

## 1. Introduction

Spin-state transitions appear in a wide range of sciences. The spin-state transition denotes a change between two well-defined spin states, whose boundary ranges provide an immense scope for fascinating phenomena. Conventional low-spin (LS) and high-spin (HS) states are known to exist in compounds of octahedrally coordinated  $3d^6$  transition metal ions ( $\text{Co}^{3+}$  and  $\text{Fe}^{2+}$ ). Recently, an unconventional  $\text{Co}^{3+}$  spin state, that is, an excitonic-insulating (EI) state described by the quantum linear combination of different spin states, has been theoretically predicted. This novel state is expected to appear when the energy gap between the two spin states ( $E_g$ ) is critically narrow.

However, no compound has yet been recognized as a spin-state criticality in the ground state without requiring special external conditions, which hampers the experimental identification and clarification and thereby the design of advanced materials with respect to the spin-state critical range. This study reports on the characteristics of the novel substitution system,  $\text{LaCo}_{1-y}\text{Sc}_y\text{O}_3$ .

## 2. Experiment

Neutron spectroscopy was performed on the chopper spectrometer AMATERAS (BL14) at the MLF of the J-PARC. The incident energy  $E_i$  was set to 4.7 meV, and the energy resolution under elastic condition was approximately 2.3% to  $E_i$ . The main disk chopper speed was fixed at 300 Hz. A He closed-cycle refrigerator was used. The data were obtained by the UTSUSEMI software provided by the MLF.

## 3. Results and Conclusions

Figure 1 shows the measured energy spectra at elevated temperatures in comparison with the reference  $y = 0$  data [1]. For the  $y = 0$ , no signal is observed at the minimal temperature and the gapped magnetic excitations grow with increasing the temperature, indicating that the high-spin states are thermally activated. In contrast, for the  $y = 0.05$ , the clear signal is already obtained from the minimal temperature.

This study is just before the submission by combination with macroscopic data.

### Reference

[1] A. Podlesnyak *et al.*, Phys. Rev. Lett. **97**, 247208 (2006).

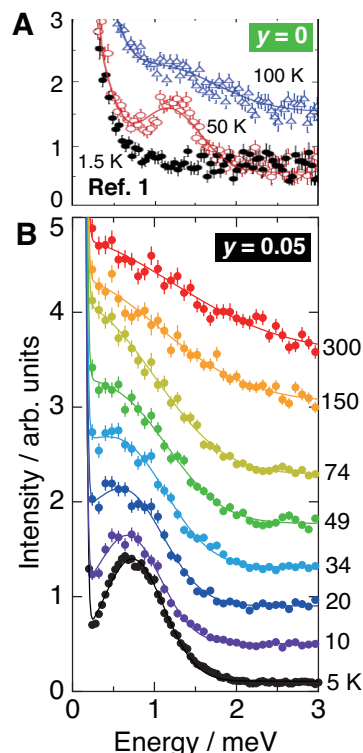


FIG. 1: (A) Energy spectra measured at the elevated temperatures for the reference  $y = 0$  composition and (B) those for the  $y = 0.05$  composition. The curves are obtained from the least-square fitting with Gaussians.