Heat Capacity of $^3$He Solid Films on Graphite in Weak Magnetic Fields

Masashi Morishita
Graduate School of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-8571, Japan
E-mail: morishit@sakura.cc.tsukuba.ac.jp

Abstract. The heat capacities of the second layer $^3$He solid films adsorbed on a graphite surface were measured in magnetic fields up to 300 G. The applied magnetic fields are considerably weaker than the exchange interaction between the nuclear spins of $^3$He. The results reveal unexpected behaviors. In the antiferromagnetic film, the double peak structure of heat capacity is enhanced by the magnetic fields. In the ferromagnetic films, the heat capacity peaks are remarkably reduced. These results might indicate that the ferromagnetic films do not demonstrate simple fully saturated ferromagnetism and that the magnetic fields strongly affect magnetic frustration.

1. Introduction
A $^3$He solid film adsorbed on a graphite surface is an almost ideal two-dimensional quantum spin system [1]. The magnetic behavior of the second layer $^3$He solid film changes from antiferromagnetism to ferromagnetism with increasing areal density [2]. It is believed that the magnetism of the $^3$He solid film arises from a competition between multiple spin exchange (MSE) interactions, and the evolution of magnetism is a consequence of different density variation of magnitude of each MSE interactions [3]. The competition between the MSE interactions can be a source of strong magnetic frustration. From the previous heat capacity measurements of the second layer $^3$He solid films, the antiferromagnetic phase has been considered to be strongly frustrated, while the ferromagnetic phases to be almost simple Heisenberg systems [4, 5].

In this paper, the results of heat capacity measurements of the second layer $^3$He solid films in weak magnetic fields are reported. Although the applied magnetic fields are considerably weaker than the exchange interactions between the nuclear spins of $^3$He, the behavior of the heat capacity changes considerably. The results suggest that the magnetism of $^3$He solid films is more complicated than ever has been believed.

2. Experimental
Grafoil sheets, whose surface area is estimated to be 420 m$^2$, are used as graphite substrates. The magnetic field is applied parallel to the surfaces of the Grafoil sheets. Heat capacity was measured by the adiabatic heat pulse method above 1 mK and by the relaxation method below 3 mK. The other experimental setups and techniques are almost the same as those employed in our previous studies[5, 6].
3. Results and Discussion
The results of the heat capacity measurements of $^3$He solid films for four different areal densities in three different magnetic fields are shown in figure 1. Here, the contribution to the heat capacity from the amorphous $^3$He adsorbed on the heterogeneous surface of the graphite substrate [7] is not subtracted, as its dependence on magnetic field is not known. Owing to the apparatus circumstances, the amplitude of the magnetic field is limited below 150 G for the adiabatic heat pulse method, while the relaxation measurements below 3 mK are performed in magnetic fields up to 300 G. No difference can be distinguished above 3 mK between the results for the magnetic fields of 0 G and 150 G. In the previous heat capacity measurements of the first layer $^3$He solid films, which was limited to observing the high temperature slopes of heat capacity peaks, anomalously large shifts toward higher temperatures with increasing magnetic field were observed [6]. The magnitudes of the shifts are ten times larger than the values expected due to

![Figure 1](image_url)

**Figure 1.** The heat capacity of $^3$He films with the areal densities of 18 nm$^{-2}$ (a), 23 nm$^{-2}$ (b), 26 nm$^{-2}$ (c), and 35 nm$^{-2}$ (d) in the magnetic fields of 0, 150 and 300 G. The lines are a guide for the eye. The contribution from amorphous $^3$He has not been subtracted. The contribution from the fluid overlayers has been subtracted in (b)-(d).
the Zeeman effect. Even if a similar shift exists in the case of the second layer solid $^3$He, the magnitude of the shift is 30 $\mu$K at 150 G, and it is too small to be detected at temperatures above 3 mK. Below 3 mK variations due to the magnetic field are observed. They are not simple shifts as those observed in the high temperature slope of heat capacity of the first layer solid. In figure 2, the differences between the heat capacity in magnetic field of 300 G and the smoothed values of measured heat capacity in a zero magnetic field are shown. The magnitude of magnetic field dependent variations changes with the areal density; hence, the magnetic field dependent variation must not arise only from extrinsic sources. The contribution from the amorphous $^3$He is thought to almost independent on the areal density between 18 and 35 nm$^{-2}$ [7]. Figure 2 strongly suggests that the contribution from amorphous $^3$He is not responsible for the magnetic field dependent variation. In the following sections, the results are discussed individually.

