

論文の内容の要旨

論文題目 : Two-dimensional Quantum Phases of Helium Three on Graphite

(グラファイト上へリウム3の2次元量子相)

氏名 佐藤 大輔

A few layers of helium three (^3He) thin films adsorbed on an atomically flat graphite surface are ideal 2D Fermion systems with nuclear spin-1/2. Particularly, in the second layer, particle correlations can widely be changed from the dilute Fermi gas regime to the highly compressed solid by varying areal density of ^3He (ρ). As a result, a rich quantum phase diagram with various exotic quantum phases is proposed [1,2]. They include an anomalous quantum fluid phase [3], the gapless quantum spin-liquid (QSL) state in the low-density commensurate solid (the 4/7 phase) [2,4] and a ferromagnetic state in the high-density incommensurate solid [2,5]. However, detailed density evolutions of the various phases have not been explored until today.

In this thesis, after introduction to ^3He systems in two dimensions (2D) in Chap. 1 and 2 and describing experimental methods in Chap.3, results of a comprehensive heat-capacity measurement of the 2nd layer solid ^3He adsorbed on graphite preplated with a monolayer ^4He ($^3\text{He}/^4\text{He}/\text{gr}$) in a wide temperature ($0.1 \leq T \leq 80$ mK) and density ($6.8 \leq \rho \leq 16.5$ nm $^{-2}$) range are discussed in Chap. 4. Here, I could disclose how the gapless QSL evolves into a frustrated ferromagnet through a first-order transition (possibly a commensurate-incommensurate (C-IC) transition) including density evolutions of the multiple-spin exchange (MSE) interactions up to six-spin exchanges [6]. In Chap. 5, results of similar heat-capacity measurements at very low densities for the 1st to 4th layers are given. Here, I could answer a long-standing question if 2D ^3He systems self-condense or not. The answer is yes but with a surprisingly low critical liquid density (0.6-0.9 nm $^{-2}$) ever found. In Chap. 6, I show results of nuclear magnetic resonance (NMR) measurements on the second layer ^3He including the first experimental information on the spin dynamics of the gapless QSL.

Frustrated magnetism in the 2nd layer solid ^3He (Chap. 4)

As was first pointed out by Thouless [7] nuclear magnetism of solid ^3He can be frustrated by competition among the MSE interactions where exchanges of odd (even) number of atoms favour (anti-) ferromagnetism. This magnetic frustration manifests most exotically in the 4/7 phase as the gapless QSL state. We found that the 4/7 phase ($\rho = 6.8$ nm $^{-2}$) is stable against adding ^3He atoms up to 8.1 nm $^{-2}$. With increasing density further, the heat-capacity double peak of the 4/7 phase below 1 mK characteristic of the gapless QSL dramatically changes to the ferromagnetic single peak at 3 mK in the density region of $8.1 \leq \rho \leq 9.5$ nm $^{-2}$. Measured heat capacities in this region are consistent

with the two-phase coexistence model ($C = (1 - x)C_C + xC_{IC}$) between the 4/7 (C) and the incommensurate (IC) solid at $\rho = 9.5 \text{ nm}^{-2}$ (Fig. 1). This is the first thermodynamic evidence for this previously anticipated phase transition. In addition, a slightly positive deviation of the density evolution of x from the linear relation indicates that this would be somewhat more complicated by the domain-wall structure characteristic of the C-IC transition than the conventional two-phase coexistence.

Above 9.5 nm^{-2} , the ferromagnetic IC solid is uniformly compressed with increasing density. Figure 2 shows measured heat capacities in this region normalized by the number of ^3He atoms N and the effective exchange interaction J_c . The solid line in the figure is a theoretical calculation of the spin-1/2 Heisenberg ferromagnetic model on a 2D triangular lattice (HFT) [8]. The data at the highest density (14.45 nm^{-2}) are represented very well by this HFT model. With decreasing density, the data show a remarkable deviation from the model. This is because of increasing contributions from the higher-order exchanges such as four- and six-spin exchanges (J_4 and J_6) at lower densities. Since the ground state of the IC solid is known to be ferromagnetic [5], we call this phase a *frustrated ferromagnet*. The inset of Fig. 2 shows density variations of the MSE parameters, $-J = J_2 - 2J_3$, J_6/J_4 and $-K/J$, deduced by fitting our data to the high- T series expansion of heat capacity for the MSE Hamiltonian [9]. Here, J_n is the n -spin exchange interaction, and $K = J_4 - 2J_5$. The MSE parameters are determined here with much higher precisions than the earlier study [10].

