Heat Capacity of Kagome $^3$He Monolayer Film on Graphite

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Abstract. The heat capacities of second layer $^3$He-$^4$He mixture solid films are measured. The total areal densities are adjusted to be the 4/7 phase commensurate to the underlying $^4$He first layer, and the fractions of $^3$He in the second layer are adjusted to be almost three-fourths of the amount of second-layer helium atoms. According to the recent theoretical prediction of the adsorption structure of the 4/7 phase by Takagi, $^3$He atoms in such a $^3$He-$^4$He mixture film are expected to be a nuclear spin system of the pure kagome lattice. The heat capacity measurements of such $^3$He-$^4$He films between 0.1 and 80 mK reveal double peak structures. The higher-temperature peaks appear at around 30 mK; these are thought to arise from the replacement of $^3$He and $^4$He atoms. The magnitude of the higher-temperature peak strongly supports the adsorption structure proposed by Takagi, and not the one previously proposed by Elser, and gives strong supporting evidence of the realization of a pure kagome $^3$He lattice. The lower-temperature peaks must arise from the short-range spin ordering of $^3$He nuclear spins on the kagome lattice. As compared to the heat capacity of the pure $^3$He in the 4/7 phase, i.e., in the triangular phase, slower short-range ordering is observed in the kagome lattice, which agrees with the theoretical predictions.

1. Introduction
Researchers have actively pursued the investigation of quantum disordered ground states. Many geometrically frustrated magnets have been investigated, and spin liquid states have been proposed as the ground state in some of them. Antiferromagnetic spins on both triangular lattices and kagome lattices are typical two-dimensional geometrically frustrated magnets. Although they have some different properties, specific characteristics due to the differences in the lattice structure have not yet been observed clearly.

The 4/7 phase of the second adsorbed $^3$He layer on graphite is one of the most typical antiferromagnetic quantum spins on a triangular lattice [1]. V. Elser suggested the formation of an effective kagome lattice of $^3$He nuclear spins [2] to explain the missing entropy problem reported by Greywall and Busch [3]. He also proposed the adsorption structure of the 4/7 phase as follows—one fourth of the $^3$He atoms adsorb on just above the first layer $^4$He atoms, and three-fourths adsorb on saddle points between adjacent dimples in the first layer [2]. Although later experimental measurements revealed that an effective kagome lattice is not essential to the missing entropy problem [5], the adsorption structure proposed by Elser has been considered to be valid for a long time.

Recent theoretical calculations by T. Takagi have revealed that one-fourth of the second
layer $^3$He atoms in the 4/7 phase adsorb on stable sites ($B'$-sites), as shown in figure 1 [4]; in contrast, Elser had proposed a different adsorption structure wherein three-fourths of the $^4$He atoms adsorb on stable sites. This leads to the possibility of a realizing a $S = 1/2$ quantum spin system on a pure kagome lattice by substituting $^4$He for one-fourth of the second layer $^3$He in the 4/7 phase, because a $^4$He atom has smaller zero point energy than a $^3$He atom and the $B'$-sites should be occupied by $^4$He atoms at low temperature. This paper reports the results of the heat capacity measurements of such $^3$He-$^4$He films between 0.1 and 80 mK.

2. Experimental
Grafoil sheets having an estimated surface area of 420 m$^2$ are used as graphite substrates. Three $^3$He-$^4$He films were prepared. The first layer comprises of pure $^4$He, whose areal density is believed to be 12 nm$^{-2}$. In the second layer, the amount of $^4$He atoms was fixed to just one-fourth of that of second layer atoms in the 4/7 phase, while the fractions of $^3$He atoms were 0.72, 0.75, and 0.78 times the amount of second layer atoms in the 4/7 phase. Other experimental setups and techniques are similar to those in our previous studies [5].

3. Results and Discussion
The results of the heat capacity measurements of the $^3$He-$^4$He solid films are shown in figure 2. Double peak structures can be observed, although there exists a $^3$He areal density variation. The magnitude of the exchange interaction in these $^3$He-$^4$He solid films is believed to be similar to that in the 4/7 phase, i.e., of the order of several mK. Therefore, the lower-temperature peak must arise from the spin short-range ordering. The origin of the higher-temperature peak is not trivial. The entropy changes calculated from the measured heat capacity exceed the expected overall change in the spin entropy within the measured temperature range. Therefore, it is evident that the higher temperature peaks originate from a source other than spin system.

Similar double peak structures have been observed in the pure $^3$He second-layer films on the $^4$He first layer adsorbed on graphite [6]. However, it has been reported that the magnitude of the higher-temperature peak decreases with increasing areal density and that this peak disappears entirely at the areal density of the 4/7 phase. The higher-temperature peak is believed to be arise from the zero point vacancies. The areal densities of the $^3$He-$^4$He films investigated in this study are similar to that of the 4/7 phase, and the higher-temperature peak due to zero point vacancies should not appear.

