Novel quantum spin liquid state in $Ba_3ZnRu_2O_9$

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We discovered a novel type of quantum spin liquid in $Ba_3ZnRu_2O_9$, which has a hexagonal lattice of Ru^{5+} dimers.[1,2] In the temperature (T) dependence of the magnetic susceptibility (χ) of $Ba_3ZnRu_2O_9$, no trace of the Curie tail or glassy behavior has been detected down to 50 mK. We studied the magnetic behavior of the Nb-doped system, $Ba_3Zn(Ru_{1-y}Nb_y)_2O_9$, where Nb^{5+} $(4d)^0$ is a non-magnetic ion that disturbs the formation of the Ru^{5+} dimer. The χ-T curves of the Nb-doped system also show no trace of the Curie tail at low temperatures, indicating that the local Ru^{5+} spin induced by Nb-doping does not act like a free spin. The spin liquid state of $Ba_3ZnRu_2O_9$ has been found to be robust by impurity doping.

To study magnetic dynamics at low temperatures, we attempted to perform ZF-μSR and LF-μSR measurements on three samples of $Ba_3Zn(Ru_{1-y}Nb_y)_2O_9$ ($y = 0$, 0.06, and 0.12), down to 0.3 K. First, we performed ZF-μSR and LF-μSR measurements on $Ba_3ZnRu_2O_9$ using ARGUS. Figure 1 shows the ZF-μSR time spectra obtained at various temperatures for $Ba_3ZnRu_2O_9$. The muon-precession behavior with small amplitude was observed below ~35 K. The initial asymmetry began dropping below ~100 K and the relaxation rate λ_2 rapidly increased at ~100 K with decreasing T. In contrast, the internal field started developing from 80 K and saturated below 3 K at 450 G. These results simply indicate the appearance of long-range magnetic ordering with a tiny long-range ordered moment; however, a spin-liquid state is expected in this sample, as suggested from other studies. Nevertheless, the results of LF-μSR measurements suggest that the internal field is 500 G with a 40% volume fraction. These results indicate that the spin is still dynamic at 2 K and slows at 0.3 K. The spin system coexists with an antiferromagnetic long-range state with tiny ordered moments and dynamical spin liquid state.

Next, we performed similar measurements on $Ba_3Zn(Ru_{1-y}Nb_y)_2O_9$ with $y = 0.06$ and 0.12. However, owing to issues with the dilution refrigerator, we performed ZF-μSR and LF-μSR measurements at temperatures above 2 K on the sample with $y = 0.06$ using ARGUS. Moreover, we performed ZF-μSR measurements above 10 K on the sample with $y = 0.12$ using EMU. Although the temperature range was limited, we observed similar decreasing behavior in the initial asymmetry at ~100 K with decreasing T for $y = 0.06$ and 0.12. However, the muon-spin precession behavior was not observed in the measured temperature region for $y =$ 0.06 and 0.12, which may be due to the randomness effect by Nb-doping. Then, the overall scheme of the magnetic behavior was found to be similar for all samples with y = 0, 0.06 and 0.12.

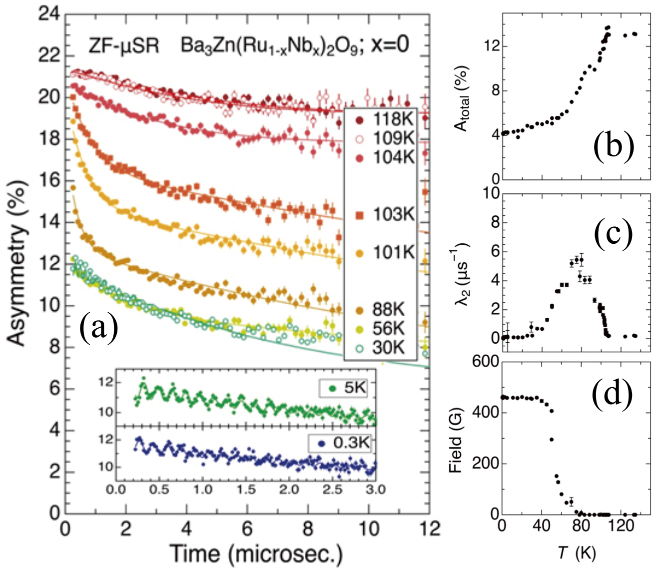

Fig. 1. (a) ZF-μSR time spectra of $Ba_3ZnRu_2O_9$ measured at various temperatures. Temperature dependence of (b) the initial asymmetry Atotal, (c) relaxation rate λ_2, and (d) internal field of $Ba_3ZnRu_2O_9$ deduced from the analysis of ZF-μSR time spectra.

Using these results, we consider a novel scenario in this system wherein a spin liquid state accommodates long-range magnetic ordering with a tiny ordered moment. When Nb substitution is increasing, the coexistence of a spin-liquid state with long-range magnetic ordering is suppressed by the random disconnection of the frustrated magnetic path, and magnetic ordering becomes smeared. Thus, detailed ZF-μSR and LF-μSR measurements are required, along with other measurements, for $Ba_3ZnRu_2O_9$ ($y = 0$, 0.06, and 0.12).

Recently, Tanaka and Hotta reported a theoretical study based on the intriguingly characteristics of the $Ba_3MRu_2O_9$ family.[3] They adopted Heisenberg exchange interactions, J, J', J'', and biquadratic interaction, B. Using a phase diagram with reasonable J, J', J'', and B parameters, they proposed the parameter regions of novel magnetic states such as the nonmagnetic singlet, Ferroquadrupolar nematic Bose-Einstein condensation (FQ-p-BEC), AFM-solid, AFM-BEC-coexistence, *etc.* states. It is significantly interesting in the correspondence between the theoretically proposed novel magnetic states and the experimental results of present materials.

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References

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