Investigation of magnetic ground states in mixed kagome systems $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ II

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The Heisenberg kagome antiferromagnet (HKAF) has attracted much attention in magnetism because lots of frustration and quantum effects have been indicated by theories. For example, in the classical spin model case for HKAF, the $q = 0$ or $\sqrt{3} \times \sqrt{3}$ magnetic structure is stabilized when the next-nearest-neighbor interaction is considered. In the case of $S = 1/2$ kagome lattice, exotic magnetic ground states have been theoretically predicted. For example, numerical calculations revealed that the ground state is a magnetically disordered spin liquid. In the ground state, triplet excitations are gapped, and there exists the continuum of low-lying singlet states below the triplet gap. Valence-bond crystals having a periodic arrangement of singlet dimers have also been proposed as the magnetic ground state of $S = 1/2$ HKAF. Experimentally, many types of HKAFs have been investigated as the candidate for the ideal kagome spin lattice material. For example, "jarosite" materials $AM_3(OH)_6(SO_4)_2$ (A = NH_4, Na, or K, M = Fe, Cr), m−MPYNN·BF_4, $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$, $[Cu_3(titmb)_2(OCOCH_3)_6 \cdot H_2O$, $\beta-Cu_3V_2O_8$, etc.

A new candidate for the ideal kagome lattice which has an exotic magnetic ground state was reported[1]. The cupric compound $A_2Cu_3SnF_{12}$ (A = Cs, Rb), which is the subject in this study, is a newly synthesized family of $S = 1/2$ HKAF. For $Cs_2Cu_3SnF_{12}$, the $q = 0$ magnetic structure is observed below $T_N = 20$ K. On the other hand, for $Rb_2Cu_3SnF_{12}$, the first realization of the "pinwheel" valence bond solid ground state in the $S = 1/2$ HKAF is confirmed by inelastic neutron measurements.

Recently, the ground state of mixed kagome $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ was investing by Katayama *et al*[2]. By magnetic susceptibility and specific heat measurements on single crystals, they reported a ground-state phase diagram, which shows the existence of a quantum phase transition from the valence-bond-glass (VBG) phase to the AF phase at $x_c = 0.53$. In this critical concentration, the spin gap and the ordered state disappear at least down to 1.8 K from the magnetic susceptibility measurement results. The characteristic properties at the quantum phase transition point is of great interest, and the information about it will contribute to a further understanding of the properties of the ground states in the kagome lattice compound. In the previous beam time, we carried out the longitudinal-field muon-spin-relaxation (LF-μSR) measurements in the sample of $x_c = 0.53$ down to 0.3 K, and we reported that internal magnetic fields fluctuate at 0.3 K, and that no tendency towards a magnetic phase transition is observed. The purpose of this study is to microscopically investigate the spin dynamics including fluctuating frequencies and values of internal magnetic fields down to lower temperatures.

In zero field (ZF), a low frequency rotational behavior due to dipole-dipole interactions between F^- and μ^+[3] is observed, and to extract the Cu spins dynamics from the entire spectra includung rotational signals, longitudinal fields above 100 gauss are applied. Figure 1 (a) shows the LF-μSR time spectra at 26 mK in the case of $x_c = 0.53$ above 100 gauss. The time spectra are analyzed using the two components function as follows:

$$A(t) = A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) \qquad (1)$$

Here, λ_1 and λ_2 are the muon spin relaxation rates. A ratio of A_1/A_2 is fixed to be 0.97. Fig. 1 (b) shows the obtained LF dependence of muon spin relaxations λ_1 and λ_2. No step-like change in the LF-dependence appears, and λ_1 and λ_2 monotonously decrease with increasing the LF. These results suggest that internal magnetic fields at the muon sites fluctuate with widely spread frequencies down to 26 mK.

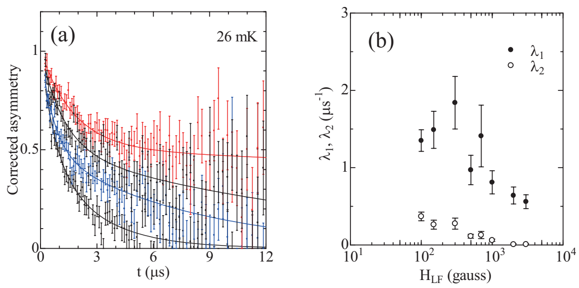

Fig. 1. (a) LF-μSR time spectra at 26 mK in 100, 700, 1000, and 3000 gauss shown from bottom to top. The solid lines represent the fitted results. (b) LF-dependence of the muon spin relaxation rates at 26 mK.

References
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