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論 文 要 旨

THESIS SUMMARY

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学籍番号： Student ID Number	13D02027	指導教員（主）： Academic Advisor(main)	田中秀数　教授
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Theoretical consensus on S = 1/2 Heisenberg kagome lattice antiferromagnet (KLAF) is that the ground state is disorder owing to the interplay of strong frustration and quantum fluctuation. However, the nature of the disordered ground state has been under debate. Recent theory on quantum KLAFs with bond randomness predicts that when the randomness exceeds a critical value, the ground state changes from the disordered state to a gapless spin-liquid-like state. Motivated by these theoretical debate and predictions, I investigate the magnetic properties of $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$, which is a mixture of two S = 1/2 KLAFs: $Rb_2Cu_3SnF_{12}$ with gapped ground state and $Cs_2Cu_3SnF_{12}$ with ordered ground state.

The most important point to obtain reliable experimental results is to prepare high-quality single crystals. I first grow single crystals of two parent systems $Rb_2Cu_3SnF_{12}$ and $Cs_2Cu_3SnF_{12}$ from a melting method, using platinum tubes as crucibles. Mixing stoichiometric amount of these parent crystals, I prepare sizable single crystals of $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ with various cesium concentration x from the melting method. Details of the sample preparation are described in section 2.2. The homogeneity of crystals was confirmed by X-ray diffraction.

Using single crystals obtained, I have performed magnetization, thermodynamic and μSR measurements. The magnetic susceptibilities for $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ shows systematic change with x, i.e., the magnetic susceptibility is not given by the superposition of those for $Rb_2Cu_3SnF_{12}$ and $Cs_2Cu_3SnF_{12}$, which confirms the homogeneity of crystals. From high-temperature magnetic susceptibility data for $T \geq 60$ K, I evaluated the four kinds of exchange interaction J_i (i = 1 – 4), using the exact diagonalization for the 12-kagome cluster. It was found that all the exchange interactions approach a uniform value for $x \to 1$, as shown in subsection 3.1.1. The substitution between rubidium and cesium ions will also create exchange randomness. Therefore, it is considered that the magnitude of exchange randomness increases with decreasing x from $x = 0$.

From the low-temperature magnetic susceptibility measurements, I observed that with increasing cesium concentration x, $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ undergoes the quantum phase transition from the disordered state to the ordered state at $x_c = 0.53$. I also found that the ground state for $0 < x \leq 0.53$ has a finite magnetic susceptibility, nevertheless the ground state exhibits no long-range magnetic ordering. For $0 < x \leq 0.53$, the excitation gap decreases with increasing x, and vanishes at the critical point $x_c = 0.53$. The absence of the magnetic ordering was confirmed via the μSR measurements conducted down to 0.3 K on the sample with critical concentration $x_c = 0.53$. Thus, the ground

state for $0 < x \leq 0.53$ is just like a spin liquid. I discussed this ground state nature according to recent theory that investigate the ground state for $S = 1/2$ Heisenberg KLAF with exchange randomness. The theory predicts that with increasing the magnitude of randomness, the ground state changes from quantum disordered state to a gapless spin-liquid-like state, which is described as valence-bond-glass (VBG). The VBG state is a state in which tightly bound spin singlets are localized on stronger bonds, while loosely bound singlets are situated on weaker bonds, as shown in Fig. 1.9. The characteristic properties associated to the VBG are as follows: (1) magnetic excitation is gapless, (2) long-range magnetic ordering is absent, and (3) low-temperature specific heat is in proportion to temperature. The first two theoretical results are consistent with our experimental results for the ground state for $0 < x \leq 0.53$, though I failed to observe the T – linear specific heat at low temperature, because the exchange interaction is rather large. Therefore, I concluded that the ground state in $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ with $0 < x \leq 0.53$ is the VBG state. This is the first clear report on the ground state of VBG with random kagome lattice.

Experimental magnetic susceptibility for $0 < x \leq 0.53$ shown in Fig. 3.5 displays small Curie term, though theoretical magnetic susceptibility for $S = 1/2$ random bond Heisenberg KLAF exhibits fairly large Curie term in the gapless spin-liquid-like state. I consider that this discrepancy arises from the finite size effect in calculation. The theoretical magnetic susceptibility was calculated, using exact diagonalization for up to 30-site clusters. Therefore, unpaired spins created on the boundary cannot form spin singlet, which leads to the Curie term in magnetic susceptibility. These unpaired spins will form any singlet when the system size is increased, so that the Curie term decreases.

For $Cs_2LiMn_3F_{12}$, which is described as S= 2 KLAF, I have investigated the magnetic properties via the magnetic susceptibility, specific heat and neutron scattering. I succeeded in synthesizing purified and sizable single crystals of $Cs_2LiMn_3F_{12}$. I confirmed the crystallographic axis using x-ray diffractometer for the first time. I measured the magnetic measurements and specific heat for $H \parallel c$ and $H \perp c$ using single crystal sample. It was found that $Cs_2LiMn_3F_{12}$ exhibits the antiferromagnetic order at $T_N \simeq 2.1$ K in both susceptibility and heat capacity measurements. I analyzed the results of magnetic susceptibilities using the high temperature expansion to evaluate the exchange interaction of $J/k_B = 4.4$ K. It was observed that the lowtemperature specific heat is proportional to T^2, which indicates that $Cs_2LiMn_3F_{12}$ has a good two-dimensional character. In addition, I observed that the excitation gap is induced by the applied magnetic field, which should be attributed to the alternating D vector of the DM interaction. I conducted the powder neutron diffraction to unveil a magnetic ground state. Several magnetic reflections were observed below $T_N \simeq 2.0$ K. However, observed magnetic reflections can be explained in terms of neither $q = 0$ structure nor $\sqrt{3} \times \sqrt{3}$ structure. It was found that the best description of the magnetic structure is given by a propagation vector $q = (1/3, 0, 0)$. To the best of my knowledge, such an unusual magnetic ordering is the first observation in KLAFs. At present, I have no reasonable solution of the spin structure that produces the observed diffraction pattern. To solve the spin structure of the ordered state in $Cs_2LiMn_3F_{12}$ is an important future problem. Because there is no theory on the magnetic excitations for classical and quantum KLAFs with a magnetic order characterized by a propagation vector $q = (1/3, 0, 0)$, INS experiments on $Cs_2LiMn_3F_{12}$ are of great interest.