ESRF	**Experiment title:** *Unusual effect of crystal and magnetic structure transformation in the metastable $La_{0.5}Ba_{0.5}CoO_{2.87}$ and … modified by pulsed huge-field*	**Experiment number:** **HE-4996**
Beamline: **ID-22**	**Date of experiment:** from: **29.11.2022** to: **01.12.2022**	**Date of report:** 01.03.2023
Shifts: 3	**Local contact(s):** **Dr. Giorgia Confalonieri**	*Received at ESRF:*
Dr. Stanislav Savvin, ILL, France		

The cobaltites with perovskite structure are systems with fascinating transport and magnetic properties [1-11]. The parent compound $LaCoO_3$ (LCO) is a paramagnetic (PM) insulator at low temperature (LT). As temperature increases a gradual spin transition (ST) from a low-spin (LS, $t_{2g}^6e_g^0$) to intermediate (IS, $t_{2g}^5e_g^1$) or high (HS, $t_{2g}^4e_g^2$) spin-state occurs, accompanied with insulator to metal transition at ~550 K [1-5]. Previous studies showed that chemical substitutions in LCO can induce a long-range magnetic antiferromagnetic (AFM) or ferromagnetic (FM) order [2-10]. For example, low doped $La_{1-x}Ba_xCoO_{3-d}$ ($LBCO_{3-d}$) become to be AFM insulators [4,7,8,10]. One possible explanation for these phenomena is stabilization of the HS state. Surprisingly, the T_c at these compositions increases under applied pressure [7,8,10] while the origin of the magnetostructural phase transition is still unclear. In a medium doped oxygen deficit compound $La_{0.5}Ba_{0.5}CoO_{2.75}$ all cobalt atoms are in the oxidative state of 3+ and FM ordering is gradually suppressed upon temperature decrease. It leads to a large magnetic anisotropy (Fig.1a) inducing a giant magnetoresistance as well as transformation of crystal structure including the separation into two different pseudocubic phases for multi-valent $La_{0.5}Ba_{0.5}CoO_{2.87}$ (**Fig.1 and inset**).

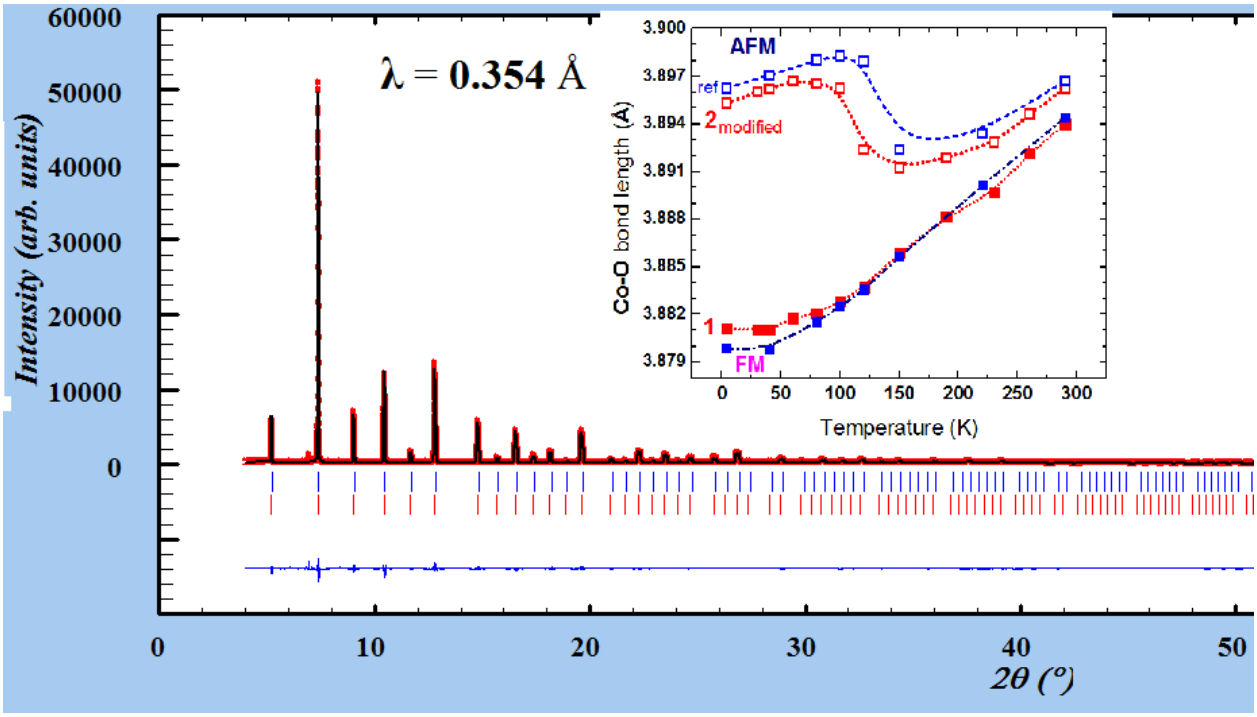

Fig. 1. Rietveld refinement of the x-ray powder diffraction pattern of $La_{0.5}Ba_{0.5}CoO_{2.87}$ at 2 K. Experimental data (open circles), refinement (solid line), and difference curve (solid line at the bottom). Tick marks indicate the calculated positions of the Bragg peaks. Inset shows the temperature dependence of the two Co-O bond length for pristine (blue color) and modified (red color) by pulsed 40 Tl.

