Spin dynamics in the $S = 1/2$ zigzag spin chain magnets $K_2CuCl_2SO_4$ and $Na_2CuCl_2SO_4$

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The $S = 1/2$ Heisenberg chain is an outstanding and versatile model system in quantum many-body physics. The cuprate is known to exhibit magnetic properties of the quantum spin chain, thus providing access to the experimental study of spin chains with various perturbations. Among these are frustrated intrachain exchange interactions, staggered g tensors, staggered Dzyaloshinskii-Moriya (DM) interactions, or disorder.

We recently succeeded in synthesizing the pure-phase of a new compound $Na_2CuCl_2SO_4$. This compound has the same crystal structure as $K_2CuCl_2SO_4$ and $K_2CuBr_2SO_4$.[1)] On first inspection, the ferromagnetic nearest-neighbor interaction J_1 and the antiferromagnetic next-nearest-neighbor interaction J_2 through the Cu-Cl(Br)-Cu path seem to compete, however, it was reported that the linear spin chains along the a-axis are formed by the exchange interaction J through the Cu-Cl(Br)-Cl(Br)-Cu path in $K_2CuCl_2SO_4$ and $K_2CuBr_2SO_4$. In addition, from the crystal structure information, these two compounds feature substantial DM interactions that are uniform within each chain, but are anti-parallel in adjacent chains. Therefore, we consider that $Na_2CuCl_2SO_4$ has the unique spin frustration induced by the DM interaction.

Further, we succeeded to grow large single crystals ($\sim$10 mm^3). Therefore, $Na_2CuCl_2SO_4$ is suitable for observing the spin dynamics along and perpendicular to the spin chain by both muon spin rotation and relaxation (μSR) and inelastic neutron scattering. The temperature dependence of the total specific heat divided by temperature C/T and the magnetic susceptibility are shown in Figs. 1(a) and (b). Short-range correlation is developed around 5 K, and the long-range magnetic ordering is observed at $T_N = 0.5$ K. The INS data measured at 1.5 K in the $(h, 1.5, 0)$ scattering plane reveals a spinon continuum, indicating that the Tomonaga-Luttinger liquid state is realized in $Na_2CuCl_2SO_4$ above T_N. Then, in order to investigate the spin fluctuations in $Na_2CuCl_2SO_4$, we performed μSR measurements at the RIKEN-RAL Muon facility at the Rutherford-Appleton Laboratory, UK.

The crystal orientations were determined by Laue X-ray diffraction. The crystal was cut into slices along the bc-plane with a homogeneous thickness of 3 mm, and it was mounted on a silver sample holder. Powder sample of $Na_2CuCl_2SO_4$ was prepared by milling single crystals.

A clear oscillation indicative of the long-range magnetic order was observed at 0.3 K in zero field (Fig. 2(a)). The weak LF asymmetries for $T > 0.6$ K

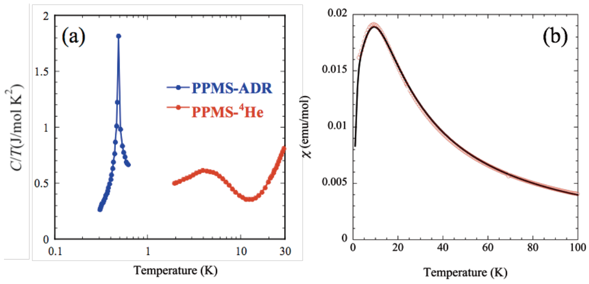

Fig. 1. (a) Temperature dependence of the total specific heat divided by temperature C/T. (b) Temperature dependence of magnetic susceptibility measured at 1 T (open red circles). The grey solid line represents the theoretical curve of the linear spin chain model with a AFM interaction $J = 15.5$ K.

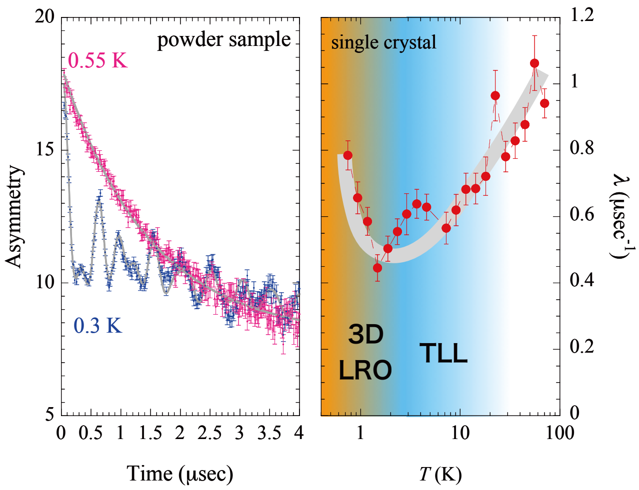

Fig. 2. (a) Selected ZF-μSR signals (asymmetry vs time). The gray lines are the corresponding fits to the data. (b) Temperature dependence of the muon spin relaxation rate in $Na_2CuCl_2SO_4$ measured in longitudinal field 50 G.

are fitted by the stretched exponential function $a(t) = a_1 \exp[-(\lambda t)^\beta] + a_{BG}$, where a_1 is an intrinsic asymmetry, a_{BG} is a constant background, λ is the muon spin relaxation rate, and β is the stretching exponent. We observe a decrease in the λ with increasing temperature above 1 K in the full time window, 0–20 μs as shown in Fig. 2(b). This behavior is expected in the TLL system; they have been seen in other spin-liquid candidates.[2)] Further the analyses of our μSR spectrum measured in both the single crystal and the powder sample are now in progress.

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References

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