Gapless Spin Liquid Behavior in Two-Dimensional Solid ^{3}He

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(Received 28 April 2003; published 16 January 2004)

Direct demagnetization has been made for two-dimensional solid ^{3}He in both the paramagnetic and the antiferromagnetic phases. The lowest temperature is about 10 μK, judging from the observed magnetization for the paramagnetic solid ^{3}He. The magnetization of the antiferromagnetic solid ^{3}He shows a gradual increase to about 10 μK for the 4/7 phase adsorbed on both one layer of ^{4}He and two layers of HD preplated graphite. This strongly suggests that the triangular antiferromagnet with the higher order multiple exchange has a quantum spin liquid ground state with nearly zero or extremely small spin gap less than 10 μK.

DOI: 10.1103/PhysRevLett.92.025301 PACS numbers: 67.80.Jd, 67.70.+n, 75.10.Jm

Highly frustrated low-dimensional quantum antiferromagnets have attracted much attention because a magnetically disordered ground state may be realized. A low-density solid ^{3}He film adsorbed on graphite surface is one of the most ideal two-dimensional (2D) quantum spin systems with nuclear spin $S = 1/2$ on a triangular lattice [1]. Because of the hard-core potential between ^{3}He atoms, the higher order multiple spin exchange processes as well as two-particle exchange play important roles [2]. The exchange of an even number of particles is antiferromagnetic (AFM), while that of an odd number is ferromagnetic. The competition between them, in addition to the geometrical frustration inherent to a triangular lattice structure, makes the solid ^{3}He film strongly frustrated. Recent theoretical studies in the exact diagonalization method for a finite system show that none of the long range ordered AFM phases survive even at $T = 0$ K due to quantum fluctuations. The exchange parameters so far obtained in the AFM region predicts a quantum spin liquid (QSL) ground state with a finite excitation gap [3].

Experimentally, Collin *et al.* measured the magnetization for the AFM solid ^{3}He phase, the so-called 4/7 phase, adsorbed on one layer of ^{4}He above 100 μK, and suggested a QSL ground state with a spin gap of the order of 100 μK [4]. On the other hand, Ishida *et al.* observed a double-peak structure followed by an unusual temperature dependence in the heat capacity [5]. The features led the authors to the suggestion of a disordered ground state for the AFM solid ^{3}He. The other 4/7 phase formed on graphite plated with a bilayer of HD has a much lower density than that of the second layer on the monolayer ^{4}He. The low-density significantly enhances the exchange interaction up to several mK [6,7], making it possible to study a low-dimensional frustrated quantum antiferromagnet at much lower temperatures than the exchange interaction (J_χ) obtained from the Weiss temperature $\theta = 3J_\chi$. Even for $|T/J_\chi| \sim 0.04$, the magnetizations for ^{3}He adsorbed on both two and three layers of HD show no evidence corresponding to a spin gap [8,9]. Hence the ground state of the 4/7 phase is still an open question. In this Letter, the first direct demagnetization of the 2D solid ^{3}He down to 10 μK region is reported.

We used a two-stage nuclear demagnetization refrigerator. The first stage is effectively 30 mol of copper nuclei at 9 T, and the second one is partly composed of the ^{3}He sample itself (~ 0.2 mmol) adsorbed on an exfoliated graphite, a so-called Grafoil (GTY grade [10]) with a total surface area of 12 m^2. Two Grafoil sheets (9 mm $\times$ 8 mm) were diffusion bonded on both sides of a copper foil 25 μm in thickness. The tabs extending from 25 copper foils were sandwiched with two copper sheets (0.5 mm thick and 8 mm wide) and diffusion bonded together. Two of these pieces, 38 mmol of copper nuclei in the effective field of 0.55 T, serve as the second stage. It is connected to the first stage via a zinc heat switch, the details of which have been described elsewhere [11]. As long as the heat switch is on, the ^{3}He temperature (T) can be measured with a platinum NMR thermometer on the first stage, which is calibrated against a ^{3}He melting curve thermometer. The whole second stage including the heat switch is housed in a Pyrex glass tube whose pressure inside is monitored with a cold pressure sensor at the first stage.

