Magnetic susceptibility and saturation magnetic field in the $t$-$J_2$-$J_3$-$K$ model: $^3$He on graphite

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Abstract. We consider dynamics of zero-point vacancies introduced in the two-dimensional (2D) solid $^3$He adsorbed on a graphite surface. In our previous paper, we calculated the temperature dependence of the heat capacity, single-particle spectra and effective mass, as well as the spin and density excitation spectra of the $t$-$J$-$K$ model, and discussed their experimental implications. As an extension of this model, we here propose an improved model, i.e., the $t$-$J_2$-$J_3$-$K$ model, which takes into account the proper cut of the ring-exchange paths by the zero-point vacancies, and we discuss doping dependence of the magnetic susceptibility and saturation magnetic field in the 2D $^3$He. We thus show that the improved model is essential in particular for considering the magnetic field dependence of the system.

1. Introduction
A system of $^3$He atoms adsorbed on a graphite surface is known to be an ideal two-dimensional correlated spin-1/2 fermion system [1]. A solidified commensurate phase of $^3$He atoms is stabilized at a 4/7 density of the underlying layer of $^4$He atoms, i.e., at the density $\rho = \rho_{4/7}$, and a triangular lattice of $^3$He is formed. Multiple-spin exchange (MSE) interactions become important in this system due to the hard-core repulsion between $^3$He atoms [2, 3]. Theoretically, the 4/7 phase has been studied using the triangular-lattice MSE model and the $uuud$ magnetisation plateau at half of the saturation magnetisation $M = M_s/2$ has been predicted by use of this model [4, 5], where the four-spin exchange interactions are taken into account. Recently, the $M = M_s/2$ plateau has been observed experimentally [6], which indicates however that the plateau is narrower than a theoretical prediction. Then, introduction of the higher-order exchange interactions may be required to reproduce the experimental result more quantitatively within the framework of the MSE model [7].

The doped Mott region of the monolayer $^3$He also shows interesting phenomena such as the double-peak structure in the temperature dependence of the heat capacity, which has been considered to be an experimental evidence of the presence of zero-point vacancies [8]. Recently, the triangular-lattice $t$-$J$-$K$ model has been proposed as a natural extension of the MSE model under the introduction of zero-point vacancies and the ground-state phase diagram in the parameter space is obtained [9]. Then, we have demonstrated [10] that the double-peak structure of the heat capacity is actually reproduced by the $t$-$J$-$K$ model in a parameter space where the spin degrees of freedom are highly frustrated. This is an indication of the clear separation in the energy scales between spin and density excitations [10]. Formation and stability of the 4/7 phase have also been discussed using an extended Hubbard model [11]. However, theoretical
models that explain the experimental results for the monolayer $^3$He in the doped Mott region consistently have not yet been established.

In this paper, we will propose the triangular-lattice $t$-$J_2$-$J_3$-$K$ model for the doped Mott region of the monolayer $^3$He. We will then calculate the uniform magnetic susceptibility and magnetisation curve and show that the introduction of this model is essential in particular for considering the magnetic properties of the monolayer $^3$He.

2. Model and method

The triangular-lattice $t$-$J_2$-$J_3$-$K$ model we propose is defined by the Hamiltonian

$$
\mathcal{H} = -t \sum_{\langle ij \rangle} (\hat{c}_{i \sigma}^\dagger \hat{c}_{j \sigma} + \text{H.c.}) + J_2 \sum_{\langle ij \rangle} \left( \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} \right) + J_3 \sum_{\langle ijk \rangle} (P_3 + P_3^{-1}) + K \sum_{\langle ijkl \rangle} (P_4 + P_4^{-1}) - h \sum_i S_i^z
$$

