Stability of 4/7 phase of He particles adsorbed on the graphite surface

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Abstract. We consider two layers of helium particles adsorbed on a graphite surface. The first layer of $^4$He particles is a solid phase with number density $12.0 \text{nm}^{-2}$. The second layer of $^3$He particles forms a 4/7 commensurate phase with its number density $6.9 \text{nm}^{-2}$. The stability of these two layers and the stable configuration of the 4/7 phase are studied by path integral Monte Carlo method. Possibility of mixing between the first layer particle($^4$He) and the second layer one($^3$He) is investigated, and we find that the mixing is prohibited in this stacking configuration of layers. Next a stable configuration is studied by operating a translation of the second layer against the first layer, and it is found that a proper translation stabilizes the system. The density profile of particles and thermodynamic quantities of the stable configuration are also computed.

1. Introduction
Many fascinating phenomena were discovered in the system of $^3$He particles adsorbed on the graphite. The structure phase diagram for temperature and particle density was given by Schick.[1] This system shows interesting character due to the large amplitude of a zero point motion and potential corrugations of the graphite substrate and of the adsorbed He particles. Magnetic property of the system strongly depends on the particle density of the system. Because the amplitudes of multi-particle exchanges are sensitive functions of the particle density. Furthermore, their amplitude of exchanges compete with each other. Therefore, this two dimensional system is quite suitable one to study the relation of spatial correlation and origin of magnetism in $^3$He particles.

In the two layer adsorbed system, a commensurate structure, 4/7 phase, is realized. Many anomalies in transport, magnetism and thermodynamic quantities are observed around number density of the 4/7 phase.[2,3] Recently, the experiment of the 4/7 phase are planned by H. Fukuyama group of Tokyo university; the system consists of the first layer of $^4$He and the second layer of $^3$He particles. The experiment aims to find the role of the purely second layer particle exchange. Stimulated by the plan we study the stability and properties of the system using a path integral quantum Monte Carlo(PIQMC) computation. The first we prepare this 4/7 phase constructed by $^3$He and $^4$He particles. A possibility of a mixing between $^3$He and $^4$He is investigated to check the stability of both layers. The stable relative configuration of the second layer for the first layer is searched. Melting temperature of the system is also calculated using a size effect of a Binder parameter in order to compare the result with that of the experimental data. In the last section, we devote ourselves into the discussion.
2. Hamiltonian of the system
In order to carry out the PIQMC we prepare the 4/7 phase. The adsorbed $^4\text{He}$ particles on the graphite surface form a solid phase as the first layer which has a number density $12.0 \text{nm}^{-2}$. Therefore, the same number density is adopted as a triangular lattice of the first layer. We add the $^3\text{He}$ particles to the system as the second layer of the 4/7 phase. Two dimensional periodic boundary condition is imposed on the directions of graphite plane. Hamiltonian of the system is given as

$$H = -\sum_{i=1}^{N} \frac{\hbar^2}{2m_i} \nabla^2_i + \sum_{i<j} V(r_{ij}) + \sum_{i=1}^{N} U(z_i),$$

(1)

where the notation $m_i$ stands for the mass of the $i$-th particle which is the mass of $^3\text{He}$ or $^4\text{He}$, and $N$ is the total number of $^3\text{He}$ and $^4\text{He}$ particles. The C-atom of the graphite is placed in the $xy$-plane and coordinate $z_i$ stands for the distance of the $z$-direction from the graphite plane, and He-particles are placed in the $0 < z$ region. Potential terms, $V(r_{ij})$ and $U(z_i)$, mean Aziz potential of the He-He interaction[4], and the potential He-C atoms of graphite[5], respectively. In the He-C interaction potential, we neglect the corrugation of the potential in $xy$-plane due to the first layer being incommensurate to the substrate lattice. We consider the structure of the system which has the order of the energy per particle $\sim 1\text{K}$, so the exchange energy per particle $\sim 1\text{mK}$ is negligible. We deal with both $^3\text{He}$ and $^4\text{He}$ particles as a distinguishable one. Hence no path exchange is performed in the PIQMC computation. The difference between $^3\text{He}$ and $^4\text{He}$ is just introduced by the mass of the particle in our computation. We adopt the density matrix which is readily attained by the matrix squaring method at the temperature 58.3K. During the computation we flip all the particle paths, including that of the first layer, globally and locally. The acceptance ration of the path flip is almost kept as 50% with changing the flipping amplitude.

