Application of μSR technique to frustrated spin magnets

T. Goto, T. Suzuki[1], I. Watanabe[1], F.-L. Pratt[2], H. Manaka[3] and T. Sasaki[4]

(Physics Division, Sophia Univ., [1]RIKEN Nishina Center, [2]ISIS, Rutherford Appleton Laboratory, [5]Graduate School of Science and Engineering, Kagoshima Univ., [4]Institute for Materials Research, Tohoku Univ.)

Introduction

μSR explores in magnetic materials with a microscopic probe of muon spins capable of detecting phenomena with no spatial coherency, and hence invisible through scattering techniques. It also specializes in the investigation on properties of electron spins that are dynamically fluctuating; by measuring the muon spin depolarization rate under longitudinal field (LF) parallel with muon spin polarization, one can determine both the spin fluctuation amplitude and its characteristic frequency separately.

In this presentation, we introduce our recent study on the spin dynamics in frustrated spin systems κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, the organic magnet with triangular lattice[1], and (CH3)$_2$CHNH$_3$Cu(Cl$_x$Br$_{1-x}$)$_3$, the composite ladder system with bond randomness[2-5]. The ground state of the both systems which show no long range order was revealed by μSR technique. Finally, we also refer to the combinatorial usage of μSR and NMR can be a powerful tool to investigate many exotic states in solids [5].

Experimental

μSR measurements were performed at ISIS Riken-RAL Muon Fascility. A fully polarized surface muon bunch with the momentum of 27 MeV/c was injected perpendicularly to the surface of single crystals, which are set aligned on the silver plate of the cryostat. Time evolution of the muon spin polarization, measured as an *asymmetry*, was traced in the duration of 10 μsec after each injection. A typical statistics to obtain a single depolarization curve was 20–30×10^6 events.

The Fourier component of the dynamical fluctuation of electron spins in the sample is estimated from the Lorentzian component λ in muon spin depolarization curves measured under LF. An entire Fourier spectrum of electron spin fluctuation is obtained by measuring the LF-dependence of λ.

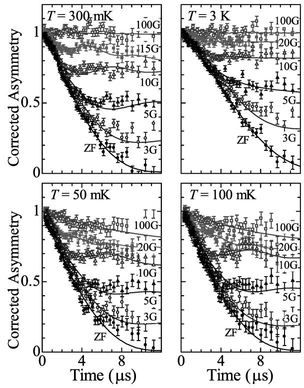

Fig. 1 Typical curves of muon spin depolarization under longitudinal field H_{LF} at various temperatures.

Results and Discussion

Organic spin magnet with triangular-lattice κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

The ground state of the quantum spin system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, in which antiferromagnetically interacting S=½ spins are located on a nearly equilateral triangular lattice, attracts considerable interest from both experimental and theoretical aspects, because a simple antiferromagnetic order may be inhibited owing to geometrical frustration, and hence, an exotic ground state is expected. Several recent reports on the ground state of this system have increased the intrigue by showing completely controversial results: the NMR and the specific-heat, indicating the gapless state, and the thermal conductivity indicating gapped state. By utilizing the μSR microscopic probe, we have investigated its spin dynamics below 0.1 K, unveiling its microscopically phase-separated ground state at zero field[1].

Figure 1 shows typical muon spin depolarization curves under various longitudinal fields H_{LF}'s. Below 300 mK, depolarization curves contain two Lorentzian components with λ_1 and λ_2, indicating that the system microscopically phase separates (Fig. 2). The behavior of λ_2 is explained as the spin fluctuation in conventional paramagnets as described by Redfield function [2], while λ_1 shows quite anomalous LF-dependence — it increases with increasing LF and then decreases, the behavior of which looks close to the spin-singlet state[1].

Based on these experimental results, we have developed a model for the spin state in this system. At high temperature of T > 3 K, the system is homogeneous and all the spins fluctuate paramagnetically. When the charge fluctuation slows down around 6 K, the averaged position of each spin on the dimer is also decentered, following the

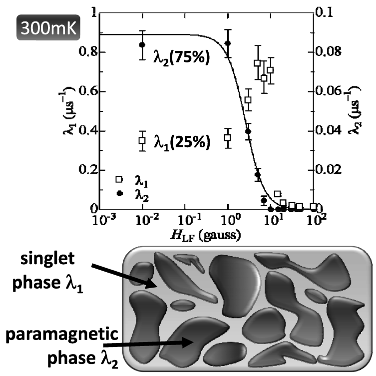

Fig. 2 LF-dependence of muon spin depolarization rate in a phase-separated state (above), and a schematic drawing of the spin state deduced from it (below).

charge redistribution. This brings the random spatial modulation to the effective exchange interaction J and thus transforms the nearly equilateral triangular lattice to the random lattice. This randomness may cause microscopic phase separation in the system at lower temperatures. The phase corresponding to λ_2 is inheritance of the paramagnetic phase at high temperatures, and the newly appeared λ_1 phase is considered to be a spin singlet. In both phases, there is no static spin component, so that it is considered to be difficult to detect this phase separation under zero field by the other probes than μSR.

