Evidence for a Self-Bound Liquid State and the Commensurate-Incommensurate Coexistence in 2D $^3$He on Graphite

D. Sato · D. Tsuji · S. Takayoshi · K. Obata · T. Matsui · Hiroshi Fukuyama

Received: date / Accepted: date

Abstract We made heat-capacity measurements of two dimensional (2D) $^3$He adsorbed on graphite preplated with monolayer $^4$He in a wide temperature range (0.1 $\leq T \leq$ 80 mK) at densities higher than that for the 4/7 phase ($\rho = 6.8$ nm$^{-2}$). In the density range of $6.8 \leq \rho \leq 8.1$ nm$^{-2}$, the 4/7 phase is stable against additional $^3$He atoms up to 20% and they are promoted into the third layer. We found evidence that such promoted atoms form a self-bound 2D Fermi liquid with an approximate density of 1 nm$^{-2}$ from the measured density dependence of the $\gamma$-coefficient of heat capacity. We also show evidence for the first-order transition between the commensurate 4/7 phase and the ferromagnetic incommensurate phase in the second layer in the density range of $8.1 \leq \rho \leq 9.5$ nm$^{-2}$.

Keywords quantum fluid · quantum solid · two dimensional system · helium-3

PACS 67.30.ej · 64.70.F- · 67.80.dm · 64.70.Rh

1 Introduction

Monolayer $^3$He adsorbed on a graphite substrate is an ideal model system for studies of strongly interacting two-dimensional (2D) Fermions. In this system, we can vary areal density of Fermions over a wide range where various quantum phases are found [1, 2]. Especially, in the second layer, a commensurate (C) solid phase, “the so called 4/7 phase”, appears at the four sevenths density of that of the first layer [2], where the multiple spin exchange (MSE) interactions compete each other yielding strong magnetic frustration. It is believed that the magnetic ground state of this phase is the gapless quantum spin-liquid (QSL) [3, 4]. According to the first principles calculations based on the WKB approximation [5], the three-particle ring exchange becomes dominant with increasing density due to its less steric hindrance than the other exchange processes. Therefore, the magnetic ground state changes from the gapless QSL state to...
the frustrated ferromagnetic (FF) one with increasing density as was observed in the previous magnetization measurements in low fields [6,7]. Schiffer et al. [6] claimed the first-order transition between the two phases from the measured linear density dependence of $T = 0$ magnetization for the pure $^3$He system (hereafter, $^3$He/$^3$He/gr). The neutron scattering experiment [5] shows that, at high enough densities, the structure of the second layer is the incommensurate (IC) solid with a triangular lattice. However, the nature of the magnetic and structural transitions among those phases are not known in detail until now.

In this letter, we present results of heat-capacity measurements of 2D $^3$He adsorbed on graphite preplated with monolayer $^4$He (hereafter, $^3$He/$^4$He/gr) in a wide temperature range ($0.1 \leq T \leq 80$ mK) at higher densities beyond the 4/7 phase ($\rho \geq \rho_{4/7} = 6.8$ nm$^{-2}$). The results show clearly how the QSL-FF transition evolves as a function of increasing density.

2 Experimental

The surface area ($A$) of Grafoil (exfoliated graphite) substrate used here is 556 m$^2$. The first layer $^4$He of 12.09 nm$^{-2}$ was prepared at $T = 4.2$ K, and then $^3$He overlayers were made at 2.7 K. We assume almost complete isotropic stratification between the first and second layers.

The heat-capacity was measured with the relaxation method at $0.1 \leq T \leq 1$ mK and the adiabatic heat-pulse one at $T \geq 0.3$ mK. The temperature of the samples was determined by a platinum-pulsed NMR thermometer ($T \leq 30$ mK), and a carbon resistance one ($T \geq 20$ mK) which are calibrated by a $^3$He melting curve thermometer. Details of the experimental setup were described in the previous paper [9].

---

![Fig. 1](Color online) Heat capacities ($C$) of 2D $^3$He on graphite at four representative densities involving the commensurate 4/7 phase ($\rho = 6.79$ and 7.96 nm$^{-2}$), the C-IC coexistence region (8.82 nm$^{-2}$), and the high density bound of the coexistence region (9.50 nm$^{-2}$) for the second layer.
3 Results and Discussion

Fig. 1 shows the measured heat capacities ($C$) at four representative densities. The data can be classified into two distinct density regions. We discuss each region separately in the followings.

It is known that, in $^3$He/$^3$He/gr, the 4/7 phase ($\rho_{4/7} = 6.4$ nm$^{-2}$) has two broad $C$ peaks at 1.8 and 0.3 mK, which is characteristic of highly frustrated magnetic systems [3]. For the present system ($^3$He/$^3$He/gr), we also observed a similar double-peak structure in the 4/7 phase but at somewhat lower temperatures, i.e., 1.2 and around 0.2 mK (see Fig. 1). The peak-temperature differences are presumably caused by the different densities of the 4/7 phase with respect to the first layer ones.

