Experimental report

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Proposal: **4-01-1346** **Council:** 4/2014

Title: Spin dynamics of frustrated spin-2chain system Ca3Co2O6 in a magnetic field

Research area: Physics

This proposal is a continuation of 4-01-1215

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Samples: Ca3Co2O6

Instrument	Requested days	Allocated days	From	To
IN20 Flatcone	11	7	29/07/2015	06/08/2015
IN8 Flatcone	8	0		

Abstract:

Ising-like spin-chain compound Ca3Co2O6 has recently attracted a lot of experimental and theoretical attention because of its interesting magnetic properties, such as field-induced magnetization steps, time-dependent magnetic order. In our recent inelastic neutron scattering study, the observed large ratio of the spin gap to the bandwidth (~8) and the absence of dispersion perpendicular to the chains indicate the highly 1D nature of this material in spin space and in real space. In the proposed work, we want to investigate the effect of a magnetic field on the spin dynamics by measuring the magnetic excitations in the longitudinal and transverse field. It is theoretically predicted that the gap in the magnetic excitation spectrum of Ising spin-chain softens as a magnetic field is applied transverse to the preferred spin direction and goes to zero at critical field. However, different ratios of interchain to intrachain interactions and different degrees of deviation from the ideal Ising spin Hamiltonian, is expected to modify the magnetic excitations.

Spin dynamics of frustrated spin-2 chain system $Ca_3Co_2O_6$ in a magnetic field

Experiment No.: 4-01-1346, Instrument: IN20

Ising-like spin-chain compound $Ca_3Co_2O_6$ has recently attracted a lot of attention because of its interesting magnetic properties, such as field-induced magnetization steps, time-dependent magnetic order, and nanoscale magnetic fluctuations [1–9]. It crystallizes in the space group $R\bar{3}c$ and the crystal structure contains spin-chains, made up of alternating face-sharing CoO_6 octahedra (OCT) and CoO_6 trigonal prisms (TP). Ferromagnetic (FM) intrachain and antiferromagnetic (AFM) interchain interactions combined with a triangular lattice arrangement of the spin chains give rise to a geometrical frustration.

In the dc magnetization [1, 6, 10] with $H \parallel c$, field driven magnetization steps were observed at low temperatures. The effective 2D Ising model was initially proposed to explain the magnetization steps. However, the recently observed incommensurate longitudinal spin-density wave (SDW) structure below the Néel temperature, $T_N \approx 24$ K [4] and the time dependent magnetic order-order transition from the SDW to a commensurate AFM (CAFM) structure below ~ 10 K in polycrystalline $Ca_3Co_2O_6$ [3, 9] have challenged the validity of this model. The 3D character of the magnetic interactions was proposed to explain the time dependent magnetic order [11]. The very recent small angle neutron experiments showed the presence of nanoscale magnetic fluctuations firmly embedded inside the SDW magnetic structure [8]. These fluctuations were reported to slow down into a super-paramagnetic regime of stable spatiotemporal nano-structures at lower temperatures. A frustrated quantum Ising model was recently proposed to explain the magnetic properties [12]. A small transverse field in this model was reported to stabilize a ferrimagnetic phase. However, this model does not explain the order-order transition from the SDW to CAFM phase. A theoretical model to describe the 1/3 magnetization plateau and magnetic properties of $Ca_3Co_2O_6$ is still missing, mainly because direct determination of exchange parameters and single-ion anisotropy parameters has not been carried out.

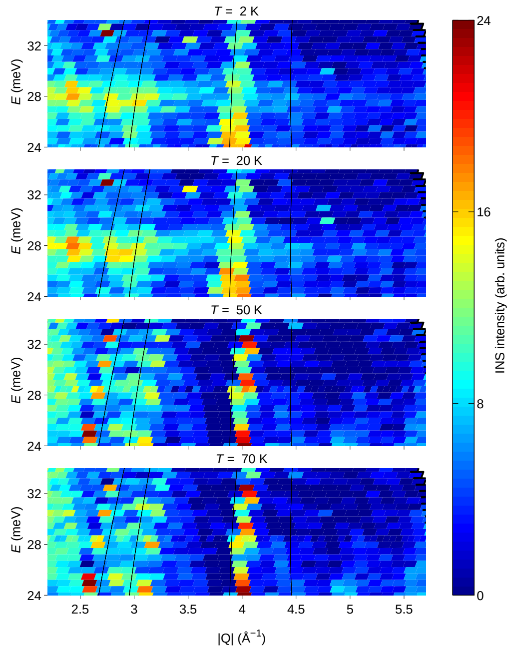

Fig. 1: Intensity color maps of the observed inelastic neutron spectrum at 2, 20, 50, and 100 K under an applied magnetic field of 13.5 T at the zone center. Thin black lines represent the position of Aluminium powder lines.

We have recently investigated magnetic excitations in single crystals of $Ca_3Co_2O_6$ by means of inelastic neutron scattering (INS) [13]. At 1.5 K, a gapped band of magnetic excitation, dispersing between ~ 27 and 30.5 meV, with a bandwidth of ~ 3.5 meV, was observed. The gap was minimum at zone center and maximum at zone boundary. The observed large ratio of the gap to bandwidth (~ 7.7) and no dispersion perpendicular to the chain direction confirmed that this material is highly 1D.

To understand the origin of 1/3 magnetization plateau and steps in the magnetization, we investigated the magnetic excitations in $Ca_3Co_2O_6$ under an applied magnetic field of 13.5 T [Fig. 1]. The INS measurements were performed using the thermal-neutron spectrometer IN20 (in the Flatcone multi-analyzer configuration using unpolarized neutrons) at the ILL, Grenoble. The sample was mounted such that horizontal scattering plane was ($HK0$) and magnetic field was applied along the chain direction (c-axis). In this scattering plane, we could access only the center of the Brillouin zone. The spin wave gap is found to increase (by 1.3 meV) under an applied magnetic filed of 13.5 T [Fig. 1]. Surprisingly, the magnetic signal shifts towards higher energies upon warming (above $T_N = 25$ K). The temperature dependence of the spin wave gap at higher temperature could not be investigated because of (i) weakening of the magnetic signal, (ii)exceptionally high background from the magnet (as coils were not covered with Cd), and (ii) quenching of the magnet during the beam time. To fully understand the spin dynamics in $Ca_3Co_2O_6$, measurements of magnetic excitation (with field along the c-axis) at higher temperatures (above 50 K) will be required.

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