We have prepared the following three different samples: (I) the paramagnetic solid ^{3}He formed on graphite as the first layer, (II) the antiferromagnetic solid ^{3}He adsorbed on one layer of ^{4}He preplated graphite, and (III) the antiferromagnetic solid ^{3}He adsorbed on two layers of HD preplated graphite. Sample (I) was prepared by introducing ^{3}He gas [4.21 ccSTP (cm^3 in standard temperature and pressure)] and annealing overnight at 7.3 K. The areal density is estimated to be 9.4 nm^{-2} from the amount of introduced ^{3}He gas and the available surface area determined from the BET method. Sample (II) was prepared as follows. After an initial introduction of ^{4}He gas (6.45 ccSTP) and annealing overnight at 7.2 K, ^{3}He gas (2.84 ccSTP) was added and annealed at 2 K. To make a pure 4/7 phase without any influence of the heterogeneities of Grafoil, the amounts of both helium gas were chosen based on the previous work

in the same system [4]. For sample (III), special care was taken in the preparation of HD gas as described elsewhere [8]. The 2.03 layers of HD were formed and annealed at 14 K overnight. Then ^{4}He gas (0.52 ccSTP) and ^{3}He gas (1.92 ccSTP) were introduced and annealed in the same way as for sample (II). Judging from the previous measurement [9], the solid ^{3}He density is estimated to be comparable with 5.2 nm^{-2}.

The first step of a direct demagnetization experiment is to precool the second stage in the field of 0.55 T. The first stage was demagnetized step by step down to about 200 μK in 2 days. Then the temperature was kept for 5, 20, and 24 h for samples (I), (II), and (III), respectively, to have a complete thermal equilibrium. For sample (I), at $T = 150$ μK and $B = 0.55$ T, the ^{3}He spin system is highly polarized ($p > 99\%$) and most of the entropy is removed ($S < 0.032R\ln 2$). The second step is to demagnetize it in about 17 h from 0.55 T to the final and measuring field of 5 or 2.5 mT. The field sweep rate was controlled to be $dB/dt = -0.009$ mT/s so as to reduce the eddy current heating and to ensure a thermal equilibrium in the second stage. The temperature of the first stage was kept below 1.5 mK during the whole measurement (~ 3 days). NMR measurements were made with a continuous wave method at a frequency of 162 kHz (81 kHz) corresponding to a static field of 5 mT (2.5 mT) parallel to the graphite sheet. A silver rf coil was wound on the glass sample cell and the resonant frequency was adjusted by switching a tank circuit remotely at room temperature. The magnetization (M) was obtained from a numerical integration of the absorption line. The results are divided into three parts.

(1) The paramagnetic sample (I).—The direct demagnetization of sample (I) is useful, because the magnetization of this solid is expected to follow the Brillouin function, and therefore we can easily obtain the temperature of demagnetized ^{3}He itself. The magnetization of the ^{3}He and Cu nuclei in the second stage were measured at 5 mT (2.5 mT) and about 15 mT, respectively, in thermal equilibrium with the first stage over the temperature range of 0.25–3 mK. Both magnetizations obey a Curie law in this temperature region. Figure 1(a) shows the time dependence of the ^{3}He polarization p ($= M/M_s$) for the demagnetized sample. Here M_s is a saturation magnetization estimated from a Curie constant in the high temperature measurement. The polarization is converted to the ^{3}He spin temperature by using a Brillouin function as shown in Fig. 1(b). For the demagnetization from $B_i = 0.55$ T, $T_i = 240$ μK (250 μK) to $B_f = 5$ mT (2.5 mT), the ^{3}He is cooled down to $T_f = 12 \pm 0.6$ μK (8 ± 0.8 μK), where B_i (B_f) and T_i (T_f) are the initial (the final) field and temperature. The lowest temperature is higher than $T_f = 2.3$ μK (1.1 μK) for the ideal demagnetization. The discrepancy is explained as follows. In 2D ^{3}He adsorbed on graphite, the time constant for thermal equilibrium with Grafoil is typically 20 min at 100 μK.