(1)

where $\hat{c}_{i \sigma} = c_{i \sigma}(1 - n_{i,-\sigma})$ is the projected annihilation operator of a fermionic particle ($^3$He atom) at $i$-site and spin $\sigma(=\uparrow, \downarrow)$ allowing no doubly occupied sites, which reflects the hard-core repulsion between $^3$He atoms. $\mathbf{S}_i$ is the spin-1/2 operator. $P_3$ ($P_4$) is the cyclic permutation operator of particles on $i, j, k$ ($i, j, k, l$) sites. These operators are defined as $P_3 = P_{ik} P_{ij}$ and $P_4 = P_{ik} P_{kj} P_{ij}$, where $P_{ij} = P_{ij} = \frac{1}{2}(\sigma_i \sigma_j + 1)$ is the two-particle permutation operator with the Pauli-spin matrix $\sigma_i$. The signs of the exchange parameters are $J_2 > 0, J_3 < 0$ and $K > 0$, as are determined from the number of permutations of fermions [2]. The summation $\sum_{\langle ij \rangle}$ ($\sum_{\langle ijk \rangle}$, $\sum_{\langle ijkl \rangle}$) in the second (third, fourth) term of Eq. (1) is done when all of $i, j$ ($i, j, k$, $i, j, k, l$) sites are occupied by particles. The last term $-h \sum_i S_i^z$ is the Zeeman energy under the uniform magnetic field $h$. We define the filling $n$ of particles as $n = N/L$, where $N$ is the total number of particles and $L$ is the total number of the lattice sites of the system.

Here, let us refer to the relation between the $t$-$J_2$-$J_3$-$K$ and MSE models. The second term of Eq. (1) can be written as $J_2 \sum_{\langle ij \rangle} \left( \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} \right) = \frac{J_2}{2} \sum_{\langle ij \rangle} P_2$. So, our definition of $J_2$ is twice as large as that of the MSE model. In the absence of vacancies, the $t$-$J_2$-$J_3$-$K$ model is identical, apart from a constant term, with the MSE model defined by the Hamiltonian $\mathcal{H} = \frac{J_2}{2} \sum_{\langle ij \rangle} P_2 + K \sum_{\langle ijkl \rangle} (P_4 + P_4^{-1})$ with the relation $J = J_2 + 4J_3$.

The reason we treat $J_3$ term independently is the following (also see Fig. 1). At half filling, the $t$-$J$-$K$ and $t$-$J_2$-$J_3$-$K$ models are both reduced to the MSE model with the relation $J = J_2 + 4J_3$ as mentioned above. When a vacancy is introduced into a triangle, the $t$-$J$-$K$ model gives the ferromagnetic interaction between two spins. However, since the three-spin ferromagnetic interaction no longer exists, the interaction between the two spins must be antiferromagnetic. Thus, we should treat the $J_3$ term independently.

We use the Lanczos exact-diagonalization technique on small clusters to study the magnetic properties of this model. In particular, we calculate the magnetisation curve $M(h)$, the temperature dependence of the zero-field magnetic susceptibility $\chi(T) \mid_{h=0}$ and the saturation magnetic field $h_s = h \mid_{M=M_s}$, where $M_s$ is the saturation magnetisation. Throughout the paper, we use $t = 1$ as the unit of energy and show the results on a parameter region $J = J_2 + 4J_3 = -0.3$, $K \leq 0.1$, where the spin degrees of freedom are highly frustrated.

3. Results of calculation

Calculated results for the temperature dependence of the magnetic susceptibility $\chi(T)$ are shown in Fig. 2. We assume $|J_3/J_2| = 1.5$ [12] for the $t$-$J_2$-$J_3$-$K$ model. One may confirm in Fig. 2 that, when vacancies are introduced into the MSE model, the magnetic susceptibility of the $t$-$J$-$K$ model is larger than that of the $t$-$J_2$-$J_3$-$K$ model. This behaviour can be understood as follows:
Figure 1. Schematic image of the difference between the t-J-K and t-J2-J3-K models. On one hand, the $J_3$ term is easily cut by the introduction of the vacancies than the $J_2$ term is, so that the t-J2-J3-K model shows more antiferromagnetic behaviour. The t-J-K model, on the other hand, shows more ferromagnetic behaviour because the $K$ term is easily cut by the vacancies rather than the $J$ term is. Although finite-size scaling analysis is not possible in the present method, we believe that the behaviours observed by our small-cluster studies should be retained in the infinite-size systems.

