### 論文の内容の要旨

# 論文題目: Quantum Phase Diagram of Two-dimensional Helium on Graphite

(グラファイト上2次元ヘリウムの量子相図)

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The phase diagram of two atomic layers of helium four (<sup>4</sup>He) adsorbed on graphite was studied using an experimental method. Heat capacity and vapor pressure have been measured as a function of temperature for a series of coverage. The system <sup>4</sup>He on graphite comprises a pure Bose system in the presence of a periodic potential. The second layer, especially, is subjected to a weakly corrugated potential, and various phenomena are expected to arise: for example, reentrant superfluidity was observed only in the second layer [1]. However, in reality, the phase diagram of the second layer is still controversial. Although heat capacity peaks usually indicate phase transitions, peaks observed by a previous experiment were too obscure to construct a phase diagram [2]. This thesis describes improved measurements of heat capacity using high-quality graphite, ZYX, as a substrate. As ZYX comprises much larger crystals than Grafoil used in the previous measurement, the correlation length of films on ZYX was not overly suppressed. Vapor pressure was also measured to confirm phase boundaries determined from the heat capacity.

The important features observed to determine the phase boundaries are (a) a sub-step in vapor pressure isotherms at T = 1.75 K, (b) constant chemical potential of <sup>4</sup>He films for wide coverage region indicating gas-liquid coexistence, and (c) substrate-dependence of heat capacity peaks from comparison with previous study results employing Grafoil.

#### (a) Sub-step in vapor pressure isotherms

Vapor pressure of <sup>4</sup>He bilayer films was measured in the coverage range of 19.0 nm<sup>-2</sup> to 20.7 nm<sup>-2</sup> at temperatures less than 2.9 K. In the coverage range between 19.4 nm<sup>-2</sup> and 19.7 nm<sup>-2</sup>, the vapor

pressure at T = 1.75 K stays constant at  $2 \times 10^{-3}$  mbar. In an adsorption isotherm with axes of coverage and pressure, this behavior looks like a slight sub-step. Since the chemical potential of the vapor is equal to that of the films in thermal equilibrium, which is a function of pressure and temperature, the sub-step shows directly that the films have constant chemical potential throughout the coverage range. Therefore, distinct phases are realized at both ends of the coverage range,  $\rho = 19.4 \text{ nm}^{-2}$  and  $19.7 \text{ nm}^{-2}$ , coexisting within the sub-step. This coverage range overlaps that of the superfluidity observed in recent torsional oscillator experiments [3].

#### (b) Constant chemical potential indicating liquid-gas coexistence

The chemical potential of <sup>4</sup>He films at coverages less than 22.0 nm<sup>-2</sup> and that of a layer of <sup>3</sup>He on <sup>4</sup>He at coverages less than 8.0 nm<sup>-2</sup> were derived from the vapor pressure. The chemical potential of bilayer <sup>4</sup>He stays almost constant over the coverage range between 13.0 nm<sup>-2</sup> and 16.0 nm<sup>-2</sup>, while that of the <sup>3</sup>He-<sup>4</sup>He layered films gradually increases with the coverage in the corresponding coverage range. It suggests that two phases coexist in <sup>4</sup>He films, while <sup>3</sup>He films form a uniform phase [4]. Beyond 16.0 nm<sup>-2</sup>, the chemical potential of <sup>4</sup>He films starts to increase gradually with the coverage, which indicates compression of a uniform phase.

#### (c) Substrate-dependence of heat capacity peaks

Heat capacity of <sup>4</sup>He films was measured at temperature in the temperature range of 0.1–1.7 K for coverages between 2.5 nm<sup>-2</sup> and 23.0 nm<sup>-2</sup>. Comparing data after the system-size change is a useful tool for distinguishing ordinal transitions from a KT transition and one-particle phenomena; thus, all heat capacities obtained were compared with precision with those in the previous study using Grafoil as a substrate [2]. On the whole, various substrate-dependencies were observed.

In the coverage range between 13.0 nm<sup>-2</sup> and 16.0 nm<sup>-2</sup>, heat capacity peaks were clearly enhanced by using ZYX. A sharp kink was observed throughout the coverage range, compared to a rounded kink when using Grafoil [2]. The heat capacity diverges logarithmically around the peak temperature at 15.0 nm<sup>-2</sup>, where the maximum peak height was obtained. On the other hand, the peak temperature was independent of both substrate and coverage. Because the chemical potential stays constant over this coverage range, there is the coexistence of two phases, the gas and liquid phase.