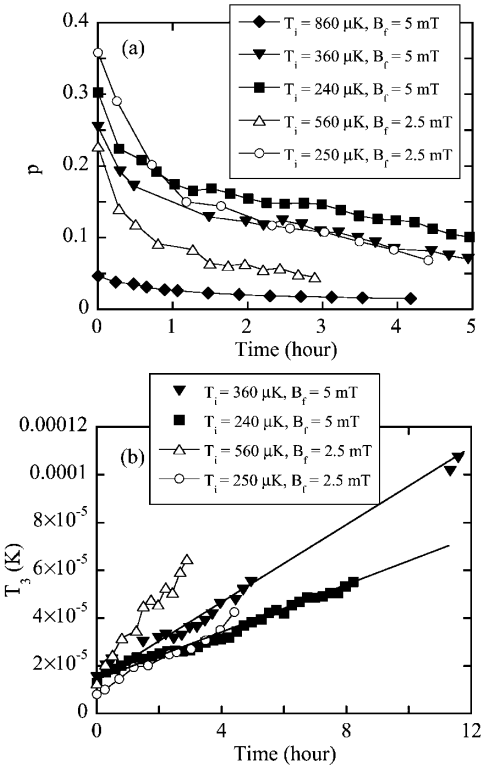

FIG. 1. (a) The time dependence of the polarization for the paramagnetic solid ^{3}He after demagnetization ($B_i = 0.55$ T). (b) The time dependence of the ^{3}He temperature obtained from its polarization. Solid lines represent the linear fitted ones.

Since it is expected to be shorter even below 100 μK than the demagnetization time of 17 h and the heat capacity of the paramagnetic ^{3}He is several hundred times smaller than that of the Cu nuclei, the ^{3}He nuclei is strongly connected with the demagnetized Cu nuclei via conduction electrons regardless of self-cooling of the ^{3}He nuclei. Now the Cu nuclear spin system in the second stage is cooled down to about 3 ± 0.2 μK for the demagnetization to $B_f = 5$ mT. The temperature difference between the Cu nuclei and the conduction electrons is estimated to be less than 1 μK, using the exact formula between them [12] and the total heat leak to the second stage, less than 10 pW, obtained from its temperature rise. Therefore the observed temperature difference between the ^{3}He and the copper nuclei should be due to the thermal conductance K_1 between the ^{3}He and the conduction electrons. K_1 is known from the heat capacity measurement for the first layer above 100 μK to have a temperature dependence of $1.8 \times 10^{-3}T$ (W/K) [13]. By use of this formula, a heat leak K_1 directly into the ^{3}He system, $\dot{Q} = \int K_1 dT$, is calculated to be approximately 0.1 pW. This value is quite reasonable judging from the estimated rf absorption during NMR measurement. Thus, the above heat bath model can be used to estimate the temperature of the demagnetized AFM solid ^{3}He.

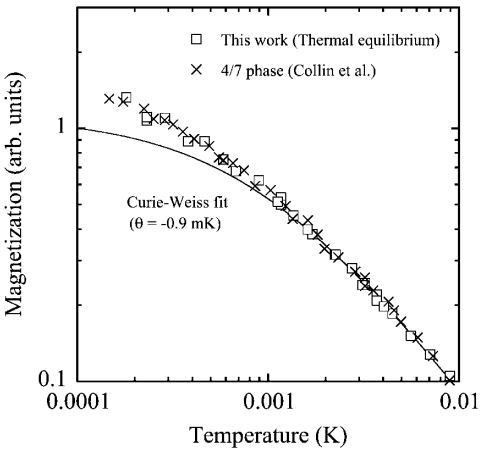

FIG. 2. Magnetization of the AFM solid ^{3}He adsorbed on one layer of ^{4}He in thermal equilibrium (□: our data; ×: Collin *et al.*). The solid line represents a Curie-Weiss fitting for our data.

