ESRF	**Experiment title:** Orbital-ordering in $LaCoO_3$ studied by resonant inelastic x-ray scattering	**Experiment number**: HE-1590
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Report:

We report here on the investigation of low-energy electronic excitations by resonant inelastic x-ray scattering (RIXS) in $LaCoO_3$, a candidate to orbital ordering. In a previous series of experiments on $LaCoO_3$, we had monitored the spin state transitions as function of temperature and pressure. In the present study, we focused on the resonant inelastic spectra in the intermediate spin (IS) state observed at room temperature and ambient pressure, where orbital ordering is reportedly predicted.

In the intermediate spin state, Co has a doubly degenerated electronic configuration $t_{2g}^5 e_g^1$. The Co sites become Jahn-Teller active and orbital ordering may arise through a cooperative Jahn-Teller mechanism. Recent calculations further indicate that the free energy of the system is minimized when orbital ordering is introduced in the IS state [1]. At high temperature, $LaCoO_3$ undergoes an insulator-metal transition, which leads to the disappearance of the orbitally-ordered phase. The purpose of the present experiment was to investigate the occurrence of an orbitally ordered phase and follow its evolution through the transition.

The experiment was carried out on the inelastic scattering beamline ID-16, using the existing RIXS spectrometer. Our primary aim was to obtain the best spectral resolution in the considered energy region. This was achieved by: i) narrowing the incident energy bandwidth down to ~100 meV thanks to a Si(440) channel-cut inserted after the main monochromator; ii) extending the spectrometer arm to 2 m to minimize the source size contribution to the resolution, and iii) using a high-resolution Si analyzer specifically designed for the Co K edge energy. The analyzer was prepared at the Université Paris 6 by ionically bonding a Si(533) wafer onto a spherically shaped glass blank with a 2-m radius. The glue-free technique ensures minimum strain, which mostly arises from the gluing process, thus a sharp resolution function. The total resolution was estimated at ~280 meV at 7.7 keV from the FWHM of the elastic peak obtained in a thin plastic foil.

The energy loss spectra were measured by tuning the incident energy (E_1) to the Co K-edge region, and scanning the emitted energy (E_2) within a few eV from the elastic (recombination) peak. The sample, a single crystal of $LaCoO_3$, was orientated in the beam to align the momentum transfer along the (111) or (110) directions. Typical RIXS spectra are shown on figure 1 with the incident energy tuned on (7.707 keV) and off (7.691 keV) resonance. A weak feature shows up in the foot of the elastic peak (arrow) around 0.35 eV energy transfer. This can be better visualized in the difference spectrum between on and off resonance, as illustrated in the inset to figure 1. Figure 2 shows the variation of intensity of the 0.35 eV feature as function of the incident energy. The excitation presents a broad resonance around the white-line position – energies above ~7710 eV could not be reached because of geometrical constraints related to the spectrometer setup.

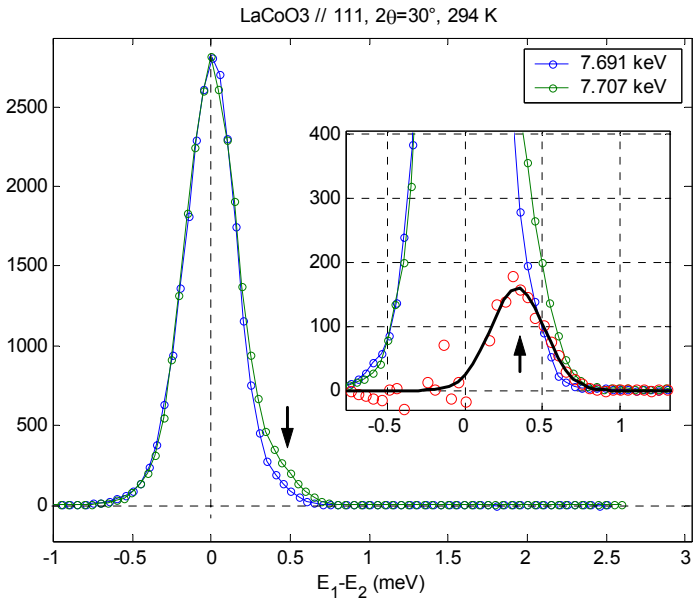

Figure 1 : RIXS spectra in $LaCoO_3$ on (green line) and off (blue line) resonance on a transfered energy scale. The on/off resonance difference reveals a clear excitation around 0.35 eV as shown in the inset (red circles). The black line is a Gaussian fit to the difference curve

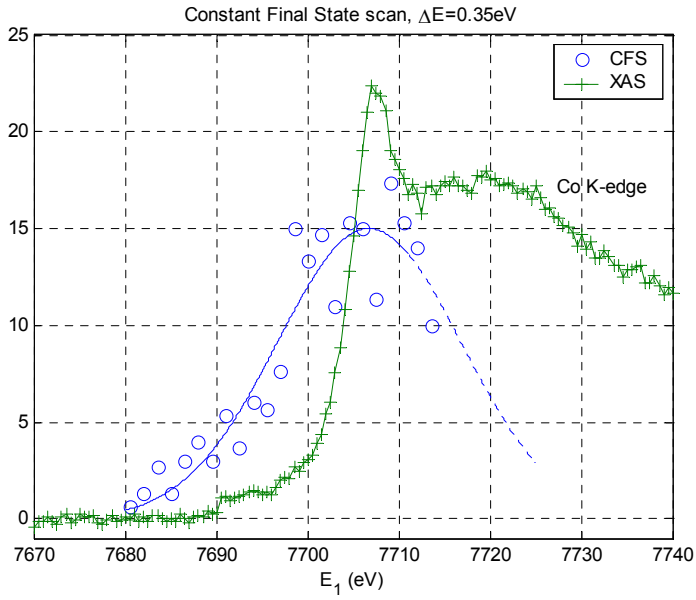

Figure 2 : Co K-edge absorption spectrum in $LaCoO_3$ in total fluorescence yield mode (green line). The intensity variation of the 0.35 eV excitation as function of the incident energy (also called constant final state scan) is indicated in blue. It shows a broad resonance around the white line energy.

Excitations related to orbital ordering were observed in the insulator compound $LaMnO_3$ at 2.5 eV energy transfer [2]. The lower energy of the observed excitation here is consistent with the semi-conducting character of $LaCoO_3$ in the intermediate spin state, which imposes an electronic gap (of the order of 0.1-0.2 eV) largely reduced compared to manganates. We did not notice any dispersion of the observed excitation either as function of the momentum transfer or when changing the sample orientation.

Unfortunately, we were not able to monitor the temperature dependence of the RIXS spectra as the sample was heated above the metalization temperature (500 K). Measurements in the oven turned out to be difficult because of the bulky sample environment. Complementary RIXS experiments are necessary at this point. Alternatively, we plan to carry out a temperature-dependent resonant diffraction experiment to assess the orbital-ordering hypothesis.

[1] M.A. Korotin et al., Phys. Rev. B **54**, 5309 (1996)
[2] T. Inami et al, Phys. Rev. B **67**, 45108 (2003)