Meanwhile, in the coverage range of 16.5 nm<sup>-2</sup> to 18.7 nm<sup>-2</sup>, the heat capacity data obtained by using ZYX and those with Grafoil look alike. Both have a bump, instead of a peak, which suggests that the characteristic length of the phenomenon, the cause of the bump, is shorter than the platelet size of Grafoil, namely 10–20 nm. Therefore, this bump does not indicate any long-range order, though it may indicate a quasi-long-range order that arises in a KT transition. In

this coverage range, the chemical potential indicates a uniform phase. It is probable that the <sup>4</sup>He films form a uniform fluid here.

In the coverage range of  $19.2~\text{nm}^{-2}$  to  $20.4~\text{nm}^{-2}$ , a broad peak was observed at  $\approx 1.4~\text{K}$ . By using ZYX, this peak was enhanced by a maximum of 15% and the peak temperature shifted by  $\approx 40~\text{mK}$  [2], thus indicating an ordinal phase transition. The peak amplitude increases linearly with the coverage in the coverage range between  $19.3~\text{nm}^{-2}$  and  $19.7~\text{nm}^{-2}$ , where the adsorption isotherm shows a sub-step. These facts are consistent with the assumption of two-phase coexistence, suggested by the sub-step. However, even though being enhanced, the peak was still broad. Though bilayer  $^3\text{He}$  behaved similarly to bilayer  $^4\text{He}$  in the relevant coverage range, no feature was observed for a layer of  $^3\text{He}$  on  $^4\text{He}$  in corresponding coverage range. The heat capacity of the  $^3\text{He}$ - $^4\text{He}$  layered films was dominated by the contribution from the mixing of the isotope atoms between layers. The energy scale for exchange of helium atoms between layers should be as large as the onset temperature of the contribution,  $\approx 0.4~\text{K}$ , which is much less than the peak temperature of pure isotope systems. From these observations, it can be concluded that the peak indicates a phenomenon of bilayers.

Also, higher coverages were examined, and a signature of the melting of an incommensurate solid and the gas-liquid transition of the third layer were observed; though the margin of error was large, owing to a high vapor pressure.

As a reference system, the first layer of <sup>4</sup>He films was also investigated. For the first layer, the corrugation in the substrate potential of graphite is strong enough to cause a structural order called  $\sqrt{3} \times \sqrt{3}$  registry. The registry is completed at the density of 6.366 nm<sup>-2</sup>, below which, the ordered solid covers a certain portion of the graphite surface. At coverages higher than 5 nm<sup>-2</sup>, a sharp peak observed at  $T \approx 3$  K indicates an order-disorder transition of the structure. Since the transition had been previously investigated using ZYX [5], the peak was measured this time to confirm the quality of our ZYX substrate. Strong enhancement of the peak height and a small peak-temperature shift by ≈ 20 mK were reproduced; furthermore, coverage-dependency of the peak was investigated in detail, and a strong anisotropy in the peak amplitude was observed: excess atoms easily destroy the peak while holes do not affect it much. As this anisotropy was not observed in the previous experiments with Grafoil [2], it suggests higher homogeneity in the ZYX surface. (The anisotropy is almost opposite to that of the second layer in the coverage range between 19.2 nm<sup>-2</sup> and 20.4 nm<sup>-2</sup>). At coverages less than 6 nm<sup>-2</sup>, an extra bump was observed at low temperatures in several experiments using different graphites [2,6], whose cause is still not explained. To study the bump, monolayer <sup>4</sup>He films in the coverage range between 2.5 nm<sup>-2</sup> and 5.5 nm<sup>-2</sup> were examined; this bump was clearly enhanced in amplitude, but the peak temperature depends significantly on coverage and the peak was still rounded. These features are different from other anomalies observed in the second layer.

The experimental results of the phase transitions were thus collected for <sup>4</sup>He films adsorbed on ZYX graphite. In summary of these observations, a new phase diagram of the second layer of <sup>4</sup>He on graphite is proposed, as shown in the figure below.

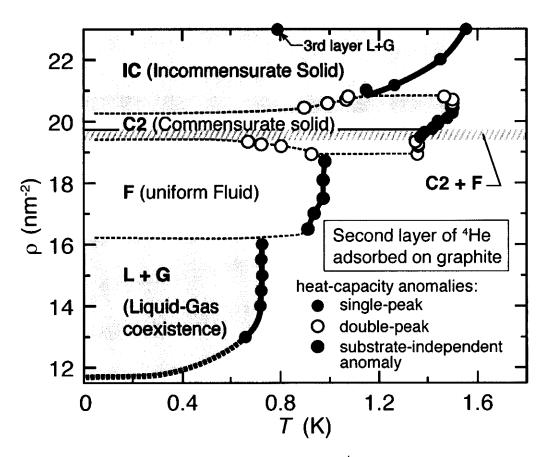


Figure: Proposed phase diagram of bilayer <sup>4</sup>He adsorbed on graphite.

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