glassy solid appears in the beginning of the layer but does not change until a coverage a little before the next layer promotion [14].

Now we turn to the results in the solid region. The magnetization from the glassy solid should be subtracted from the raw data to get the solid ^{3}He magnetization in the flat surface. To estimate the former contribution, we use the extrapolated values of N_G shown by the solid line in Fig. 2(b). This procedure seems to be appropriate, judging from the fact that the heat capacity of the glassy solid does not change abruptly before and after solidification as mentioned above [14]. The temperature dependence of the magnetization thus obtained is shown in Fig. 3. Here the result for 5.61 nm^{-2} is also shown at 72.5 G corresponding to a Zeeman splitting of 11 μK. It can be reduced from that in the higher field (191 G) by multiplying a constant factor, justifying that the used field was too low to destroy the spin gap even if it exists. The high temperature behavior can be well fitted to a Curie-Weiss law (solid lines in Fig. 3). For 5.20 nm^{-2}, the fitting gives us a large Weiss temperature θ of -24 mK. The value, higher than

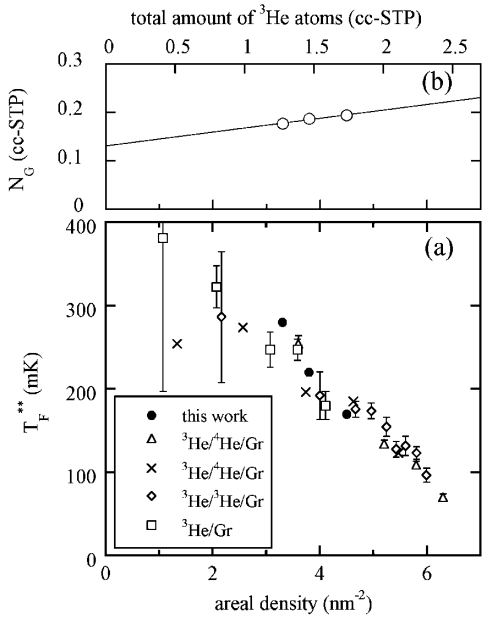

FIG. 2. (a) The obtained effective Fermi temperature (•), ^{3}He on a monolayer ^{4}He preplated graphite (△: [16], ×: [17]), second layer pure ^{3}He (◇: [18]), and monolayer ^{3}He on bare graphite (□: [19]). (b) The obtained N_G as a function of the areal density. The upper scale shows the total number of ^{3}He.

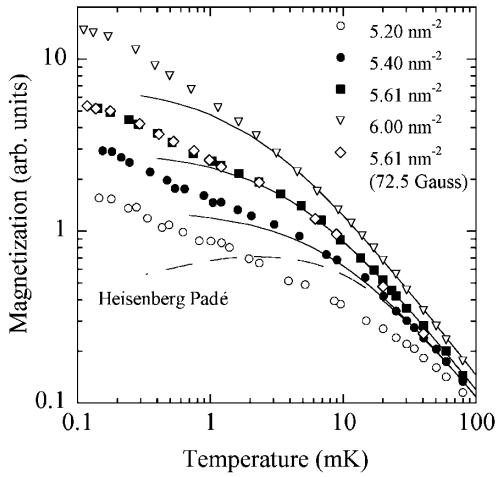

FIG. 3. Magnetization of the AFM solid. Solid lines represent a Curie-Weiss fitting. The [7,7] Padé approximant to the HTSE of the Heisenberg Hamiltonian for a triangular lattice ($J_\chi = \theta/3 = -3.0$ mK) is also shown for 5.40 nm^{-2} (dashed line).

the previous result [8], may be due to the existence of a small fraction of liquid component. At completely solidified density of 5.40 nm^{-2}, the behavior above 20 mK can be well fitted to the Curie-Weiss law with $\theta = -9$ mK, but M obeys the Curie-Weiss law down to 5 mK. Such a behavior was also pointed out by Siqueira *et al.* [8]. On the other hand, as shown in Fig. 3, a [7,7] Padé approximant to the high temperature series expansion (HTSE) of the Heisenberg Hamiltonian for a triangular lattice cannot reproduce our experimental data, suggesting the necessity of the higher order exchange interactions than the three-particle one. At further lower temperatures the data slowly deviate from the Curie-Weiss law and increase gradually down to 100 μK without any phase transitions or spin gap behavior. This behavior is consistent with the magnetization observed in the second layer ^{3}He [7] and is attributable to a huge number of low-lying excited states peculiar to the strongly frustrated system of two dimensional monolayer ^{3}He.

The obtained effective exchange interactions $J_\chi = \theta/3$ are summarized in Fig. 4(a). In the AFM region $|J_\chi|$ decreases rapidly as the areal density ($|J_\chi| \propto \rho^{-15\pm2}$). This steep density dependence is also seen in the effective exchange energies from the heat capacity J_c [9]. With increasing the coverage the liquid overlayer promotion is known to begin at 6.8 nm^{-2}, and the exchange interaction becomes FM. In the FM region, the magnetization curves after subtraction of the glassy solid contribution are fitted to the sum of a Curie-Weiss component and a Fermi liquid one. As shown in Fig. 4(b), the Curie constant of solid ^{3}He (C) saturates, indicating the solid density approaches constant. This result is not consistent with that of Casey *et al.,* who found a sudden reduction of C in the FM region [20].

