Investigation of the magnetic ground state in a new one-dimensional quantum spin system $K_2Cu_3O(SO_4)_3$

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The magnetic ground states of linear chain systems are known to follow the Haldane's conjecture [1)] that the half-integer spin chains are gapless and integer spin chains are gapped, which is supported by a vast amount of theoretical and experimental results. On the other hand, competing interactions or inter-chain interactions can often induce a spin gap, even in half-integer spin systems.[2)] These exciting results indicate that exotic quantum states will be discovered in one-dimensional spin systems with competing magnetic interactions based on the unique arrangement of magnetic ions. In zero-dimensional $S = 1/2$ spin systems on an isolated spin cluster, the magnetic ground state can be determined exactly. If a cluster contains many spins, it is difficult to intuitively understand, and there is a possibility of the presence of an exotic quantum state. In the present study, we report the magnetism of the edge-sharing $S = 1$ tetrahedral cluster chain system $K_2Cu_3O(SO_4)_3$. The unique competing interaction in a spin edge-sharing tetrahedron and the weak one-dimensional inter-cluster couplings lead to the exotic quantum state.

Single-phase $K_2Cu_3O(SO_4)_3$ was verified to possess a monoclinic crystal structure identical to the mineral (Fig. 1). As illustrated in Figs. 1(b) and (c), magnetic ions of Cu^{2+} form the edge-sharing spin tetrahedral cluster that are connected with each other by SO_4^{2-} ions along the b-axis direction. The inter-cluster magnetic interactions should be weaker than intra-cluster magnetic interactions because they are next-nearest-neighbor magnetic interactions through the Cu-O-S-O-Cu exchange paths, hence we called this system the "edge-sharing spin tetrahedral cluster chain." The nonmagnetic potassium ions are located as shown Fig. 1(b), thus resulting in a long inter-chain distance along the a-axis. In this compound, Cu^{2+} ions have tetra-coordination, as shown Fig. 1(d). Therefore, only the presence of very weak inter-chain magnetic interactions through the Cu-O-K-O-Cu exchange paths is expected. Therefore, we conclude that $K_2Cu_3O(SO_4)_3$ has good one dimensionally.

We observed a Schottky-like anomaly at around 4 K, and any anomaly indicative of long-range ordering is absent down to at least 0.7 K. The anomalies at around 4 K and 100 K were seen in the temperature dependence of the magnetic susceptibility. Moreover, a clear spin-gap is seen in the inelastic neutron scattering spectrum.[3)] In order to investigate the spin dynamics in $K_2Cu_3O(SO_4)_3$, we performed muon spin rotation and relaxation (μSR) measurements at the RIKEN-RAL Muon facility at the Rutherford-Appleton Laboratory, UK.

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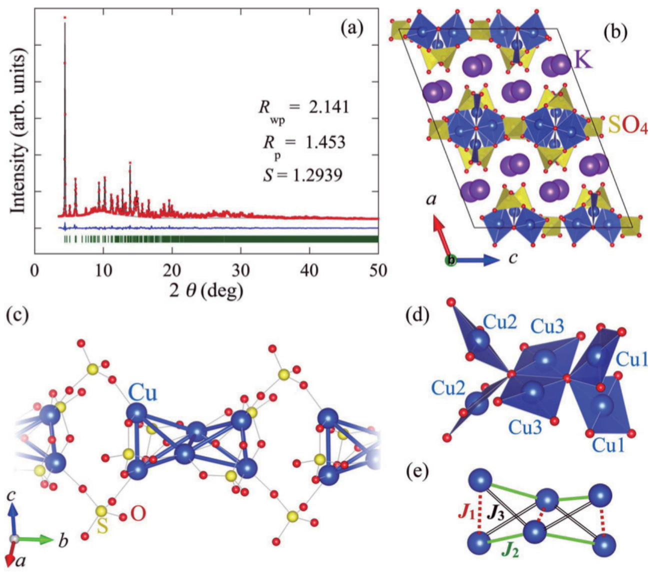

Fig. 1. (a) The observed synchrotron x-ray diffraction intensity pattern for $K_2Cu_3O(SO_4)_3$ and the result of the calculated Rietveld structure refinements. (b), (c), (d) The crystal structure of $K_2Cu_3O(SO_4)_3$. (e) An edge-sharing tetrahedron with nearest-neighbor exchange couplings.

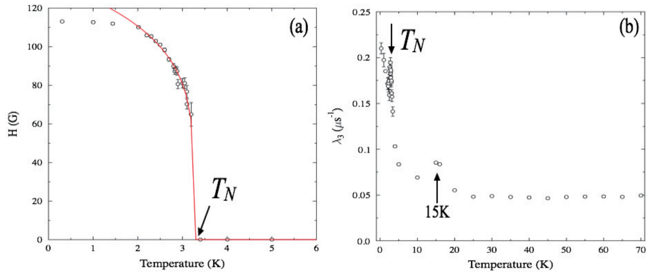

Fig. 2. (a) Temperature dependence of the internal field in $K_2Cu_3O(SO_4)_3$. (b) Temperature dependences of the relaxation rate.

Surprisingly, the μSR results are quite different from the results of the other experiments. A clear oscillation indicative of the long-range magnetic order was observed at 3.2 K (Fig. 2(a)). In addition, the anomaly was observed at around 15 K (Fig. 2(b)). Further detailed studies are required. At this point, we suspect that the sample was deteriorated by moisture.

References

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