With increasing density, the peak temperatures are unchanged until $8.1 \pm 0.1$ nm$^{-2}$, while the high-$T$ heat capacities increase progressively. This can be seen more clearly in Fig. 2 where $C$ isotherms at several fixed temperatures are shown. Excess heat capacities, $C_{FL} = C(\rho) - C(\rho_{4/7})$, can nicely be represented by the following functional form:

$$C_{FL} = \gamma T - \alpha T^2,$$

which is characteristic of degenerate Fermi fluid. In 2D systems, $\gamma = \pi k_B^2 m^* A/(3\hbar^2)$, and $\gamma$ depends only on the $^3$He effective mass $m^*$ and $A$. Therefore, we conclude that the 4/7 phase is stable up to $\rho \approx 8.1$ nm$^{-2}$ and the additional atoms are promoted into the third layer forming a degenerate 2D Fermi fluid in the temperature range we studied.
The $\gamma$-coefficients obtained in this way at densities below 10.3 $\text{nm}^{-2}$ are plotted as a function of the excess density ($\rho - \rho_4/7$) in Fig. 3. The most striking feature here is that the $\gamma$ values are smaller than that for the ideal Fermi gas spreading over the whole surface ($\gamma_0$) with $m^*/m = 1$. Moreover, $\gamma$ increases linearly with $\rho$ in the density region of $7.2 \leq \rho \leq 8.1$ $\text{nm}^{-2}$ and approaches $\gamma_0$ near 8.1 $\text{nm}^{-2}$. This indicates that the third-layer fluid forms a self-bound 2D Fermi liquid (or puddle) with $m^*/m = 1$. Moreover, $\gamma$ increases linearly with $\rho$ in the density region of $7.2 \leq \rho \leq 8.1$ $\text{nm}^{-2}$ and approaches $\gamma_0$ near 8.1 $\text{nm}^{-2}$. This indicates that the third-layer fluid forms a self-bound 2D Fermi liquid (or puddle) with a fixed density around 1.3 $\text{nm}^{-2}$ covering a limited area of the surface. The puddles cover the whole surface at $\rho \sim 8.1$ $\text{nm}^{-2}$ where the two phase coexistence between the C and IC solids starts (see below). Although similarly small $\gamma$ values were noticed in the previous heat capacity measurements in $^3\text{He}/^3\text{He}/\text{gr}$ [1, 10], they were considered as substrate heterogeneity effects not the puddle formation presumably because of the lack of data of the detailed density variations. Also, the present observation is different from the previous suggestion by Godfrin et al. [11] who assumed the puddle formation of about 4 $\text{nm}^{-2}$ at much higher densities than we observed.

It has been believed that the critical point does not exist, i.e., the absence of the gas-liquid transition, in 2D $^3\text{He}$ both theoretically [12] and experimentally [1]. However, the variational Monte Carlo calculation taking account of delocalization of the wave function perpendicular to the 2D plane [13] suggests the gas-liquid coexistence below about 2 $\text{nm}^{-2}$. It seems to be plausible that the third-layer $^3\text{He}$ on graphite can be self-bound due to the much smaller confinement potential than in the first- and second-layers.

Within a small excess-density regime below 0.5 $\text{nm}^{-2}$, the linear density dependence of $\gamma$ does not seem to hold (Fig. 3). The most plausible explanation for this is that the first 7% excess particles are somehow accommodated in the second layer and possibly in the third layer as well extending the area of the 4/7 phase. This explanation is consistent with small increases of the 0.5 and 1 mK isotherms in the corresponding density region (see Fig. 2). If this is the case, the self-bound density should be about 1.0 rather than 1.3 $\text{nm}^{-2}$. We don’t know, at present, how the particles are accommodated in the commensurate 4/7 phase; compressing with domain walls? or filling intrinsically existing vacancies?

In Fig. 3 we also plotted the $\gamma$ values at higher densities than 8.1 $\text{nm}^{-2}$. Here we fitted the raw $C$ data to the fitting function described below which is slightly different from eq. (1). One can see a clear kink at $8.1 \pm 0.1$ $\text{nm}^{-2}$, and the high density behavior seems to approach $\gamma_0$ at zero excess density. This is a further support for the present scenario since the slow increase of $\gamma$ above 8.1 $\text{nm}^{-2}$ should be due to increasing $m^*$, in the uniform 2D fluid. Note that, in general, a commensurate phase (the 4/7 phase) can coexist only with a puddle phase not with a compressible fluid where the chemical potential varies with density.

