Experimental Report

Proposal: **5-14-249** **Council:** 10/2012

Title: Single crystal neutron diffractionstudy of the magnetic order-order transition in Ising- like spin-chain compound Ca3Co2O6

This proposal is a new proposal

Researh Area: Physics

Main proposer: **JAIN Anil**

Experimental Team: JAIN Anil
INOSOV Dmytro
PORTNICHENKO Pavlo
JANG Hoyoung

Local Contact: OULADDIAF Bachir

Samples: Ca3Co2O6

Instrument	Req. Days	All. Days	From	To
D10	10	7	21/05/2013	28/05/2013

Abstract:

The present proposal on the Ising- like spin-chain compound Ca3Co2O6 is motivated by an observation of the slow magnetic order-order transition from the spin density wave (SDW) structure to the commensurate antiferromagnetic (CAFM) structure with a time scale of several hours in the neutron powder diffraction study. For the powder sample, under an applied field, the magnetic Bragg peaks corresponding to the CAFM structure disappears for H > 1 T. In powder sample (with magnetic field applied to a pellet of grains) the direction of the field relative to the easy axis changes due to the powder averaging. We, therefore, propose to carry out single crystal neutron diffraction measurements on Ca3Co2O6 using the high-flux single-crystal diffractometer D10 (wavelength = 2.36 angstrom) at the ILL in the temperature range of 2-30 K under zero as well as applied magnetic field (maximum 6T).Our proposal aims to study the effect of an external magnetic field on the ground state as well as on the time scale of the slow magnetic order-order transition from the SDW to the CAFM structure.

Single crystal neutron diffraction study of the magnetic order-order transition in Ising-like spin-chain compound $Ca_3Co_2O_6$ (Experiment No: 5-14-249, Instrument: D10)

Ising-like spin-chain compound $Ca_3Co_2O_6$ 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 [1–9]. It crystallizes in a rhombohedral structure (space group $R\bar{3}c$) which represents a hexagonal arrangement of 1D chains, made up of alternating face-sharing CoO_6 octahedra (OCT) and CoO_6 trigonal prisms (TP). Due to the crystalline electric field, Co^{3+} ions at the TP and OCT sites are in high-spin ($S = 2$) and low-spin ($S = 0$) states, respectively. Besides, the large single-ion anisotropy of the Co^{3+} ions at the TP site leads to an Ising character of the Co^{3+} spins, pointing along the c-axis. 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 [2, 8, 10] as well as neutron diffraction studies [7], 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 [5] and the time dependent magnetic order-order transition from the incommensurate SDW to a commensurate AFM (CAFM) structure below ~ 10 K in polycrystalline $Ca_3Co_2O_6$ [4, 11] have challenged the validity of this model . Afterwards, the CAFM structure was proposed as the ground state for the $Ca_3Co_2O_6$ [12]. Note that the presence of the CAFM phase strongly depended upon the cooling protocol [4, 11]. The majority phase in the neutron powder diffraction pattern was SDW if the sample was cooled rapidly, however, the SDW and CAFM coexisted if the sample was cooled slowly. Furthermore, a time dependent change in the position of the incommensurate AFM reflection [from (10δ) to (100)] was observed in a recent single crystal neutron diffraction study [3] and a square wave-wave magnetic structure was proposed.

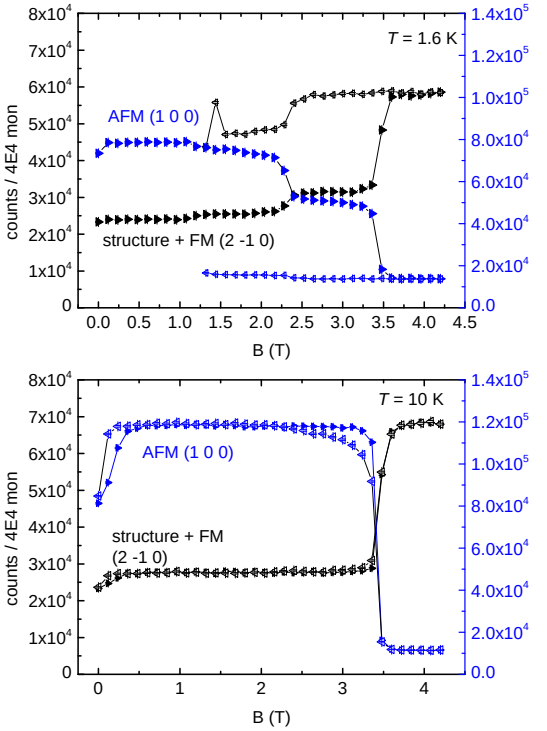

Fig. 1: The intensity of the AFM peak (100) peak and the FM peak (2-10) as a function of magnetic field at 1.6 K (top) and 10 K (bottom). The solid symbols represent the data taken while increasing the field and the open symbols show the data taken while decreasing the field.

The aim of the present proposal was to investigate the effect of magnetic field on the the time scale of the slow magnetic order-order transition using the single crystal of $Ca_3Co_2O_6$. The single crystal neutron diffraction measurements were carried out using the D10 diffractometer at the ILL. As the intensity of the AFM peak corresponding to the CAFM phase was expected to be low and dependent on the cooling conditions (rapid or slow), we co-aligned many small crystals into a mosaic (with c-axis mosaicity $\leq 3^\circ$). The sample was mounted in a 6T vertical cryomagnet with the field directed along the c-axis, allowing the measurement of the scattered intensity in the $(HK0)$ plane. The temperature dependence of the AFM peak (100), corresponding to the SDW structure, confirmed a transition to the AFM state at $T_N \sim 24$ K, in agreement with earlier results [6, 7]. A decrease in the intensity of the AFM peak (100) was also observed at low temperature. An increase in the background was observed at $Q = (\frac{1}{2}\ \frac{1}{2}\ 0)$, where one expects a strong AFM peak corresponding to the CAFM structure. A large difference in the intensity of the AFM (100) peak (corresponding to the SDW structure) was observed for the measurements under the field-increasing and field-decreasing conditions (Fig. 1), indicating that the magnetic states for a zero-field cooled sample and a sample that has previously been exposed to a high magnetic field are completely different. A time variation in the intensity of the AFM peak (100) was observed, if the sample was cooled in zero field, and a magnetic field of 4.2 T was applied and subsequently reduced to lower field (such as 1.2, 2.4, and 3.6 T, where the steps in the magnetization were reported). Neutron diffraction measurements (under an applied field) in the $(H0L)$ scattering plane will be required for the further investigation of the time dependent changes in the position of the incommensurate AFM reflection from (10δ) to (100) [3].

[1] A. Jain *et al.*, PRB **88**, 224403 (2013).
[2] V. Hardy *et al.*, PRB **70**, 214439 (2004).
[3] T. Moyoshi *et al.*, J. Phys. Soc. Jpn. **80**, 034701 (2011).
[4] S. Agrestini *et al.*, PRL **106**, 197204 (2011).
[5] S. Agrestini *et al.*, PRL **101**, 097207 (2008).
[6] O. A. Petrenko *et al.*,Eur. Phys. J. B **47**, 79 (2005).
[7] C. L. Fleck *et al.*, EPL 90, 67006 (2010).
[8] J.-G. Cheng *et al.*,PRB **79**, 184414 (2009).
[9] A. Jain *et al.*, PRB **74**, 174419 (2006).
[10] A. Maignan *et al.*, Eur. Phys. J. B **15**, 657 (2000).
[11] J. A. M. Paddison *et al.*, arXiv:1309.3222.
[12] L. C. Chapon, PRB **80**, 172405 (2009).