Magnetization curve of second layer antiferromagnetic solid $^3$He on graphite in high magnetic fields

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Abstract. Two dimensional anti-ferromagnetic solid $^3$He adsorbed on graphite, so called 4/7 phase, is a strongly frustrated quantum spin system and the ground state is considered to be a gapless spin liquid. To clarify the magnetic behavior, the magnetization curve has been investigated below 1 mK in high magnetic fields, using NMR over the wide frequency region up to 360 MHz. The magnetization is found to reach full saturation at around 10 T.

1. Introduction
Frustrated magnetic systems have attracted strong attention since older time due to the presence of remarkable properties. Among various materials, a low-density solid $^3$He film adsorbed on a graphite surface is one of the ideal systems, since it is a real two-dimensional (2D) quantum spin system on a triangular lattice with nuclear spin $S = 1/2$ [1]. Because of the hard-core potential between $^3$He atoms, higher order multiple spin exchange (MSE) processes as well as two-particle exchange play important roles. The exchange of an even number of particles is anti-ferromagnetic (AFM), while that of an odd number is ferromagnetic (FM). The competition between them, in addition to the geometrical frustration inherent in the AFM triangular lattice, makes solid $^3$He film strongly frustrated. Recent experiments suggest that the ground state of AFM 4/7 phase is a spin liquid either gapless [2, 3] or with a small gap [4], whereas the exact diagonalization for the MSE Hamiltonian predicts the existence of a finite spin gap [5]. Moreover some theoretical studies predict the appearance of plateau in the magnetization curve [5, 6]. To resolve the discrepancy, it is desirable to investigate the magnetic behaviour in high magnetic fields. The first magnetization measurement for the 4/7 phase in high fields up to 10 T has been made with a double gradient Faraday magnetometer [7]. Although the magnetization curve seems to have a plateau, it has some ambiguity due to a large background signal from graphite itself. To overcome this difficulty, we have tried to apply uhf NMR to get a more precise magnetization curve. The results below 100 MHz were given elsewhere [8]. Here we mainly present those above 100 MHz up to 360 MHz.

2. Experimental
The experimental set-up is similar to that in our previous work [8]. The substrate used is an exfoliated graphite (Grafoil GTY grade $d = 76 \mu$m) [9], which consists of partially aligned micro
crystals of graphite. It was degassed at 1000 °C in vacuum for 24 hours. Two Grafoil sheets (8 mm × 8 mm) were diffusion bonded on both sides of a silver foil 8 mm wide and 20 µm in thickness. The silver tabs extended from such 42 silver-Grafoil sandwiches were stacked into 7 grooves of well annealed silver block. They were diffusion bonded and housed into a Stycast 1266 sample cell. The end of the silver block coming from the cell was fastened tightly to a well annealed massive silver thermal link with large RRR of the order of 2000 or more. The cross section of the link is designed to increase step by step from the middle of magnet central region to the powerful nuclear stage in order to reduce the temperature gradient along it. The temperature difference there is estimated to be 50 µK even for the total heat leak into the sample cell. Thus a \(^3\text{He}\) melting curve (MCT) and a Pt wire NMR thermometer installed in the low field region were used to determine the sample temperature. Thermal relaxation was measured by monitoring \(^3\text{He}\) NMR signal whenever de-magnetized. For example, it took about 2 hours at 0.7 mK and 3 T to have a thermal equilibrium. Depending on the temperature and magnetic field, the waiting time was varied.

\[ T = 3.02 \text{ mK} \]