2D self-condensation of ^3He on graphite (Chap. 5)

It is known that the attractive interatomic potential, repulsive zero-point energy and Fermi energy are severely countervailing each other in a 2D system of ^3He . Therefore, it has been a long-standing question whether self-condensation exists or not in this system. Previous theoretical calculations suggest the absence of the self-condensation [11]. So far existing experimental studies show also no signature of the 2D condensation except one experiment, which is a heat-capacity study of submonolayer ^3He floating on thin superfluid ^4He films [12]. However, the subsequent heat-capacity, third sound and NMR measurements gave contradictory results.

To answer this question, I made detailed heat-capacity measurements of low-density monolayer ^3He in the first four layers on graphite successively. At low enough temperatures ($T \ll T_F$), heat capacity of a 2D Fermi fluid should be $C = \gamma T$ ($\gamma = \pi k_B^2 m^* A / (3\hbar^2)$). Here m^* is effective mass of ^3He quasiparticle and A is surface area of the system. In 2D, γ is independent of the number of atoms, and depends only on m^* and A . Thus, if there exists a gas-liquid transition, γ will linearly decrease to zero with decreasing density. Measured γ values actually show such linear decreases at very low densities never explored before in the 2nd, 3rd [13] and 3rd + 4th layer ^3He adsorbed on a monolayer ^4He preplated graphite (coloured regions in Fig. 3). The heat capacity of the 1st layer ^3He adsorbed directly on a bare graphite surface also show a similar linear decrease of γ followed by an initial development of a spin heat-capacity contribution with a weak T -dependence from amorphous ^3He preferentially trapped on substrate heterogeneities. These four monolayer ^3He systems have extremely different confinement potential, phonon velocities in underlayers, and substrate heterogeneity effect each other. Nevertheless the 2D condensation (*puddling*) was observed with a similar critical liquid density ($0.6\text{-}0.9 \text{ nm}^{-2}$) below which a uniform liquid ^3He is unstable against the

gas-liquid phase separation. Thus, I conclude that the self-condensation of ^3He in 2D should be an intrinsic property. This provides a severe constrain for future theoretical many-body calculations for Fermions.

Pulsed-NMR studies of the 2nd layer ^3He (Chap. 6)

The spin-spin relaxation time T_2 of the 2nd layer ^3He adsorbed on a monolayer ^4He preplated graphite was measured in a wide temperature range ($0.1 \text{ mK} \leq T \leq 1.4 \text{ K}$). In the 4/7 phase, T_2 is T -independent in a wide range of $10 \leq T \leq 300 \text{ mK}$ where it is determined by the exchange interactions (*exchange plateau*), while below 10 mK T_2 decreases gradually with decreasing T down to 100 μK . This gradual T_2 shortening is suggestive of the growth of short-range spin ordering when the system undergoes a gapless QSL state without long-range ordering. This would be the first direct experimental information on spin dynamics of such an exotic magnetic system. The density dependence of T_2 at 100 mK shows a V-shaped minimum at the density of the 4/7 phase. These T - and ρ -dependences of T_2 are qualitatively consistent with the quantum phase diagram determined from our heat-capacity measurements. However, quantitative analyses of the T_2 data are difficult at this moment because of an observed unexpected magnetic-field (B) dependence of T_2 ($1/T_2 \propto B$) [14]. The origin of this field-dependence is probably microscopic magnetic field inhomogeneities due to a mosaic angle spread of the platelet of Grafoil substrate and large diamagnetism of graphite. Future NMR measurements in much lower fields will resolve this problem.

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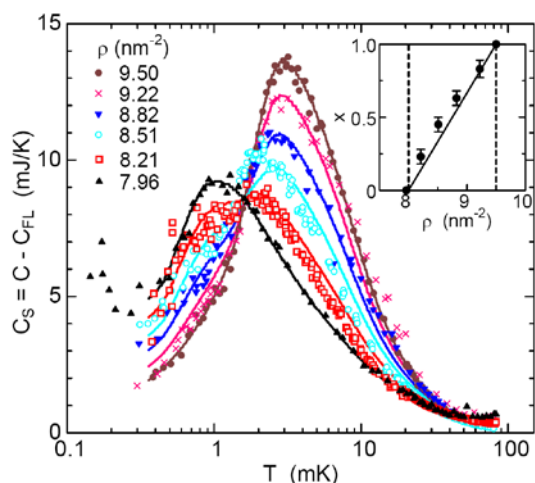


Fig. 1: Magnetic heat capacities of the 2nd-layer solid ^3He on graphite in the C-IC transition region. The solid lines are fittings to the conventional two-phase coexistence model. The inset shows an areal fraction of the C phase (x) obtained by the fittings.

