μSR investigation of novel magnetism in the 4d Heisenberg-Kitaev honeycomb compound α-RuCl₃

S. Yoon,^{*1,*2} S.-H. Do,^{*3} Y. S. Kwon,^{*4} K.-Y. Choi,^{*3} I. Watanabe,^{*1,*2} and B. J. Suh^{*2}

The Kitaev honeycomb spin model has been considered as an approach to study the spin liquid state.¹⁾⁻³⁾ This model is based on the two-dimensional honeycomb network of magnetic ions with strong spin orbit coupling. This environment can suppress the Heisenberg AF exchange,^{4),5)} and it can result in anisotropic exchange depending on the bond direction.⁵⁾ This exchange leads to a highly frustrated spin state with spatial and spin degrees of freedom.¹⁾⁻³⁾ This fractional spin excitation that reflects the frustrated spin state can be translated by an emergence of Majorana fermions with their own antiparticles.^{1)-3),5)}

Honeycomb ruthenate, α -RuCl₃, was suggested as a candidate to realize the Kitaev spin model.⁶⁾ It provides proper environments for the Kitaev spin model, a cubic environment composed of Cl⁻ ions for revealing strong spin orbit coupling, and honeycomb layers connected by weak van der Waals forces for realizing the ideal two-dimensional environment for Kitaev exchanges.⁶⁾ It exhibits two distinct magnetic anomalies, at 6 K, and 14 K in the DC susceptibility. Also, significant exchange anisotropy is found by observing the different Weiss temperatures with opposite signs in the different external field orientations.⁷⁾⁻⁹⁾



Fig. 1. (upper panel) Time differential spectra of the muon polarization at several temperatures in the zero field condition, and (lower panel) temperature dependence of the internal field extracted by fitting muon time spectra at the zero field with an assumption of the magnetic ordered state

Our synthesized honeycomb ruthenate single crystal shows three distinct anomalies at 6 K, 9.5 K, and 12.5 K in the result of the heat capacity measurement, and the magnetic susceptibility exhibits the result similar to those in the previous reports.⁷⁾⁻⁹⁾ Besides, this material is found to be similar to the ideal two-dimensional environment as a rhombohedral lattice was revealed in the crystal structure analysis. In this environment, the effects induced by stacking faults in the honeycomb layers are expected to decrease. For instance, different magnetic transitions emerge from different stacking orders in the neutron diffraction results.¹⁰⁾ Additionally, a single anomaly at 14 K is revealed in honeycomb ruthenate with a monoclinic lattice.¹¹⁾

Fig. 1 shows the muon time spectra at several temperatures in the zero field condition (upper panel), and the temperature dependence of internal fields extracted by fitting muon time spectra with an assumption of the magnetic ordered state (lower panel). The muon time spectra obtained at the zero field exhibit the clear oscillation below 6 K, and changes of the initial asymmetry, which indicate the magnetic ordered state. In addition, single exponential relaxation is exhibited over 13 K. With these phenomena, we analyzed the muon time spectra based on the magnetic ordered state below 13 K as shown in Fig. 1 (lower panel). Furthermore, we observed similar relaxations between 4 μ s and 6 μ s at 1.7 K, and 6 K. These relaxations can provide evidence of the frustrated spin state.

In summary, we performed a microscopic investigation of honeycomb ruthenate with a rhombohedral lattice using spin polarized muons. We analyzed the muon polarizations below 13 K in the zero field condition with an assumption of the magnetic ordered state revealed by the clear oscillation below 6 K, and single exponential relaxation over 13 K.

References

- 1) W. Witczak-Krempa *et al.*, Annu. Rev. Condens. Matter Phys. **5**, 57 (2014).
- 2) J. G. Rau et al., arXiv:1507.06323.
- 3) R. Schaffer et al., arXiv:1512.02224.
- 4) A. Kitaev, Ann. Phys. 321, 2 (2006).
- 5) G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009).
- 6) K. W. Plumb et al., Phys. Rev. B 90, 041112 (2014).
- 7) Y. Kubota et al., Phys. Rev. B 91, 094422 (2015).
- 8) J. A. Sears et al., Phys. Rev. B 91, 144420 (2015).
- 9) M. Majumder et al., Phys. Rev. B 91, 180401 (2015).
- 10) A. Banerjee et al., arXiv:1504.08037.
- 11) R. D. Johnson et al., Phys. Rev. B 92, 235119 (2015).

^{*1} RIKEN Nishina Center

^{*2} Department of Physics, The Catholic University of Korea

^{*3} Department of Physics, Chung-Ang University

⁴ Department of Emerging Materials Science, DGIST