Now let us focus on the AFM region. The normalized magnetization MT/C is given in Fig. 5 as a function of

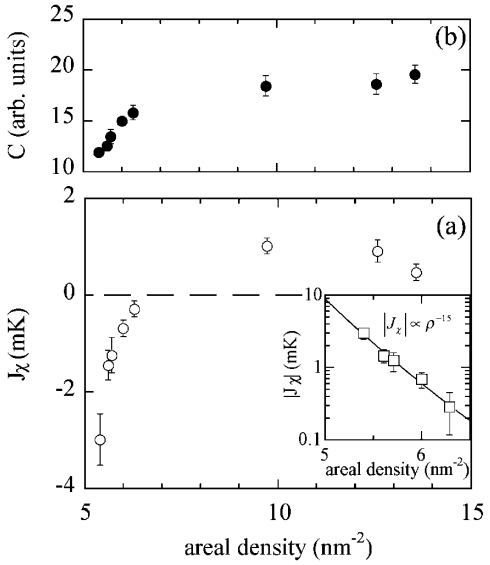

FIG. 4. (a) J_χ as a function of the areal density. The inset shows the areal density dependence of $|J_\chi|$. (b) Curie constant of solid ^{3}He as a function of coverage.

the reduced temperature $|T/J_\chi|$. In spite of a large change of J_χ by a factor of 4 from 5.40 to 6.00 nm^{-2}, the data for 5.40 and 5.61 nm^{-2} fall on the same curve and those for 6.00 nm^{-2} slightly deviate in the low-temperature region. This fact seems to be in contradiction with a naive idea that each n-particle exchange energy J_n should have a different density dependence. To resolve the difficulty, we inferred J_n by fitting the data to the HTSE of the MSE Hamiltonian including up to J_6 [21]. To reduce the fitting parameters, we assume that $J_4 = J_6$, $J_5 = 0.35J_4$, and fix J_c to the experimental value by Casey *et al.* [9]. Under this assumption, only J_4/J_2^{eff} is a fitting parameter, where $J_2^{\text{eff}} = J_2 - 2J_3$ is an effective two-particle exchange energy. After the fitting, the data are compared with $[L, M]$ Padé approximants generated from HTSE with

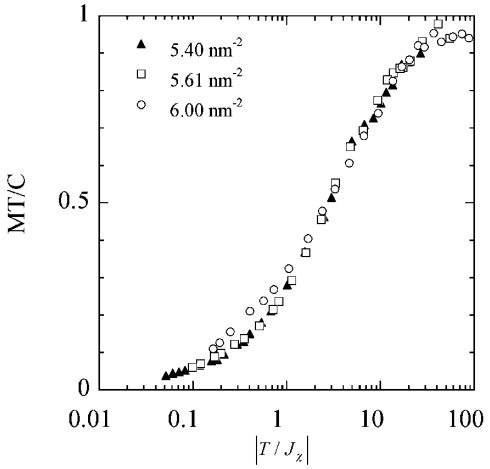

FIG. 5. MT/C as a function of $|T/J_\chi|$.

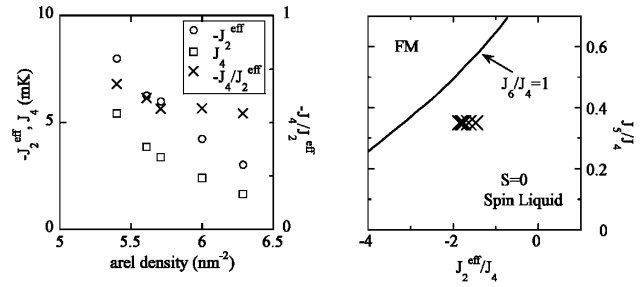

FIG. 6. (left) The obtained MSE parameters fitted to the HTSE of the MSE Hamiltonian under some assumptions described in the text. (right) Our MSE parameters ($\times$) are compared with the phase diagram of the ground state obtained by the exact diagonalization analysis [5,22]. The solid line represents the QSL-FM transition boundary for $J_6/J_4 = 1$.

the obtained parameters. [2,3] and [3,2] Padé approximants show excellent agreement with the data down to $T/J_c \sim 1.5$, where a $[L, M]$ Padé approximant is a rational function $P_M(u)/Q_L(u)$ with $P_M(u)$ and $Q_L(u)$, polynomials of degree M and L in $u = \beta/(1 - \beta)$ ($\beta = 1/k_BT$). The obtained J_2^{eff} and J_4 are summarized in Fig. 6. J_2^{eff} is negative, indicating three-particle exchange is dominant compared with two-particle exchange. $-J_2^{\text{eff}}$ and J_4 decrease monotonously with the density. Nevertheless, the frustration parameter J_4/J_2^{eff} changes very little in the region from 5.4 to 6.3 nm^{-2} in spite of a large change in J_χ. This fact is consistent with the universal behavior seen in Fig. 5. A small deviation of MT/C at 6.00 nm^{-2} may be due to a slightly stronger density dependence of J_4 than J_2^{eff}.