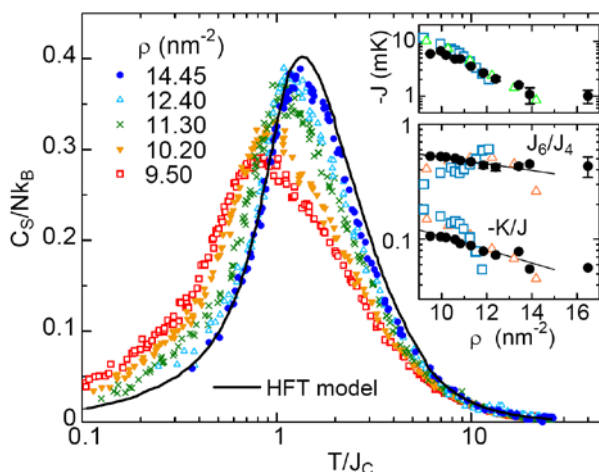


Fig. 2: Specific heats C/Nk_B as functions of T/J_c in the frustrated ferromagnetic phase of the 2nd-layer ^3He on graphite at various densities. The solid curve corresponds to a theoretical calculation of the spin-1/2 Heisenberg ferromagnetic model on a 2D triangular lattice [8]. The inset shows density variations of the multiple-spin exchange interactions. The filled circles are results in this work, and the others are those in earlier studies on pure ^3He films [10].

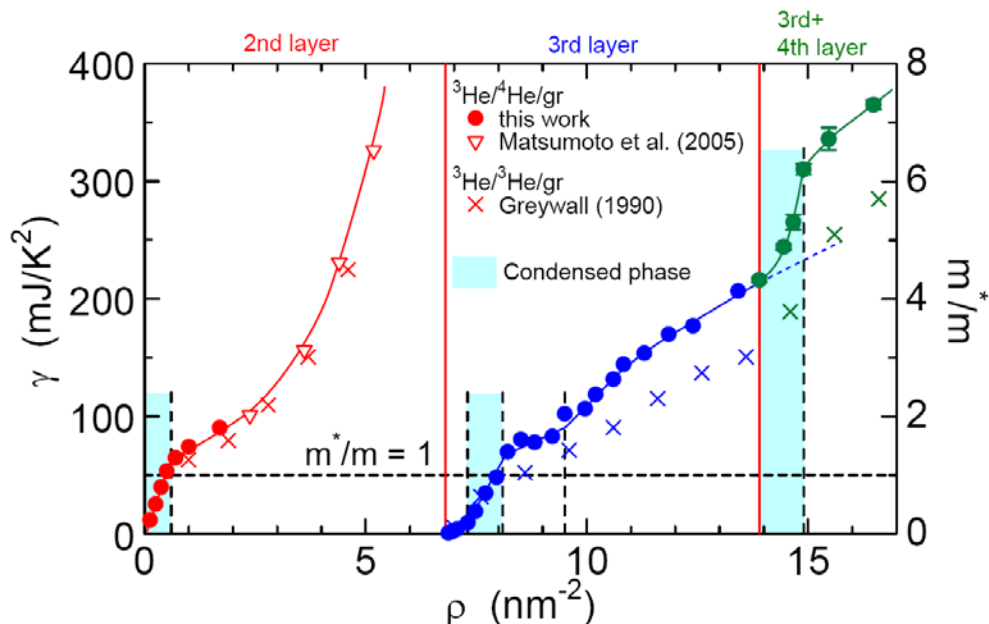


Fig. 3: Density variations of the γ coefficients in the 2nd, 3rd and 4th layers of ^3He on graphite. The filled circles (this work) and open triangles [3] are data for the $^3\text{He}/^4\text{He}/\text{gr}$ system. The crosses are for $^3\text{He}/^3\text{He}/\text{gr}$ [1]. The horizontal dashed line corresponds to the γ value of the ideal Fermi gas. The vertical solid line at 6.8 (13.9) nm^{-2} corresponds to promotion to the 3rd (4th) layer. The coloured regions are the self-condensed phase at each layer.