Resonant x-ray magnetic scattering under high magnetic fields on Ca$_3$Co$_2$O$_6$ single crystal

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The compound Ca$_3$Co$_2$O$_6$ and its derivatives [1-9 and references therein], crystallizing in the rhombohedral structure (space group R3-c), have attracted a considerable attention due to their peculiar physical properties. The compound Ca$_3$Co$_2$O$_6$ is a model system for a triangular lattice Ising antiferromagnet. We have performed resonant x-ray magnetic scattering studies on a flux grown Ca$_3$Co$_2$O$_6$ single crystal sample of size ~ 3 mm x 1 mm x 1mm. In September 2012, preliminary measurements without magnetic fields were performed on the sample using the Hutch I of the resonant scattering and diffraction beamline P09 of PETRA III synchrotron source at DESY. During 19-23 August 2013, detailed resonant magnetic x-ray scattering experiments under magnetic fields were performed using the Hutch II of the same beamline P09 of PETRA III. For the experiments under magnetic fields, Ca$_3$Co$_2$O$_6$ single crystal was mounted inside a 14 T cryomagnet. The crystal was mounted in a way so as to have [h h 0] and [h 0 0] axes in the horizontal scattering plane. Magnetic field was applied vertically up along the c-axis of the crystal. Incident photon beam was p polarized and polarization analysis of the scattered photons has been performed. The PG (600) crystal was used as analyzer.

We have investigated the evolution of magnetic ordering as a function of temperature, magnetic field and real time through the resonant x-ray magnetic scattering. Various possibilities have been explored by measuring the representative magnetic reflections, such as (7 0 0) as a function of temperature, magnetic field, and real time. Some representative plots are given below. Intensities of all these curves have been normalized for varying incident photon flux. The resonance at the Co K-edge in the energy scan of (7 0 0) reflection in $\pi$-$\pi'$ channel confirms the magnetic origin of this reflection (Fig. 1(a)). The temperature dependent study of the integrated intensity of the spin density wave reflection (700) shows that the samples orders magnetically at ~ 24 K. It is evident from the temperature variation that there is a loss of magnetic intensity at temperatures below ~ 11K. The h-scans show that magnetic wave vector remains incommensurate. It is interesting to observe that a new magnetic peak (½ ½ 0) corresponding to an commensurate antiferromagnetic (CAFM) phase appears below ~ 11 K (Fig. 1). It is evident that the second magnetic phase (CAFM) appears at $T < 11$ K at the cost of the SDW phase.

Time dependence of the SDW peak profile (700) was investigated after cooling the sample in each study from 35 K (i.e. higher than the Néel temperature) to 5, 10 and 17 K under various applied magnetic field values of 0, 0.05, 0.15, 0.3, and 0.5 T. A clear dynamic evolution of the SDW phase is found at both 5 and 10 K i.e., at temperatures below 11 K when the CAFM phase is also present (Fig. 2). The time dynamics at 10 K is even observed under magnetic field.

The SDW peak is found to enhance with the increase of magnetic field up to ~ 3 Tesla (Fig. 3). However, the SDW phase disappears under $H \geq 4.5$ Tesla, indicating a magnetic field induced complete suppression of the SDW phase. We have also measured the profiles of the (700) reflection in the $\pi$-$\sigma'$ channel under magnetic field. We find that the $ab$ component moment is order of magnitude smaller than the $c$-component moment.

The results have been interpreted in the light of our recent neutron scattering study. Based on these results a manuscript is being written up for publication in an international journal.
Figure 1: (a) Resonance at the Co K-edge in the energy scan of (7 0 0) reflection in π-π' channel confirms the magnetic origin of this reflection. Energy scan presented here has not been corrected for the absorption. (b) Temperature dependence of the integrated intensity of the SDW reflection (700) under zero magnetic field in the π-π' channel. (Right): Temperature dependence of the integrated intensity of the CAFM reflection (½ ½ 0) under zero applied magnetic field.

Figure 2: Real time dependence of the SDW reflection (700) at 10 K (under H= 0 (left) and 0.5 T (right)) and 17 K (under H = 0, middle) in the π-π' channel. At each temperature and field, the profiles (h-scans) were measured for a total time duration of 2.5 hrs with 2.5 mins for each h-scan.

Figure 3: Magnetic field dependence of the SDW reflection (700) at 2 K in the π-π' channel (left) and in the π-σ' channel (right).

References