Figure 1. Schematic diagram of uhf NMR spectrometer and a typical signal

\(^3\text{He}\) NMR measurements were made with a continuous wave method where an rf coil was connected through about 3 m coaxial cable in the cryostat to a room temperature circuit. The resonance frequency has to be varied over the wide frequency region from 100 to 360 MHz to get the magnetization curve without warming up the cryostat. For the purpose, we tried to excite various quarter wave length standing wave modes by varying a coax cable length outside the cryostat. At the top of cryostat, we put a hybrid junction combined with an impedance matching
device as is shown in Figure 1. At the same time the frequency modulation and the homodyne detection technique were used to improve the S/N ratio. As was pointed out by Rall et al. [10], NMR measurements above 100 MHz cause strong rf shielding from exfoliated graphite sheets and also large rf heating there. A possible heating effect was investigated at 263 MHz by putting a huge rf power into the tank circuit in the temperature region of dilution refrigerator. The heat input is found to be $Q = 0.12 \times V_{rf}^2$ in the present set-up. Although it is not serious in the actually used rf level, the rf field was varied at lower temperatures to confirm no saturation of $^3$He spin system and no rf heating in each static field. A static magnetic field up to about 15 T with a high homogeneity of $10^{-5} \text{cm}^3$ was applied in parallel to the Grafoil sheets and swept slowly to cover the whole NMR absorption line. An rf coil with 2 turns wound around the sample cell produced an rf field parallel to the Grafoil sheets. Magnetization ($M$) was obtained by integrating twice the derivative of absorption line. One of large errors comes from the subtraction process of the base lines, and is the order of 10%.

We have prepared the following two different samples: (I) anti-ferromagnetic solid $^3$He adsorbed on one layer of $^4$He pre-plated graphite, (II) paramagnetic solid $^3$He formed on graphite as the first layer. The sample (I) was prepared as follows. After an initial introduction of $^4$He gas and annealing 4 hours at 4.3 K, $^3$He gas was added and annealed 4 hours at 2.7 K. To make a 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 [3, 4]. The magnetization was measured at 0.392 T, and the obtained Weiss temperature of $-0.9 \text{mK}$ indicates the forming of similar 4/7 phase to those before [3, 4]. The sample (II) was prepared by just introducing $^3$He gas and annealing 5 hours at 3.0 K. The areal density is estimated to be $11.0 \text{nm}^{-2}$ from the amount of introduced $^3$He gas and the available surface area determined from an adsorption isotherm measurement. The paramagnetic sample (II) was used to confirm that the $^3$He sample was really cooled down to below 1 mK at 2.6 T, since the magnetization saturates at a few mK in the higher fields. Figure 2 shows the temperatures dependence of magnetization. It follows a Brillouin function (solid lines), indicating no serious problem in the refrigeration and measuring system.

3. Results and discussions

Now the temperature dependence of magnetization for 4/7 phase is shown in Figure 3 in several high magnetic fields. The vertical axis is the polarization, that is, the magnetization normalized by the corresponding saturation magnetization ($M_s$) in each magnetic field. $M_s$ is obtained by fitting the high temperature data to a Curie Weiss law ($\theta = 3J = -0.9 \text{mK}$) as shown in the inset (dashed lines). This procedure has to be done in each static field, since the raw NMR signal strongly depends on the resonance frequency. The magnetization at 9.93 T follows a Brillouin function and reaches the full saturation at about 10 T.

As is mentioned in the beginning, there are two theoretical predictions in the frame of MSE model that predict the saturation field ($H_s$) [5, 6]. G. Misguich et al. used the following MSE parameters, $J_{2/3}^{4/7}/J_4 = -2$, $J_5/J_4 = 0.2$, $J_6/J_4 = 0.08$ [5]. If we use $J_4 = 1.4 \text{mK}$, the experimentally obtained value [4], $H_s$ is estimated to be about 5 T, which is roughly half of the observed field. The discrepancy is possibly attributed to their much smaller $J_6/J_4$ than the experimentally derived value of 0.8 [4]. While according to T. Momoi et al. [6], $H_s$ is estimated to be about 20 T for the same $J_4 = 1.4 \text{mK}$, which is larger by a factor of two than the observed value. However $J_5$ and $J_6$ are neglected in their calculation. Even if a small ferromagnetic $J_5$ is taken into account, the effective $J_4$ gets small and $H_s$ is expected to decrease rapidly. Therefore a proper set of MSE parameters, in particular a suitable amount of five and six spin exchange term, seems to be important in order to explain the obtained result.
Figure 2. Temperature dependence of magnetization for paramagnetic sample in the magnetic fields below 2.59 T.

Figure 3. Temperature dependence of magnetization for 4/7 phase in the magnetic fields above 4.22 T. Inset: the solid lines are Brillouin functions.

4. Conclusion
The magnetization of 4/7 phase is found to saturate at about 10 T. This value is just between two theoretically predicted ones in the multiple spin exchange model. The proper amount of five and six spin exchange interaction term seems to be important to reproduce the obtained results.

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