Figure 1(a) presents the results for a film with an areal density of 18 nm$^{-2}$. At this areal density, the second layer solid film is believed to have the 4/7 structure commensurate to the first layer [8], and it demonstrates an antiferromagnetic behavior [2]. The MSE competition and the geometrical frustration are strong, and the spin liquid state has been proposed as the ground state [5]. The heat capacity in a zero magnetic field exhibits a double peak structure around this areal density [5]. By applying a magnetic field, the lower peak increases in height, and the double peak structure becomes further conspicuous. A similar but smaller change has been observed with a modest increase in the areal density [5]. The double peak structure is believed to be a result of the strong magnetic frustration due to the strong competition between the MSE interactions [5, 9]. The growth in the height of the lower peak might result in an increase in magnetic frustration. The strong magnetic frustration might be affected by the applied magnetic field.

At higher areal densities, fluid overlayers exist that are believed to behave as a two-dimensional Fermi liquid. The heat capacity of the fluid overlayers are estimated by fitting the results above 20 mK with the following formula: $\gamma T + \gamma_2 T^2 + AT^\alpha$. The estimated contributions of the fluid overlayers, $\gamma T + \gamma_2 T^2$, are subtracted from the total heat capacity, and the remainders are illustrated as spin contributions in figures 1(b)-(d). They illustrate the magnetic field dependent variations of the heat capacity of $^3$He solid films with areal densities of 23, 26 and 35 nm$^{-2}$, respectively. At these areal densities, the second layer of $^3$He films demonstrate ferromagnetic behaviors [2]. The effective exchange interactions obtained from
the magnetization measurements exhibit a rounded peak around 26 nm$^{-2}$. The ferromagnetic tendency is believed to arise from the predominance of the 3-spin exchange in the higher areal density region. Further, the competition between MSE interactions is believed to be weaker at high areal densities. At 23 nm$^{-2}$, the heat capacity does not change with the magnetic field, except for a gradual increase below 1 mK, as shown in figure 1(b). While at the areal densities of 26 and 35 nm$^{-2}$, distinguished decreases in the peak heights are observed, as shown in figures 1(c) and 1(d). The spin entropy changes calculated from the results of the heat capacity measurements within the measured temperature range decrease with increasing magnetic field. It implies that the growth in the short range spin ordering is suppressed by the magnetic field even though the interaction is ferromagnetic, if the heat capacity does not change with the magnetic field in the higher temperature range. The results presented in figure 1(d) suggest that the heat capacity in the higher temperature range does not change for magnetic fields of up to 300 G at least for an areal density of 35 nm$^{-2}$. The short range spin ordering in a ferromagnet could be suppressed by a magnetic field if magnetic anisotropy exists. However, the demagnetization field and dipole interaction are extremely weak, and anisotropy cannot exist in this system. The RKKY interaction through the fluid overlayers has been discussed as the origin of the ferromagnetic behavior [10]. If the overlayers were to be polarized by the applied magnetic field, the ferromagnetic tendency may be reduced. However, the overlayers behave as a two-dimensional Fermi liquid, and it is impossible to polarize them with a weak magnetic field of the order of a few hundred G. Although the ground state of the ferromagnetic phase is tacitly understood to be fully polarized, it has not been confirmed. In the magnetization measurement by Shiffer et al., only 80% of the saturation magnetization is obtained at 26 nm$^{-2}$ when the measured magnetizations are extrapolated to $T = 0$ [11, 12]. If the spins have structures such as canted antiferromagnetism, a magnetic field could suppress the spin short range ordering.

4. Conclusions
The heat capacity of the second layer $^3$He solid film is found to be strongly affected by weak magnetic fields. The variations are not simple shifts as observed in the first layer. Although the origins of these variation have not been elucidated, the observed behavior suggests that the spin system of $^3$He solid films is rather complicated. Performing measurements in a stronger magnetic field is necessary to understand.

Acknowledgments
All experimental measurements has been performed using the commonly used equipments at the Cryogenics Division, Research Facility Center for Science and Technology, University of Tsukuba. This research is supported by the Grants-in Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References
[1] Godfrin H and Lauter H J 1995 Progress in Low Temperature Physics vol XIV, ed W P Halperin (Amsterdam: Elsevier) p 213
[2] Godfrin H, Morhard K D, Rapp R E and Bunkov Yu M 1994 Physica B 194-196 675
[3] Roger M 1984 Phys. Rev. B 30 6432
[4] Greywall D S 1990 Phys. Rev. B 41 1842
[5] Ishida K, Morishita M, Yawata K and Fukuyama H 1997 Phys. Rev. Lett. 79 3451
[6] Morishita M 2007 J. Low Temp. Phys. 148 761
[7] Morishita M, Nagatani H and Fukuyama H 2002 Phys. Rev. B 65 104524
[8] Elser V 1989 Phys. Rev. Lett. 62 2405
[9] Roger M 1990 Phys. Rev. Lett. 64 297
[10] Jichu H and Kuroda Y 1983 Prog. Theor. Phys. 69 1358
[11] Schiffer P, O’Keefe M T, Osheroff D D and Fukuyama H 1993 Phys. Rev. Lett. 71 1404
[12] Schiffer P, O’Keefe M T, Osheroff D D and Fukuyama H 1994 J. Low Temp. Phys. 94 489