Experimental report

23/03/2021

Proposal: **4-05-760** **Council:** 10/2019

Title: Investigation of spin excitations in candidate Kitaev magnet Na2Co2TeO6

Research area: Physics

This proposal is a new proposal

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Samples: Na2Co2TeO6

Instrument	Requested days	Allocated days	From	To
THALES	8	6	24/02/2021	02/03/2021

Abstract:

Na2Co2TeO6 has recently been proposed to be a Kitaev-like magnet and was experimentally found to exhibit some resemblances with previous Kitaev materials. We propose to perform an inelastic neutron scattering experiment on Na2Co2TeO6 single crystals. The spin excitation spectrum in the magnetically ordered states will be carefully measured. By fitting the excitation spectrum, we will determine the leading interactions in this compound, which will allow us to assess how close it is to the Kitaev model. To further explore possible unconventional spin excitations, we will also make measurement well above the magnetic ordering temperature.

Investigation of spin excitations in candidate Kitaev magnet $Na_2Co_2TeO_6$

Introduction

$Na_2Co_2TeO_6$ has recently been proposed to be a candidate of Kitaev magnet based on Co^{2+} ion. In this compound the Co^{2+} ions form a two-dimensional honeycomb lattice staggered along c-axis. At low temperature, the magnetism of this compound is believed to be governed by an effective spin-1/2 due to spin-orbital coupling and an octahedral crystal field. Although it enters into a long-range antiferromagnetic (AFM) order below T_N ~ 26.5 K, this magnetic order can be suppressed by in-plane magnetic fields, which is similar with the Kitaev magnet α-$RuCl_3$. Nevertheless, how close it is to a Kitaev magnet is unclear up to now and further studies of the magnetic excitations are warranted, especially for the phase at high magnetic field.

Experiment

The experiment was performed at Thales spectrometer in Flatcone mode. Due to the covid-19 pandemic, the experiment was delayed. During that period, we already had a full map of the magnetic excitation with a TOF spectrometer (4SEASONS) at J-Parc, Japan. Therefore, we changed the originally proposed plan and further requested for a 10 T magnet to explore the magnetic excitations under in-plane fields.

The sample was a coaligned $Na_2Co_2TeO_6$ single crystal array with total mass of about 0.6 gram. (H, 0, L) plane of the crystal was put in horizontal so that the magnetic field was applied within the honeycomb plane, parallel to a zigzag chain of the Co honeycomb layer. A fixed final neutron energy of 4 meV was selected with the Si111 analyzers of FlatCone. In the experiment we rotated the sample in 1 deg per step to cover about a quadrant of (H, 0, L) plane, but without moving the Flatcone. Magnetic excitations do not have a L-dependence, so we integrated all the measured L-range in the data presented below.

Results

We first studied the field dependence of the lowest spin wave branch at 1.5 K. With the increase of magnetic field, we found the band bottom of the lowest spin wave branch only slightly descends while the band top first descends then rises after the field exceeds 8.2 T (Fig. 1 (a) and (b)). The bandwidth of the spin wave has a minimum at ~8.2 T, while its intensity has a maximum there (Fig. 1 (c)). This indicates a field-induced phase transition at ~ 8.2 T. By extrapolating the field dependence of the intensity at the band bottom, it seems to terminate at ~10.4 T, which is beyond our maximal magnetic field of 10 T. This observation indicates the existence of another critical magnetic field slightly above 10 T, where the AFM order is expected to completely disappear.

We also noticed that the energy width of the spin wave gets smaller as the field approaching 8.2 T. By carefully inspecting the spectra, we found that there are actually two spin wave branches coexist in a field range below 8.2 T (Fig. 2). The energy dependence of the intensity around (0.25, 0) clearly shows a two-peak structure at 6.8 T and only a sharp single peak at 8.2 T. As there is only one branch observed at 0 T, the appearance of the second branch indicates the transition at ~ 8.2 T is discontinuous (or first-order). From our single-crystal neutron diffraction experiment performed in a similar field range (not shown here), we believe that the first-order transition range is between 5 and 8.2 T, where field-hysteretic behavior can be observed at low T.

As the spin-wave intensities become suppressed by the field, we found a new gapped excitation mode at 4.5 meV around the zone center (Fig. 3 (a)), which is most evidenced at the maximal magnetic field of 10 T. When warming up to 27 K, the spin wave excitation disappears while this gapped mode persists (Fig. 3 (b)). Fig. 3 (c) and (d) present more detailed temperature dependence studies at 10 T around the zone center, which reveal that the gapped mode persists well above the T_N (of about 10 K) at 10 T. This unusual temperature dependence behavior indicates that it should be of a different origin from the conventional spin-wave excitations.