Calculated results for the magnetisation curve $M(h)$ are shown in Fig. 3. We assume

Figure 3. Calculated magnetisation curves $M(h)$ for the t-J-K (dotted line) and t-J2-J3-K (solid line) models. We use the $L = 20$ cluster with two vacancies ($n = 0.9$).

Figure 2. Calculated temperature dependence of the magnetic susceptibility $\chi(T)$ where $C$ is the Curie constant. We use the $L = 12$ cluster without vacancies ($n = 1.0$) and with two vacancies ($n = 0.83$). The straight dotted line indicates the Curie’s law.

Figure 4. Four-spin exchange parameter $K$ dependence of the saturation magnetic field $h_s$ for the t-J-K and t-J2-J3-K models. For the t-J2-J3-K model, four curves are almost indistinguishable. We use the $L = 20$ cluster without vacancies ($n = 1.0$) and with two vacancies ($n = 0.9$).
$|J_3/J_2| = 1.5$. The shapes of magnetisation curves are not very different between the $t$-$J$-$K$ and $t$-$J_2$-$J_3$-$K$ models. In other words, independent treatment of the $J_3$ term simply shifts these curves upward but will not drastically change the structure of energy levels near half filling.

Calculated results for the saturation magnetic field $h_s$ are shown in Fig. 4. Some sets of the parameter values of $J_2$ and $J_3$ that satisfy the relation $J_2 + 4J_3 = -0.3$ with $1 \leq |J_3/J_2| \leq 2$ are used for calculations of the $t$-$J_2$-$J_3$-$K$ model. We find that, in contrast to the $t$-$J$-$K$ model, the $t$-$J_2$-$J_3$-$K$ model is rather hard to be spin-polarized because the effective ferromagnetic two-spin interactions $J = J_2 + 4J_3$ are cut around the vacancies in the $t$-$J_2$-$J_3$-$K$ model. However, the results of the $t$-$J_2$-$J_3$-$K$ model with $J = J_2 + 4J_3 = -0.3$ little depend on the ratio $|J_3/J_2|$. This is because the effective two-spin interaction $J$ works on most of the exchange bonds distant from the vacancies, as long as the doping rate of vacancies is small. Thus, the ratio $|J_3/J_2|$ only slightly affect the results. One may also confirm that the four-spin interaction ($K$) dependence of $h_s$ for the $t$-$J$-$K$ and $t$-$J_2$-$J_3$-$K$ models are weaker than that of the MSE model. This is because the four-spin exchange interactions are suppressed by vacancies.

4. Summary
We have considered the dynamics of zero-point vacancies introducing into the 2D solid $^3$He adsorbed on a graphite surface. We have pointed out that the $J_3$ term should be treated independently to take into account the proper cut of the ring-exchange interactions, whereby we have proposed the $t$-$J_2$-$J_3$-$K$ model for the doped monolayer $^3$He system. We have used the exact-diagonalization technique on small clusters and studied the magnetic properties of the $t$-$J_2$-$J_3$-$K$ model in order to consider the density dependence of the magnetic properties of the monolayer $^3$He. We have shown that the calculated results for the magnetic susceptibility, saturation magnetic field and magnetisation curve of the $t$-$J_2$-$J_3$-$K$ model are consistently indicative of the enhancement of the antiferromagnetic interactions, which are, in contrast to those for the $t$-$J$-$K$ model, qualitatively consistent with the experimental results [13]. Thus, the suppression of the ring-exchange interactions caused by the vacancies is an essential ingredient in the explanation of the experimental magnetic properties of the monolayer $^3$He.

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