Another possible explanation is the replacement of the $^3$He and $^4$He atoms within the second layer. At sufficiently low temperatures, the adsorption structure shown in figure 1 is expected. With an increase in the temperature, the $^4$He and $^4$He atoms can change their positions due to thermal excitations. The calculated heat capacity of such a replacement, assuming that the replacement sites are independent, is indicated in figure 2 by the dotted line. Here, the
Figure 2. Heat capacity of three $^3$He-$^4$He films. The fractions of $^3$He and $^4$He atoms to the number of sites of the 4/7 phase are shown in the figure. The solid lines serve as visual guides. The dotted line and the dash-dot line indicate the calculated heat capacity of replacement of the $^3$He and $^4$He atoms in the adsorption structures proposed by Takagi [4] and Elser [2], respectively; note that the interactions are neglected here. The broken line indicates the calculated result assuming an interaction between the replacement sites, as described in the text for the case of Takagi’s structure.

fitting parameter is only the energy per replacement. The peak of the calculated heat capacity is too sharp to reproduce the measured results. However, the entropy change calculated from the measured higher-temperature peak of the heat capacity agrees with the expected overall entropy change of the replacement, $k_B N_3 (8 \ln 2 - 3 \ln 3)/3$, if the heat capacity is extrapolated to higher temperatures assuming a single exponent behavior. Furthermore, the calculated heat capacity can be broadened by the assumed interactions between replacement sites. In fact, the calculated heat capacity assuming the energy of replacement as $\epsilon_0 (M - \gamma M^{1/2})$ almost reproduces the higher-temperature peak, as indicated in figure 2 by the broken line, except at the highest temperature regime where the interaction between the replacement sites must be considered in greater detail. Here, $\epsilon_0$ is the energy of an independent replacement; $M$ the number of replacements; and $\gamma$ a factor. They strongly support that the higher-temperature peak arises from the replacement of $^3$He and $^4$He atoms.

Even if the adsorption structure proposed by Elser is valid, similar replacements can occur, and a heat capacity peak must appear. However, the heat capacity of replacement assuming the adsorption structure proposed by Elser is very small and it cannot reproduce the observed peaks as indicated in figure 2 by the dash-dot line. This is because the maximum number of replacements is limited to one-fourth the number of $^4$He atoms in Elser’s adsorption structure, while it is three-fourths in the adsorption structure proposed by Takagi. This strongly supports the adsorption structure proposed by Takagi and invalidates Elser’s adsorption structure. Furthermore, this implies that the realization of a quantum spin system on a pure kagome lattice in the $^3$He-$^4$He solid film is quite possible.

The double peak structure of the heat capacity implies that the pure kagome lattice in the $^3$He-$^4$He solid film is realized only in the low-temperature regime where the replacements of $^3$He and $^4$He atoms can be neglected. Fortunately, the two peaks are well separated. Therefore, the spin heat capacity behavior can be obtained with sufficient accuracy in a wide temperature range by subtracting the replacement contribution, although it is difficult to accurately estimate the heat capacity of the replacement. The spin heat capacity of the $^3$He-$^4$He film, i.e., of the spins on the kagome lattice, obtained by subtracting the heat capacity of the replacement calculated above is shown in figure 3. Note that it is plotted on a semi-logarithmic scale. The heat capacity of the pure $^3$He second layer on the $^4$He first layer measured by Matsumoto et al. [6] is also indicated by the broken line as the heat capacity of the triangular lattice. Here, their measured values are normalized by the surface area of the graphite substrate of this study and multiplied by 3/4 to equalize the number of $^3$He atoms. Broad peaks observed in the $^3$He-$^4$He films suggest that the effective exchange interaction is antiferromagnetic even in the kagome lattice. The
Figure 3. Spin heat capacity of three $^3$He-$^4$He films. Fractions of $^3$He and $^4$He atoms to the number of sites of the 4/7 phase are shown in the figure. The solid lines serve as visual guide. The broken line indicates the measured heat capacity of the pure $^3$He 4/7 phase by Matsumoto et al. [6]; this is normalized by the surface area of this study and multiplied by 0.75.

The magnitude of the exchange interaction is not very different, although a weaker hindrance of the $^4$He atoms in the exchange paths of neighboring atoms due to a smaller zero point motion may increase the exchange interaction. A detailed examination reveals that a smaller heat capacity exists in the kagome lattice above 1 mK. This implies that the spin short range ordering is slower in the kagome lattice. These behaviors agree with the theoretical predictions [2, 7]. The double peak structure predicted in the theoretical calculations for the kagome lattice [2, 7] has not been observed in the $^3$He-$^4$He films. Theoretical calculations have not taken into consideration the multiple spin exchange interactions. Their competition might push down the second peak to a lower temperature.

4. Conclusions
The heat capacities of the $\sim 75\%$ $^3$He-$^4$He solid films of the 4/7 phase exhibit a double peak structure. The higher-temperature peak can be interpreted due to the replacements of the $^3$He and $^4$He atoms. This strongly supports the adsorption structure recently proposed by Takagi, and also strongly suggests the realization of a quantum spin system on the kagome lattice. The behaviors of the spin heat capacity of $^3$He in the kagome lattice partially agree with the theoretical predictions.

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