



	<b>Experiment title:</b> Orbital and Spin-State order in layered cobaltites	<b>Experiment number:</b> HE-1819
<b>Beamline:</b> ID20	<b>Date of experiment:</b> from: 3-11-2004 to: 9-11-2004	<b>Date of report:</b> 23-2-2006
<b>Shifts:</b> 18	<b>Local contact(s):</b> Dr. L. Paolasini	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants (* indicates experimentalists):</b> Dr. U. Staub* Dr. A. M. Mulders* V. Scagnoli * Paul Scherrer Institut, Swiss Light Source, CH-5232 Villigen PSI		

Cobaltates ( $\text{RBaCo}_2\text{O}_{5+\delta}$ ) systems are particularly interesting since they display a large variety of magnetic and transport properties, which depend on the tunable oxygen concentration. It is believed that the magnetic and transport behaviors are driven by the mixed valence state of the cobalt ions and possible orbital ordering and therefore by the  $\text{Co}^{2+}/\text{Co}^{3+}$  ( $\text{Co}^{4+}/\text{Co}^{3+}$ ) ratio determined by the oxygen content of the material. Particularly interesting is the  $\delta=0.5$  oxygen concentration, where  $\text{RO}_x$  (R= rare earth ion) layers order into alternating filled and empty rows along the  $a$ -axis. This produces a significant change in the local environment of the Co ions, which differentiate in octahedral ( $\text{Co}_I$ ) and tetragonal ( $\text{Co}_{II}$ ) coordination. The nominal valence of the Co ions is expected to be 3+. At  $T_{\text{MI}}=350$  K, the material exhibit a metal-insulator (MI) transition associated with a change in the volume. In addition, susceptibility measurements indicate that a spin state transition occurs at  $T_{\text{MI}}$ . A ferromagnetic component appears below approximately 300K, which suddenly vanishes at 260 K may indicate a paramagnetic-ferromagnetic-antiferromagnetic transition [1].

Recently, a model for the magnetic structure has been proposed [1] for  $\text{GdBaCo}_2\text{O}_{5.5}$ , based on magnetization measurements, which suggests an Ising-like behavior for the spin system. The two inequivalent Co ions become either intermediate spin state (IS:  $t_{2g}^5 e_g^1$ ;  $S=1$  for  $\text{Co}_{II}$ ) or low spin state (LS:  $t_{2g}^6 e_g^0$ ;  $S=0$  for  $\text{Co}_I$ ). In the case of IS,  $d_{3z^2-x^2}$  or  $d_{x^2-y^2}$   $e_g$  orbitals can be occupied. Therefore, it has been proposed that the  $\text{Co}_{II}$  ions give raise to orbital ordering along the  $a$  and  $c$  axis direction. We performed resonant X-ray scattering on the Co K-edge on a  $\text{GdBaCo}_2\text{O}_{5.5}$  single crystal. This technique is well suited to obtain information on charge and orbital ordering of such perovskite systems [2-4].

We have measure the energy dependence, in the  $\sigma$ - $\sigma$  charge channel of a reflections probing along different directions (see for (70-1) figure 1) for both the  $\sigma$ - $\sigma$  and  $\sigma$ - $\pi$  polarizations including the corresponding azimuthal angle dependences. The strong azimuthal angle dependence in both channels indicate, that the orbital occupation is significantly changed when going through  $T_{\text{MI}}$ . A detailed analysis is in progress.