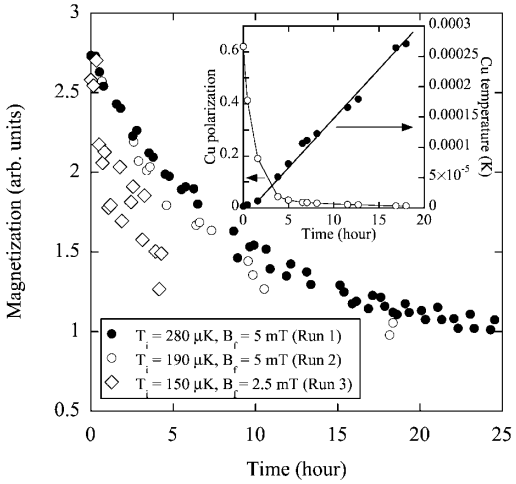

FIG. 3. The time dependences for the AFM solid ^{3}He adsorbed on one layer of ^{4}He after demagnetization. Inset: The time dependence of polarization and temperature for the Cu nuclei after demagnetization (run 2).

(2) The AFM sample (II).—Figure 2 shows the magnetization of the antiferromagnetic solid ^{3}He adsorbed on one layer of ^{4}He preplated graphite in thermal equilibrium. The behavior above 100 μK is fully in good agreement with that of the 4/7 phase which was measured by Collin *et al.* [4], indicating the formation of the same quality sample.

Figure 3 shows the time (t) dependence of magnetization after the demagnetization on three initial conditions. These magnetizations decrease smoothly with time, indicating no anomalous behavior in the system. In the warming up process, both magnetizations of ^{3}He and Cu nuclei were measured alternately at $B_f = 5$ mT by switching remotely one room temperature tank circuit (162 kHz) to the other (57 kHz). The temperature rise of Cu nuclei obtained from its polarization is given in the inset of Fig. 3. The lowest temperature of the Cu nuclei is 3 ± 0.2 μK, which is very close to that for sample (I) and is independent of the initial conditions. Since the ^{3}He is strongly connected with the Cu nuclei as mentioned above, we can estimate the ^{3}He temperature using the same heat bath model where the thermal conductance K_2 between the second layer solid ^{3}He and the conduction electrons is estimated to be $K_2 = 1.3 \times 10^{-3} T$ (W/K) from the heat capacity measurement for the second layer [13]. This estimation is reasonable, since the thermal relaxation times in the actual NMR measurement at several temperatures around 0.1 mK agree with those in the heat capacity measurement. The temperature difference (ΔT) between the ^{3}He and Cu nuclei is given for a constant heat load $\dot{Q}$ into the ^{3}He as $\Delta T = \dot{Q}/K_2 \propto \dot{Q}/T$. Since the measuring condition was the same as for sample (I), $\dot{Q}$ is roughly 0.1 pW at largest. Thus the lowest ^{3}He temperature is derived to be 14 ± 3 μK, and the ^{3}He temperature during warming is estimated from the Cu nuclear spin temperature whose rise is in proportion to time. The temperature dependence of ^{3}He magnetization thus obtained is given in Fig. 4, where the dashed line shows calculations for the spin gap of 80 μK using the formula by Collin [14]. The magnetization at 5 mT corresponding to a Zeeman splitting of 8 μK increases gradually to about 10 μK without any phase transition or spin gap behavior. This fact is consistent with the behavior obtained above 130 μK for the AFM solid on both two and three layers of HD preplated graphite [8,9] whose $|T/J_\chi|$ is close to the present value of 1/30.

(3) The AFM sample (III).—The magnetization of the 4/7 phase for submonolayer ^{3}He adsorbed on two layers of HD preplated graphite was obtained in thermal

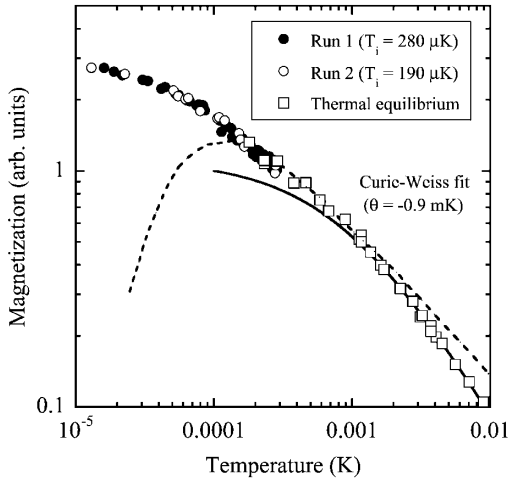

FIG. 4. Magnetization of the 4/7 phase for the AFM solid ^{3}He adsorbed on one layer of ^{4}He at 5 mT. The solid line represents a Curie-Weiss fitting with $\theta = -0.9$ mK. The dashed line shows the calculated one for the spin gap of 80 μK using the formula by Collin.