3. Calculation result
We carry out the PIQMC for the 4/7 phase which has the total particle number $N = 44$ and 77. In the computation, first $4 \times 10^5$ Monte Carlo step(MCS) are discarded as the thermalizing process. It takes quite long MCS to reach the equilibrium state, because the potential corrugation of the first layer is very shallow. Since the system being in the equilibrium state, we measure thermodynamic quantities, density profiles of particle distribution and Binder parameter. Binder parameter is given for one dimensional system size $L$ [6],

$$G_L(T) = 1 - \frac{1}{3} \left\langle \left| \hat{\rho}(G_1) \right|^4 \right\rangle_L,$$

(2)

where $G_1$ stands for one of reciprocal vectors of the 2nd layer. Fourier amplitude $\hat{\rho}(G_1)$ of a particle density $\rho(r)$ is given,

$$\hat{\rho}(G_1) = \frac{1}{\Omega} \int \text{d}r \exp(-i \mathbf{G_1} \cdot \mathbf{r}) \rho(r),$$

(3)

where $\Omega$ is area of the system.

3.1. Stability of the layer
First we check the stability of the system. We put $^4\text{He}$ particles as the first layer and $^3\text{He}$ particles as the second layer, and run the simulation program. In the simulation a particle exchange between the first layer and the second layer by a whole path exchange is tried; it means a mass exchange of considering $^3\text{He}$ and $^4\text{He}$ particles. During the simulation no exchange is observed. Furthermore, we find no exchange in a system which has more second layer density.
than the 4/7 phase. The system with reversed configuration, $^3\text{He}$ being in the first layer and $^4\text{He}$ being in the second layer, is prepared in order to confirm the stable configuration. In this system the inter layer particle exchange occurs frequently. Finally all of the $^4\text{He}$ particles are demoted to the first layer. Hence the stability of the standard configuration is conformed. Number density distributions of both layers for $z-$direction is shown in Fig.1. The number densities are normalized as the integration of each layer for $z-$coordinate becomes $12.0\text{nm}^{-2}$ and $6.9\text{nm}^{-2}$, respectively.

![Figure 1. Particle density profiles, the 1st layer and the 2nd layer in z-direction.](image1)

The particle distribution of the first layer is localized comparing with that of the second layer. Particles in the first layer feel stronger adsorption potential from the graphite and have smaller zero point vibration amplitude due to the mass difference. Cross over of the two layers are quite small reflecting the small exchanging amplitude.

### 3.2. Stable configuration of the second layer

The 4/7 phase structure proposed by Elser has two types of sites, A-site and B-site, in the second layer.[7] The A-site is just above the particle of the first layer, and the B-site is on the saddle point of the second layer. Both sites are located on unstable position of the particle-particle potential of an inter layer. Therefore, we investigate the stable position of the second layer using a degree of freedom of a translation versus the first layer. We search the stable configuration of the 4/7 phase by PIQMC with using various initial states. Among the trial initial states it becomes most stable one that the A-site particle is in the lowest potential point. (See Fig. 2) The density profile in the graphite plane at $T = 0.73\text{K}$ is shown in Fig. 4. In this configuration the B-site is located near the saddle point of the potential. This configuration has lower energy, 1K per particle, than that of the original configuration.

![Figure 2. Stable configuration of the 4/7 phase.](image2)

### 3.3. Melting temperature

Measured temperature dependent Binder parameters of the system for lattice sizes $N = 44$ and $N = 77$ are shown in Fig. 4. Binder parameter takes a value $1/3$ and $2/3$ for a perfect liquid state and a perfect solid state, respectively. Melting temperature of the system is determined as a cross point of the curves by the size effect; melting temperature is evaluated 0.9K in our system. In the case of $N = 77$, error bars become large in the region $1\text{K} < T$. By analysing a
density profile of the system, fluctuation of domains between a liquid state and a solid state are observed in this temperature region.

Figure 3. Particle density profiles in $xy$-plane at $T = 0.73$K. Numbers at the frame stand for the coordinate of C-atoms.

Figure 4. Temperature dependent Binder parameters.

4. Summary and discussion
We studied the 4/7 phase on the graphite by PIQMC. The exchange of particles between two layers hardly occurs. Therefore, pure two dimensional exchange effect in the second layer should be observed in the experiment. The more stable configuration of second layer for the first layer is found. In this configuration the role of two kinds of site reverses against the previously proposed configuration by Elser. It should directly affect an amplitude of particle exchanges, and the magnetic properties should be changed. The computation of the exchange amplitude for each particle is the future problem. Melting temperature of the system was determined using the size effect of Binder parameter as 0.9K. Greywall observed a sharp peak in the specific heat curve which should be caused by solid-liquid phase transition around 1K in the 4/7 phase composed by pure $^4$He.[8] Our obtained melting temperature well agrees to that of empirical data. Analogous computations for various particles of the first layer and the second layer, $^3$He and $H_2$, are planned to confirm the effect of zero point vibration. Advance of the experiment for these system is highly expected.

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