Magnetic excitations in the honeycomb materials $Cu_3Ni_2SbO_6$ and $Cu_3Co_2SbO_6$

L.-J. Chang[A], S. Shamoto[B,C], K. Nawa[D], K. Kinjo[D]

[A]*National Cheng Kung Univ.*, [B]*q-RADA.*, [C]*JAEA*, [D]*Tohoku Univ.*

Delafossite derivatives $Cu_3Ni_2SbO_6$ (CNSO) and $Cu_3Co_2SbO_6$ (CCSO) crystallize in a 2*H* polymorph (C2/c) with honeycomb lattices of alternating NiO_6/SbO_6 and CoO_6/SbO_6 layers. These geometrically frustrated systems exhibit higher-order magnetic interactions, making them promising candidates for Kitaev physics, a unique, solvable quantum spin-liquid model. Recent studies suggest that CCSO exhibits bond-dependent anisotropy and that its spin dynamics resemble a generalized Heisenberg-Kitaev model [1].

Polycrystalline CNSO and CCSO were synthesized using the conventional solid-state reaction method. The structure of the samples was confirmed to belong to the *C*2/*c* space group through room-temperature powder X-ray diffraction (PXRD). The results reveal the magnetic transitions at 24 K for CNSO, and at 18 K for CCSO. The Curie-Weiss temperatures (θ_{CW}) estimated by the Curie-Weiss law are -11.47 K for the CNSO and 1.80 K for the CCSO, suggesting these two compounds may have different magnetism, and the latter may have ferromagnetic correlations.

About 10 grams of powder samples of CNSO and CCSO each were wrapped in aluminum foil and mounted on a sample holder for the experiments. Because both compounds have magnetic transition temperatures around 20 K, we used a closed-cycle cryostat to control the temperature down to the order-state regime. In addition, experiments above the transition temperature were also carried out for the background clarification and comparison. The neutron scattering experiments were carried out on 4G-GPTAS, JRR-3, a general-purpose triple -axis thermal neutron spectrometer. The Q-range scans were between 0.5 ~ 2.5 $Å^{-1}$, and the E-range scans were set from -2 ~ 30 meV.

Figure 1 shows the spectra of CNSO (upper panel and CCSO (lower panel) taken at 4 K on GPTAS. A flat branch of the magnetic excitation was observed at 10 meV for CNSO powder sample. The weak flat signal at 18 meV may be attributed to background contamination or to a very small spin-orbit coupling. For the lower panel, a typical flat-band excitation observed at ~22 meV could be attributed to spin-orbit coupling in Co^{2+}. The higher intensity of this mode in the low-Q regime confirms its magnetic behavior rather than a phonon contribution. The spin-wave configuration could be observed between 0 and 8 meV. The spectra for both compounds have been simulated using linear spin-wave theory in the SpinW software. However, the Kitaev term in the CCSO Hamiltonian is necessary to achieve better results, suggesting that CCSO may be a candidate for a Kitaev quantum spin liquid.

[1] Gye-Hyeon Kim *et al.* Sci. Adv., **10**, eadn8694 (2024).

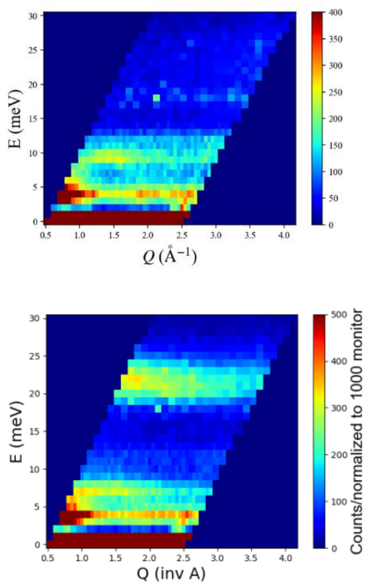

Fig. 1. The spectra for CNSO (upper panel) and CCSO (lower panel) obtained from GPTAS at 4 K.