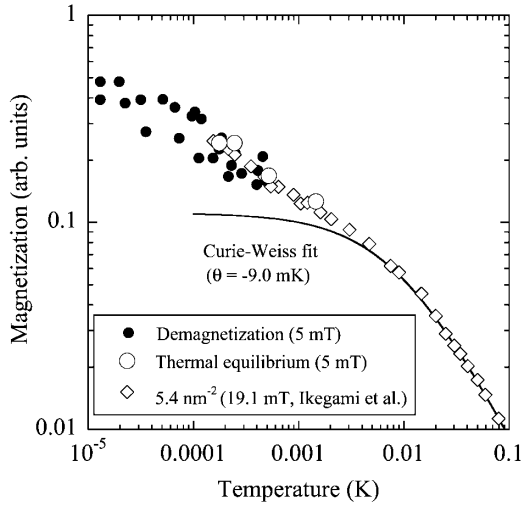

FIG. 5. Magnetization of the AFM solid ^{3}He adsorbed on two layers of HD. The high temperature data (△) above 100 μK is from Ref. [8]. The solid line represents a Curie-Weiss fitting with $\theta = -0.9$ mK.

equilibrium at 162 kHz corresponding to the field of 5 mT. The temperature dependence of the derived susceptibility is in good agreement with that obtained by Ikegami *et al.* [8], indicating θ of about -9.0 mK. The exchange interaction by almost one order of magnitude larger than sample (II) is very advantageous to reach much lower effective temperatures in the demagnetization runs. After the demagnetization from $B_i = 0.55$ T, $T_i = 176$ μK to $B_f = 5$ mT, both magnetizations of ^{3}He and Cu nuclei were measured alternately at $B_f = 5$ mT. In addition, the proton NMR signal in HD was monitored at 4 mT corresponding to 162 kHz in a separate run. The ^{3}He magnetization during the warm-up process was found to decrease smoothly with time, indicating no anomalous behavior in the system. The lowest temperature of Cu nuclei was 3 ± 0.2 μK, and its rise was similar to that for sample (II). The lowest temperature of the proton system was estimated to be about 10^{+9}_{-4} μK. These facts suggest that at 5 mT the Cu nuclei indirectly cool the proton and then the ^{3}He system. The temperature difference between the latter two should be negligible, judging from their close resonance frequency at such low fields and the much smaller heat capacity of the AFM ^{3}He phase than that of the proton system. Thus, the temperature rise of ^{3}He can be estimated from the Cu nuclear spin temperature by using the same heat bath model. Here we use the same K_2 as in sample (II), because the thermal relaxation time observed in the NMR measurement for samples (II) and (III) was almost the same in the temperature region around 0.1 mK. As is shown in Fig. 5, the magnetization increases gradually to 10 μK, which is more than 300 times smaller than the exchange interaction, without any phase transitions or spin gap behavior.

In conclusion, a direct demagnetization has been successfully performed for both two-dimensional paramagnetic and two-dimensional antiferromagnetic solid ^{3}He. The magnetizations for the antiferromagnetic solid ^{3}He adsorbed on both one layer of ^{4}He and two layers of HD preplated graphite increase gradually to about 10 μK. No anomalous behavior corresponding to the spin gap is observed, even for $|T/J_\chi| \sim 1/300$. This fact indicates that there are a huge number of low excited states peculiar to the strongly frustrated system of two-dimensional monolayer ^{3}He, suggesting a strong possibility of a quantum spin liquid ground state with a nearly zero or an extremely small spin gap less than 10 μK in the 2D triangular antiferromagnetic system with the multiple spin exchange. The above results are consistent with the heat capacity data observed in the second layer [5], but in contradiction with the theoretical prediction in the exact diagonalization for a finite size. The system size in these calculations might be too small because the higher order multiple spin exchange processes play important roles. Further theoretical calculations for a larger size of the system are eagerly desired.

We are grateful to M. Morishita and H. Fukuyama for showing us their thermal conduction data prior to publication.

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