Density Variation of the Frustrated Ferromagnetism in 2D Solid $^3$He

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Abstract We measured the heat capacities of the second layer incommensurate solid $^3$He adsorbed on graphite preplated with monolayer $^4$He ($^3$He/$^4$He/gr) in a wide temperature range ($0.3 \leq T \leq 80$ mK). From the high temperature series expansion fitting, we obtained the density dependences of multiple spin exchange interactions. The value of effective two-particle exchange interaction $J$ is quantitatively similar to that for $^3$He/$^3$He/gr at the highest densities, while it is clearly smaller than that for $^3$He/$^3$He/gr at the lowest densities. We conclude that this difference of $J$ is caused by the different second layer density for the additional potential from the first layer. We also show the spin entropy of the second layer solid. Curiously, the total spin entropies are about 18% smaller than expected value.

Keywords quantum solid · two dimensional system · multiple spin exchange · helium-3

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1 Introduction

Solid helium three ($^3$He) is well known as a quantum solid in which atom-atom exchanges occur even at zero-temperature by quantum tunnelling due to a large zero-point energy. Because of the steric hindrance caused by the large hardcore repulsion between atoms, the multiple spin exchange (MSE) among $^3$He nuclear spins ($S = 1/2$) plays an important role in deciding the nuclear magnetism of bulk solid $^3$He [1,2]. Even permutations like three- or five-particle ring exchanges generally favor ferromagnetism, while odd permutations like two- or four-particle exchanges favor antiferromagnetism [3]. The competition of these MSE induce a frustrated magnetism to the system.

Solid $^3$He films adsorbed on the atomically flat surface of graphite make an ideal two-dimensional quantum solid on triangular lattice, and it shows interesting magnetic

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properties [4, 5]. Especially in the second layer, large exchange interactions $J$ of several mK is observed, and it shows a highly frustrated magnetism [6, 7]. These values are about one order higher than those in the first layer [8]. So, it is easier in the second layer to measure the heat-capacity or the nuclear magnetic susceptibility down to much lower than $J$ [9, 10].

In $^3$He/$^4$He/gr system in which the first layer atoms are $^4$He instead of $^3$He, $^3$He atoms behave as a 2D Fermi fluid when their areal density ($\rho$) is relatively low. After the intermediate region where the vacancy doped phase is proposed [11], they localize at a commensurate density $\rho = 6.8$ nm$^{-2}$ which is four seventh of that of the first layer. This commensurate solid is called the “4/7 phase”, and the magnetic ground state of this phase is believed to be the gapless spin liquid [9, 10]. The 4/7 phase is stable against additional particles up to about 20%, and after that, the second layer changes to a incommensurate (IC) solid with a first order transition at $8.0 \leq \rho \leq 9.5$ nm$^{-2}$ [12]. In this work, we measured heat-capacities of the second layer IC solid $^3$He ($\rho \geq 9.5$ nm$^{-2}$) adsorbed on graphite preplated with monolayer $^4$He ($^3$He/$^4$He/gr) in wide temperature range ($0.3 \leq T \leq 80$ mK). Experimental setup is the same as that in [12], and also described in [13].

Fig. 1 (Color on-line) Temperature dependences of $C$ at the second layer IC solid region.

### 2 Results and Discussion

Fig. 1 shows the heat-capacities ($C$) of all five samples taken at the IC region ($\rho \geq 9.5$ nm$^{-2}$). In this region, the magnetic susceptibility shows ferromagnetic behaviours [14]. We can indentify a ferromagnetic peak below several mK and a heat capacity of degenerate Fermi fluid from overlayer at high temperature region. The ferromagnetic peak gradually decreases its temperature ($T_{peak}$) and increases its height with increasing
density. We can also see the $C \propto T$ behaviors at $T \ll T_{\text{peak}}$ in all samples. This $T$-linear heat-capacity is understood by the 2D ferromagnetic spin wave theory, and also observed in earlier experiment in $^3\text{He}/^3\text{He}/\text{gr}$ [9]. So, we can say that the measured is low temperature enough to see the whole $T$-dependences of $C$ of the second layer solid.

To study MSE interaction, we fit the measured $C$ by the following expression:

$$C = N_2 k_B P(2, 3) + \gamma T + \Gamma T^2$$

(1)

at $T < T_{\text{peak}}$. Now, $N_2$ is the number of atoms in the second layer and $k_B$ is the Boltzmann constant and $P(2,3)$ is a $(2,3)$ Padé approximant after a Euler transformation to the high-temperature series expansions (HTSE) of the specific heat up to the fifth order for the MSE Hamiltonian [15]. The last two terms are the leading terms in degenerated 2D Fermi fluid [16,17]. $N_2$ is determined from the theoretical calculation of the density of the second layer IC solid by M. Roger et al. [18] which is fitted to the two data points of the neutron diffraction on pure $^3\text{He}$ film ($^3\text{He}/^3\text{He}/\text{gr}$) [19]. For applying this calculation to our system ($^3\text{He}/^4\text{He}/\text{gr}$), we assume that the density of the second layer solid is not related to the kind of the particle in the first layer at the IC region. MSE Hamiltonian in [15] includes up to six-particle permutation: effective two-particle exchange interaction $J = J_2 - 2J_3$, effective four-spin exchange constant $K = J_4 - 2J_5$, the relative ratios of five- and six-particle exchange interaction $\eta = J_5/J_4$, $\nu = J_6/J_4$ are used as fitting parameters.