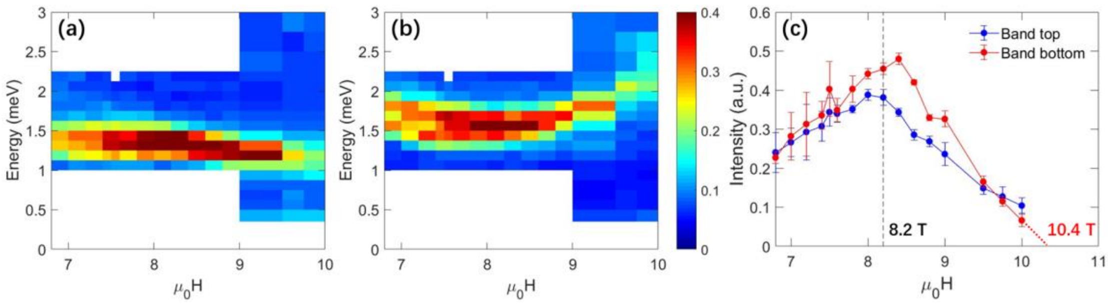

Fig. 1 (a) and (b) Field dependence of the band bottom and top of the lowest spin wave branch at 1.5 K. (c) Field dependence of the intensity at the band bottom and top. The intensity is extracted by fitting the energy dependence of the intensity (e.g. Fig. 2 (d) and (d)) with a single gaussian profile.

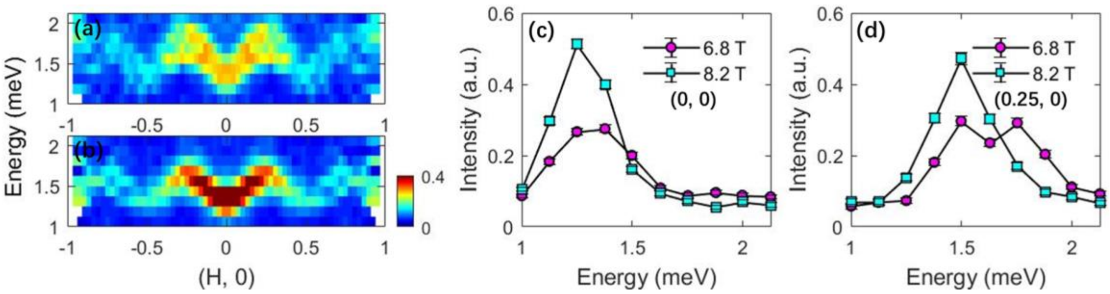

Fig. 2 (a) and (b) The lowest spin wave branch at 1.5 K under magnetic fields of 6.8 T and 8.2 T, respectively. (c) and (d) The corresponding energy dependences of the intensity at (0, 0) and (0.25, 0).

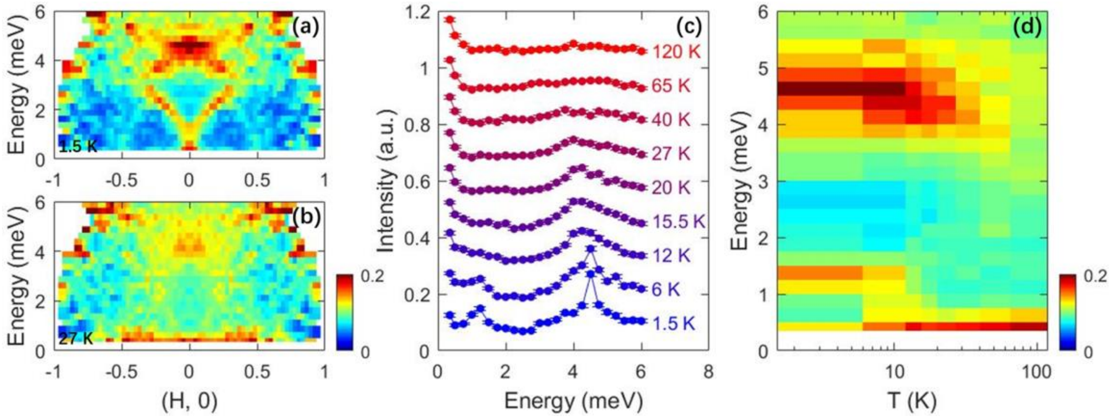

Fig. 3 (a) and (b) The magnetic excitation spectra under 10 T field at 1.5 K and 27 K, respectively. (c) The energy dependence of the intensity of (0, 0) at various temperatures. Offset for clarity. (d) Presenting the data of (c) in a false-color map.

Conclusion

At 1.5 K, we found a field-induced first-order phase transition at ~8.2 T, beyond which the system enters an AFM order different from the zero-field state, yet its spin-wave intensities gradually diminish with further increasing field. The final disappearance of this AFM order is expected to occur at about 10.4 T. Approaching the highest accessible magnetic field of 10 T, an unconventional gapped excitation mode around the zone center was observed. This mode persists into the paramagnetic phase and may be related with possible Kitaev quantum-spin-liquid physics in $Na_2Co_2TeO_6$.

Special Acknowledgements

The experiment was performed ‘remotely’ during the COVID pandemic, and instrument scientist Paul Steffens did all the hard work on the hardware side, ensuring an extremely smooth performance of the whole experiment. He also provided much help with the FlatCone data analysis software package (nplot & PlotMultiple), including diagnosing and removing spurious scattering in the collected data.