This observation completely resolves the previous disagreement over the results based on thermal conductivity and specific heat stated above. Since heat energy is present only in the paramagnetic phase of λ_2, it is reasonable that the specific heat contributed from the λ_2 phase exhibits the gapless-type temperature dependence. The gapped-type temperature dependence of thermal conductivity is also reasonable, because the barrier of λ_1 phase surrounding around each island blocks the heat transport that is indispensable to the thermal conductivity.

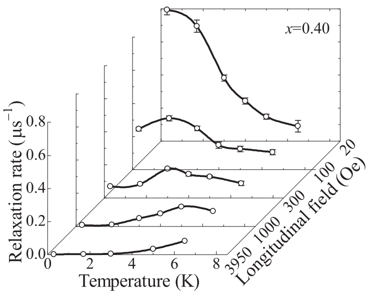

Fig. 3 Temperature dependence of Fourier spectra of electron spin fluctuation determined by LF-μSR.

Composite ladder system with bond randomness $(CH_3)_2CHNH_3$-$Cu(Cl_xBr_{1-x})_3$

Disorder often introduces a non-trivial effect into the quantum spin systems. As is well known, a small fraction of holes destroys the uniform Néel order in the parent compound of high-T_C cuprates by the frustration effect, and a small amount of non-magnetic impurities contrarily stabilizes the Néel state in spin Peierls systems by generating unpaired a spin. Recent theoretical studies have shown by mapping the thermally excited spins and their base singlet phase onto the Boson and vacuum that disorder may bring an exotic ground state Bose-glass, which however is very difficult to detect experimentally, because the critical temperature is zero[3].

The title compound $(CH_3)_2CHNH_3$-$Cu(Cl_xBr_{1-x})_3$ abbreviated as IPA-$Cu(Cl_xBr_{1-x})_3$ is a solid solution of the two spin-gapped composite ladder systems IPA-$CuCl_3$ and IPA-$CuBr_3$ with different spin gaps Δ=14 and 98 K respectively[2,3]. It is previously reported from experiments on macroscopic quantities such as the specific heat or magnetization that the spin excitation spectrum of the system possesses a finite gap when $x < 0.44$. We have investigated microscopically the ground state of the sample with $x = 0.0 - 0.5$ by μSR [3,4] and NMR [4,5], to find that neither a muon-spin rotation nor peak splitting in NMR spectra were observed in all the samples down to 0.3 K (to 15 mK for x=0.40), indicating that the system shows no long range order up to $x = 0.50$.

In this region of x, the Lorentzian component in muon spin depolarization λ curves showed characteristic behavior that indicates a spin freezing. In Fig. 3, LF and temperature dependence of λ shows that the Fourier spectrum of the spin fluctuation changes from white-noise-like flat one at high temperatures around 8 K to the steep peak weighed at around zero energy in low temperatures. We extracted characteristic fluctuation frequency ν as a peak position at each LF to obtain its temperature dependence, which is shown in Fig. 4, where the peak temperature in $1/T_1$ of ^{1}H-NMR measured under several distinct applied fields is also plotted. One can see the smooth connection between two sets of data. This behaviour is interpreted as the soft mode toward the possible phase transition at the absolute zero, or toward an exotic phase, such as Bose-glass.

Conclusion

We have investigated the ground state, especially the spin dynamics of the two frustrated spin systems κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and IPA-$Cu(Cl_xBr_{1-x})_3$ by μSR technique to show that LF- μSR is a powerful tool to the case where the target does not show magnetic order. We also note the combination of these two microscopic probes enables wide-band detection of the spin fluctuation spectrum.

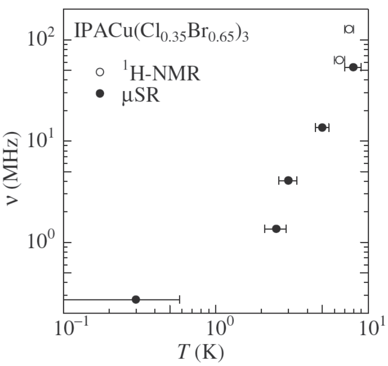

Fig. 4 Temperature dependence of the characteristic spin fluctuation frequency determined from the combination of μSR and NMR.

References

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