![Fig. 2](Color on-line) HTSE fitting to the heat capacity after removing the overlayer fluid at $\rho = 11.30$ nm$^{-2}$. The dashed line is $N_2 k_B P(2, 3)$ in (1), where $N_2 k_B = 60.0$ mJ/K, $J = -3.25$ mK, $K = 0.30$ mK, $\eta = 0.30$, and $\nu = 0.49$. The inset shows the total heat capacity including overlayer fluid. The dashed-dotted line is the fluid terms in (1) and the solid line is equation (1).
An example of HTSE fitting at $\rho = 11.30 \, \text{nm}^{-2}$ is shown in Fig. 2. The qualities of fitting are almost same in all samples. In all densities, we fixed the parameter $\eta = 0.3$ based on the first-principle calculation [20]. We also fixed the parameter $\nu = 0.45$ at $\rho = 13.42 \, \text{nm}^{-2}$ because $\nu$ is not converged at this density where only small contribution of $J_6$ exists. The MSE interactions are shown in Fig. 3. The contributions of higher order exchanges ($-K/J$) decreases with increasing density, and the ratio between four- and six-particle exchange ($\nu$) don’t shows large density dependence. These results are consistent with early studies in $^3\text{He}/^3\text{He}/\text{gr}$ [7] (The inset of Fig. 3). On the other hand, we can see a important difference in $J$ at $\rho \leq 11 \, \text{nm}^{-2}$. $J$ obtained in this work ($^3\text{He}/^4\text{He}/\text{gr}$) is much smaller than that of $^3\text{He}/^3\text{He}/\text{gr}$ [7,9]. At high density ($\rho \geq 12 \, \text{nm}^{-2}$), there is no difference of $J$ in each system.

![Fig. 3 (Color on-line) Density dependence of the effective two-particle exchange interaction $J$. The inset shows the ratio of the higher order exchanges $-K/J$ and $\nu$. Filled (open) circles are the results in this work ($^3\text{He}/^4\text{He}/\text{gr}$). Other open (filled) symbols are corresponding to the earlier studies in $^3\text{He}/^3\text{He}/\text{gr}$. Squares [7] and Diamonds [9] are parameters deduced from heat-capacity fits. Triangles [7] are deduced from susceptibility fits. The solid lines are guides for the eye.]

For the interpretation of this nature, firstly, we point out the heat-capacity peak temperatures (a higher temperature peak of two peaks) of the 4/7 phase are 1.2 mK in $^3\text{He}/^4\text{He}/\text{gr}$ [12,11] and 1.8 mK in $^3\text{He}/^3\text{He}/\text{gr}$ [9]. This difference of $T_{\text{peak}}$ can be understood by the different commensurate densities ($^3\text{He}/^4\text{He}/\text{gr} : 6.8 \, \text{nm}^{-2}, ^3\text{He}/^3\text{He}/\text{gr} : 6.4 \, \text{nm}^{-2}$) because the exchange interactions are strongly depend on the density of solid. The difference of $J$ observed in the IC region is comparable to that in the 4/7 phase, and it suggests that the most probable factor which cause the difference of $J$ will be the different density of the second layer solid. In the case of the phase equilibrium of the second layer solid and the third layer fluid, we can write a following approximate expression:

$$\mu_S(\rho_S) = \mu_L(\rho_L) + \Delta u$$

(2)
where $\mu_S(\rho_S)$ and $\mu_L(\rho_L)$ are the chemical potentials of a solid and liquid phase in ideal 2D system at their densities of $\rho_S$ and $\rho_L$, $\Delta u$ is the potential difference between the second and third layer. $\mu_S$ and $\mu_L$ in $^3$He/$^4$He/gr (including the adsorption potential in each layer) are calculated in ref.[18]. If we change the first layer from $^3$He to $^4$He, the density of the first layer increases from 11.4 to 12.0 nm$^{-2}$. The benefit of the Lennard-Jones potential increases $\Delta u$ about an order of 1 K in $^3$He/$^4$He/gr. This difference of $\Delta u$ is especially important when $\rho_L$ is low where the density variation of $\mu_L$ is gentle. On the other hand, at high densities where $\rho_L$ is high, the effect of the difference of $\Delta u$ is relatively small because it corresponds to the small difference of $\rho_L$ for the large density variation.

The temperature dependence of entropy of the second layer solid are shown in Fig. 4. Now, we extrapolate $C \propto T$ behavior for $T < 0.3$ mK. Curiously, the total spin entropies deduced from our $C$ data are about 18% smaller than the expected value ($= N_2 k_B \ln 2$) in the IC region. We note that the $N_2$ used in this analysis is proved to be wrong in $^3$He/$^4$He/gr as discussed above, but the correction of the density is a opposite direction to explain this missing entropy. Although this is not fully understood yet, it could be because of the substrate heterogeneity effect, the existence of $C$ anomalies at much lower temperatures, or hybridization between the second layer solid and the third layer liquid $^3$He.

![Fig. 4](Color on-line) Spin entropies deduced from the $C$ of the second layer solid. Solid lines below 0.3 mK are the extrapolation of the $C \propto T$. The inset is the density dependence of total spin entropies.

3 Summary

From the heat-capacity measurements in wide temperature range, we obtained the MSE interactions of the IC region of the second layer for $^3$He/$^4$He/gr. The value of $J$
is significantly smaller than that for $^3\text{He}/^3\text{He}/\text{gr}$ at lowest densities of the IC region. From this results, we conclude that the density of the IC solid of the second layer is largely affected by the difference of the first layer potential. We have also shown the spin entropy of the IC solid. Strangely, the total spin entropy is much smaller than the expected value. The origin of this missing entropy is not understood yet.

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**References**