Recently Misguich *et al.* proposed a possible phase diagram of the ground state of the MSE Hamiltonian based on the exact diagonalization analysis [5,22]. The ground state for $J_2^{\text{eff}} < 0$ is composed of two regions, a QSL state with a spin gap and a FM state as shown in Fig. 6. Our obtained MSE parameters correspond to the QSL region near the QSL-FM transition boundary. However, any indications of the spin gap are not observed down to 100 μK. This fact could be due to a rapid decrease of the gap when approaching the boundary [5]. The exact diagonalization calculation was carried out only for the case of $J_4/J_2^{\text{eff}} = -0.5$, and further theoretical works are eagerly desired, in particular, in the region near the boundary.

In conclusion, we have made NMR measurements for submonolayer ^{3}He adsorbed on a bilayer of HD preplated graphite down to 100 μK. In the AFM solid region, the magnetization follows a Curie-Weiss behavior even at $T \sim J_\chi$ and then shows a gradual increase down to 100 μK. Even for $|T/J_\chi| = 0.05$ no anomalous behavior is observed, indicating a much smaller spin gap less than 100 μK even if it exists. MT/C as a function of $|T/J_\chi|$ hardly depends on the density below 6.0 nm^{-2}, reflecting a similar density dependence of the effective two and four exchange in the region.

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[1] H. Godfrin and R. E. Rapp, Adv. Phys. **44**, 113 (1995).
[2] M. Roger, C. Bäuerle, Yu. M. Bunkov, A.-S. Chen, and H. Godfrin, Phys. Rev. Lett. **80**, 1308 (1998).
[3] M. Siqueira, J. Nyéki, B. Cowan, and J. Saunders, Phys. Rev. Lett. **78**, 2600 (1997).
[4] K. Ishida, M. Morishita, K. Yawata, and H. Fukuyama, Phys. Rev. Lett. **79**, 3451 (1997).
[5] G. Misguich, B. Bernu, C. Lhuillier, and C. Waldtmann, Phys. Rev. Lett. **81**, 1098 (1998); G. Misguich, C. Lhuillier, B. Bernu, and C. Waldtmann, Phys. Rev. B **60**, 1064 (1999).
[6] T. Momoi, K. Kubo, and K. Niki, Phys. Rev. Lett. **79**, 2081 (1997); T. Momoi, H. Sakamoto, and K. Kubo, Phys. Rev. B **59**, 9491 (1999).
[7] C. Bäuerle, A.-S. Chen, Yu. M. Bunkov, H. Godfrin, and M. Roger, J. Low Temp. Phys. **113**, 287 (1998).
[8] M. Siqueira, C. P. Lusher, B. P. Cowan, and J. Saunders, Phys. Rev. Lett. **71**, 1407 (1993).
[9] A. Casey, H. Patel, J. Nyéki, B. P. Cowan, and J. Saunders, J. Low Temp. Phys. **113**, 265 (1998).
[10] H. Ikegami, K. Obara, D. Ito, and H. Ishimoto, Phys. Rev. Lett. **81**, 2478 (1998).
[11] M. Morishita, K. Ishida, K. Yawata, H. Nagatani, and H. Fukuyama, J. Low Temp. Phys. **110**, 351 (1998).
[12] A. Casey, H. Patel, J. Nyéki, B. P. Cowan, and J. Saunders, J. Low Temp. Phys. **113**, 293 (1998).
[13] A. Golov and F. Pobell, Phys. Rev. B **53**, 12 647 (1996); J. Low Temp. Phys. **99**, 191 (1995).
[14] M. Morishita, H. Nagatani, and H. Fukuyama, Physica (Amsterdam) **284B–288B**, 228 (2000).
[15] R. N. Bhatt and P. A. Lee, Phys. Rev. Lett. **48**, 344 (1982).
[16] C. Bäuerle, Yu. M. Bunkov, A. S. Chen, S. N. Fisher, and H. Godfrin, J. Low Temp. Phys. **110**, 333 (1998).
[17] C. P. Lusher, B. Cowan, and J. Saunders, Phys. Rev. Lett. **67**, 2497 (1991).
[18] K.-D. Morhard, C. Bäuerle, J. Bossy, Yu. Bunkov, S. N. Fisher, and H. Godfrin, Phys. Rev. B **53**, 2658 (1996).
[19] K.-D. Morhard, J. Bossy, and H. Godfrin, Phys. Rev. B **51**, 446 (1995).
[20] A. Casey, H. Patel, M. Siqueira, C. P. Lusher, J. Nyéki, B. Cowan, and J. Saunders, Physica (Amsterdam) **284B–288B**, 224 (2000).
[21] M. Roger, Phys. Rev. B **56**, R2928 (1997).
[22] G. Misguich, Ph.D. thesis, Université Paris, 1999.