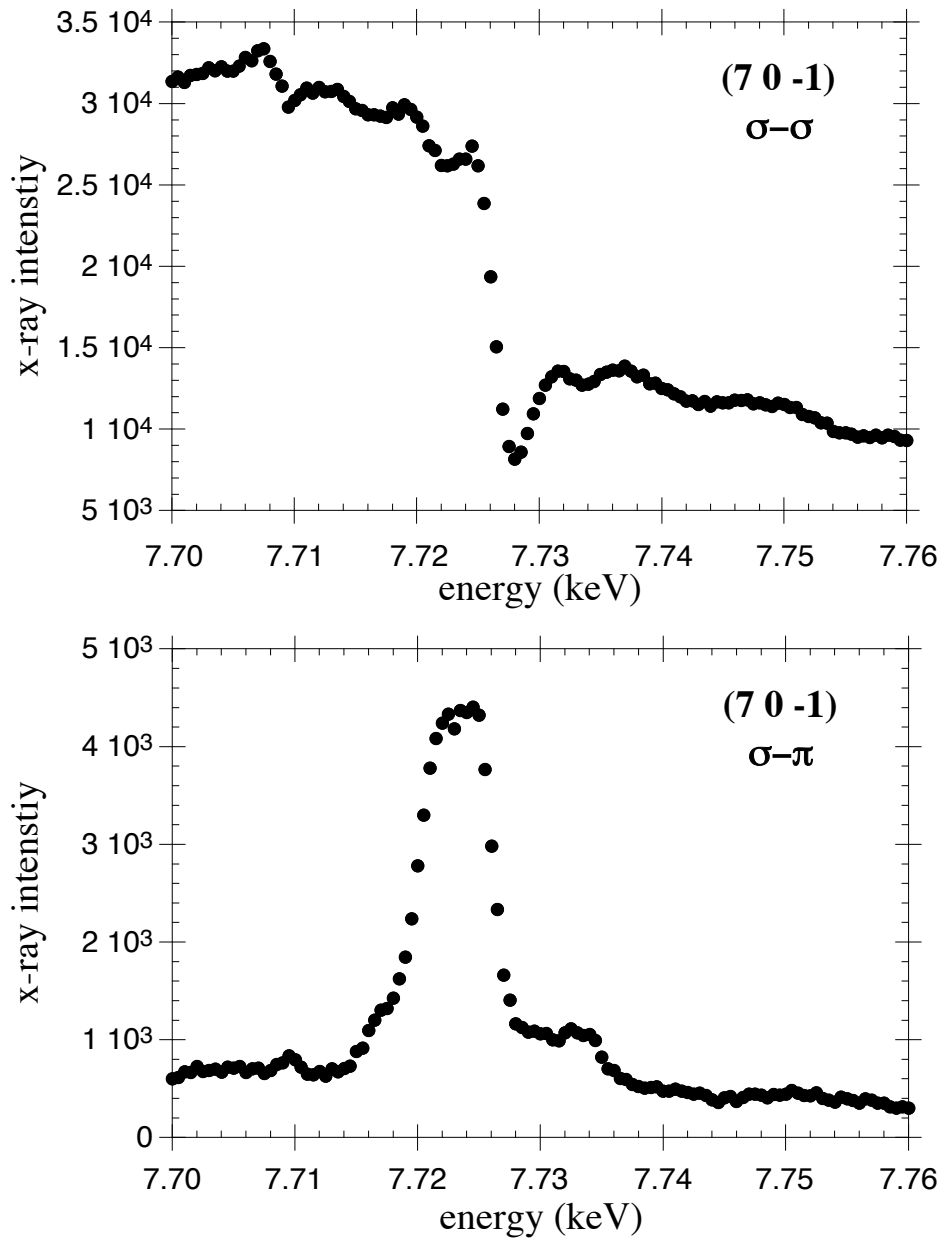


Fig. 1 shows the energy dependence of the 70-1 reflection in the two different polarization channels of  $\text{GdBaCo}_2\text{O}_{5.5}$  in the vicinity of the Co K-edge.

- [1] A.A. Taskin *et al*, Phys. Rev. Lett. **90**, 227201 (2003)
- [2] Y. Murakami *et al*, Phys. Rev. Lett. **81**, 582 (1998).
- [3] L. Paolasini *et al*, Phys. Rev. Lett. **82**, 4719 (1999).
- [4] U. Staub *et al*, Phys. Rev. Lett. **88**, 126402 (2002).