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

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The Heisenberg kagome antiferromagnet (HKAF) has attracted much attention in magnetism, because several 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[1)]. Valence-bond crystal by a periodic arrangement of singlet dimers has also been proposed as the magnetic ground state of $S = 1/2$ HKAF[2)]. Experimentally, many kinds of HKAF have been investigated as the candidate for the ideal kagome spin lattice material.

Another new candidate for the ideal kagome lattice with an exotic magnetic ground state was reported by Ono *et al.* The cupric compound $A_2Cu_3SnF_{12}$ (A = Cs, Rb), which is the subject of this study, is a newly synthesized family of $S = 1/2$ HKAF[3,4)]. For $Cs_2Cu_3SnF_{12}$, the weak-ferromagnetic behavior is observed below $T_N = 20$ K, and it is suggested that the antiferromagnetic ordered state appears[4)]. On the other hand, for $Rb_2Cu_3SnF_{12}$, the first realization of the "pinwheel" valence bond solid (VBS) ground state in the $S = 1/2$ HKAF are confirmed by inelastic neutron measurements[5)]. Quite recently, mixed kagome systems $(Rb_{1-x}Cs_x)_2Cu_3SnF_{12}$ have been prepared[6)]. By magnetic susceptibility and specific heat measurements on single crystals, they reported a phase diagram, which shows the existence of the quantum phase transition from the VBS to the AF phase at $x_c = 0.53$. In this concentration, the spin gap vanishes and the ordered state disappears from the view point of magnetization results.

We carried out LF-μSR measurements in $x = 0.53$ single crystal to investigate dynamical magnetic properties microscopically. Figure 1 (a) shows LF-μSR time spectra in 200 gauss at various temperatures. It is emphasized that no disappearance of the initial asymmetry is observed. Below 3 K, time spectra are well fitted by the two component function as follows: $A(t) = A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t)$. Here, $A_1 = 0.58$, $A_2 = 0.42$. As shown in Fig. 1 (b), λ_1 increases

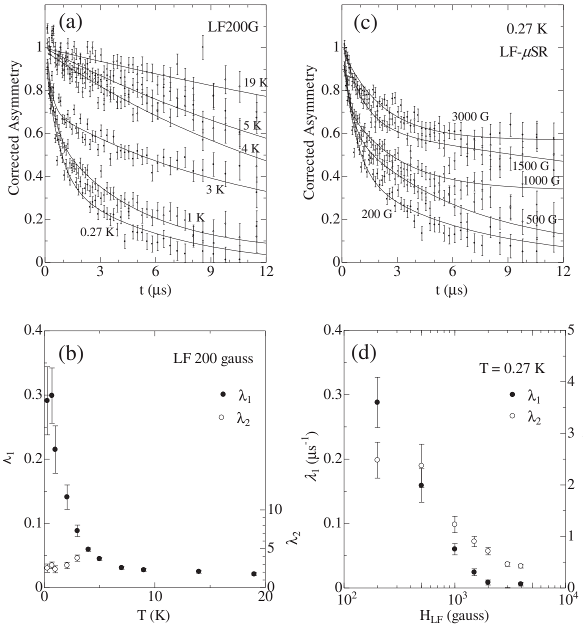

Fig. 1. (a)Temperature dependence of the time spectra in the LF of 200 gauss. (b) Temperature dependence of the muon spin relaxation rate in LF 200 gauss. (c) LF-μSR time spectra above 200 gauss up to 3000 gauss at 0.27 K. (d) LF dependence of the muon spin relaxation rate at 0.27 K.

with decreasing temperature. LF-dependence of time spectra at 0.27 K is shown in Fig. 1 (c), and LF-dependence of relaxation rates is shown in (d). Muon spin relaxation rates are inversely proportional to LF, and such change indicates "white" frequency spectra, which means the spectra are described by summation of continuously distributed frequencies using the Redfield formula. These results suggest that the internal fields fluctuate, and are consistent with reported macroscopic results at least down to 0.27 K.

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