Above 8.1 $\text{nm}^{-2}$, the temperature dependence of $C$ dramatically changes. The 1 mK peak height rapidly decreases, while a new peak develops rapidly near 3 mK. This is clearly seen in Fig. 4 where the $C_{\text{FL}}$ terms have already been subtracted. We believe that this change is due to the magnetic QSL-FF transition associated with the structural C-IC transition within the second layer and that the 3 mK peak corresponds to the FF IC phase [14] from our previous magnetization measurements [15]. We fitted the data in this region to the following equation expected from the simple coexistence model:

$$C = (1 - x)C_C + xC_{\text{IC}} + C_{\text{FL}},$$

(2)
where $C_C$ and $C_{IC}$ are smoothed heat-capacities measured at $\rho = 7.96$ (C) and 9.50 nm$^{-2}$ (IC) without the Fermi fluid contributions $C_{FL}$, respectively. $x$ is the areal ratio of the IC to C solids in the second layer. The fittings are in good agreement with the experimental data as shown in Fig. 4, and $x$ increases linearly with increasing $\rho$ (see the inset). In addition, all the data cross each other roughly at a single point (1.6 mK, 8.6 mJ/K) as expected. Thus, the present results show unambiguously the first order transition between the gapless QSL (C phase) and the FF state (IC phase) in the density region between 8.1 and 9.5 nm$^{-2}$.

The high-density bound of the coexistence ($\approx 9.5$ nm$^{-2}$) is determined as a density above which the 3 mK peak starts to shift to lower temperatures. This can be seen as a slope change at the corresponding density in the C isotherm at 0.5 mK in Fig. 2. The shift is caused by lateral compression of the IC solid [14]. We note that the fittings to Eq. (2) give systematically larger heat capacities below 1 mK for the intermediate densities. Particularly at 8.21 nm$^{-2}$, the fitting quality is worse, for which we don’t know the reason now. Since low temperature magnetic properties of the 4/7 phase should be very sensitive even to small changes in competing interactions [2], the discrepancy might be due to a possible interlayer interaction between the third-layer fluid and the underlying 4/7 phase or to a domain wall structure in the C-IC transition.

4 Conclusion

From the heat capacity measurements down to 0.1 mK, we found that the commensurate 4/7 phase in the second layer on graphite is stable against additional particles up to 20% which are promoted into the third layer forming the low-density puddles of about 1 nm$^{-2}$. We also obtained clear evidence for the first-order transition between the 4/7 phase with the gapless QSL ground state and the ferromagnetic incommensurate phase.
Fig. 4 (Color online) Heat capacities of the second layer solid $^3$He on graphite in the C-IC coexistence region. The solid lines are fittings to eq. (2) with $x = 0, 0.17, 0.63, 0.83$, and 1 where $x$ is the fraction of the IC solid. Note that the fluid overlayer contributions $C_{FL}$ are already subtracted here. The inset shows the density variation of $x$.

Acknowledgements This work was financially supported by Grant-in-Aid for Scientific Research on Priority Areas (No. 17071002) from MEXT, Japan. D.S. acknowledges support from the Global COE Program “the Physical Sciences Frontier”, MEXT, Japan.

References

1. D.S. Greywall, Phys. Rev. B 41, 1842 (1990).
2. H. Fukuyama, J. Phys. Soc. Jpn. 77, 111013 (2008).
3. K. Ishida, M. Morishita, K. Yawata, H. Fukuyama, Phys. Rev. Lett. 79, 3451 (1997).
4. R. Masutomi, Y. Karaki, H. Ishimoto, Phys. Rev. Lett. 92, 025301 (2004).
5. M. Roger, Phys. Rev. B 30, 6432 (1984).
6. P. Schiffer, M.T. O’Keefe, D.D. Osheroff, H. Fukuyama, Phys. Rev. Lett. 71, 1403 (1993); J. Low Temp. Phys. 94, 489 (1994).
7. C. Bäuerle, Ph.D. thesis (University Joseph Fourier, 1996) in French.
8. H.J. Lauter, H.P. Schidberg, H. Godfrin, H. Wiechert, R. Haensel, Can. J. Phys. 65, 1435 (1987).
9. Y. Matsumoto, S. Murakawa, K. Honkura, C. Bäuerle, H. Kambara, H. Fukuyama, Physica B 329-333, 146-147 (2003).
10. M. Siqueira, J. Nyék, B. Cowan, J. Saunders, Phys. Rev. Lett. 78, 2600 (1997).
11. H. Godfrin, R.E. Rapp, K.-D. Morhard, J. Bossy, C. Bäuerle, Phys. Rev. B 49, 12377 (1994).
12. M.D. Miller, L.H. Nosanow, J. Low Temp. Phys. 32, 145 (1978).
13. B. Brami, F. Joly, C. Lhuillier, J. Low Temp. Phys. 94, 63 (1994).
14. D. Sato, S. Takayoshi, K. Obata, T. Matsui, H. Fukuyama, this conference.
15. S. Murakawa, H. Akiyoshi, Y. Matsumoto, D. Tsuji, K. Mukai, H. Kambara, H. Fukuyama, Proc. 24th Int. Conf. Low Temperature Physics (LT24), AIP Conf. Proc. 850, 311(2006).