Our high-resolution X-ray powder diffraction studies for pristine and modified $La_{0.5}Ba_{0.5}CoO_{2.87}$ showed both decrease anisotropy for two cubic Pm-3m structures associated with FM (corresponding to the smaller 2•Co_1-O bond length) and AFM (bigger 2•Co_2-O bond length) phases (**Fig.1 inset**) and as a consequence the decrease of the isotropic thermal parameters for Co and O (**Fig. 2a,b**).

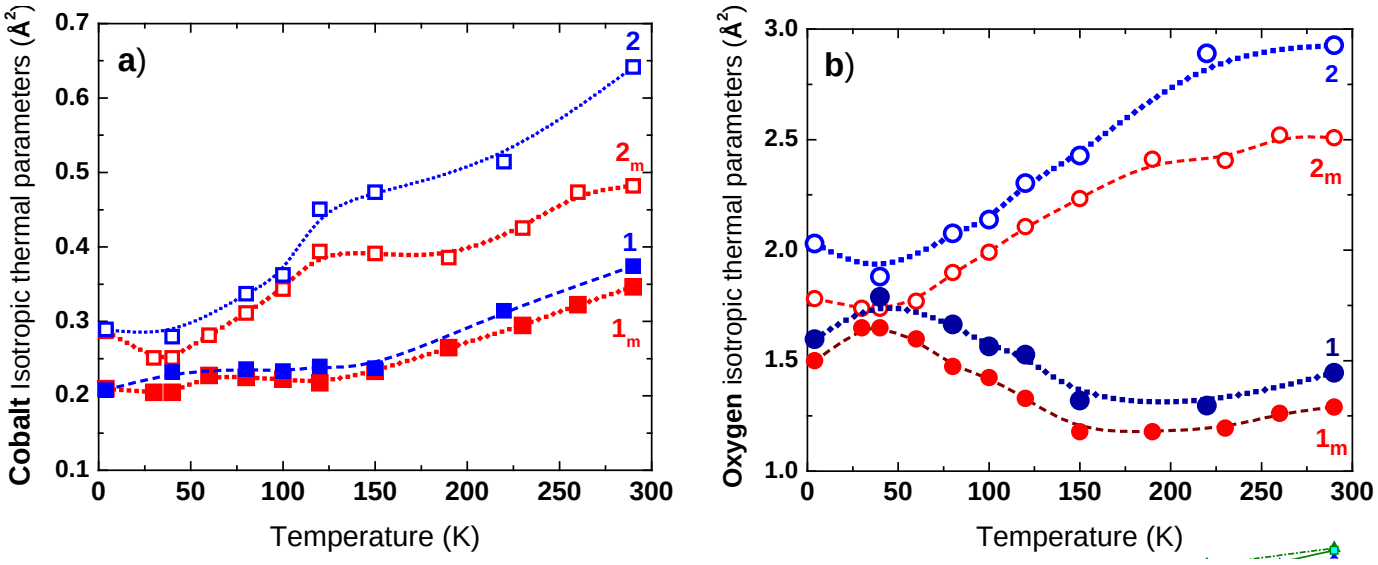

Fig. 2. Temperature dependence of isotropic ADP of Co (a) and O (b) for two Co-O bond length obtained by Rietveld analysis of the X-ray powder diffraction (XPD) data.

A small growth of the Co_1-O (or unit cell/2) below 50 K and essential decrease of the Co_2-O bond length especially in the range 100-120 K for modified one (**Fig.1 inset**) is probably connected with arrange of both the basic FM phase including chaotically reoriented magnetic moments within domain walls along of the main FM direction and smaller fraction in opposite(due to the difficulty reorientation) direction (i.e. AFM phase that corresponds to the HS state with ionic radius 0.61 Å). Such result is directly confirmed by decrease of Co and O isotropic thermal ADP especially in the temperature range 100-120 K (**Fig. 2**) for Co_2-O bond length (Fig.1), i.e. in the range of FM phase (**Fig. 3**) corresponding to the IS state with ionic radius 0.56 Å.

The temperature-dependent magnetization can be understood assuming a gradual change in the spin-state of cobalt ions with temperature decrease. The ST is accompanied by a decrease of the FM component and a growth of the AFM one, as evident from Fig. 3. An enhancement of the AFM interactions below 100 K is likely due to stabilization of the HS of the Co^{3+} ions.

It interesting to note that the influence of pulsed 40 Tesla field on $La_{0.5}Ba_{0.5}CoO_{2.87}$ transforms magnetic structure from AFM to FM state sharply increasing both magnetic ordering and T_c thus leading to the properties of a non-oxygen-deficient $La_{0.5}Ba_{0.5}CoO_3$ composition (**Fig.3**).

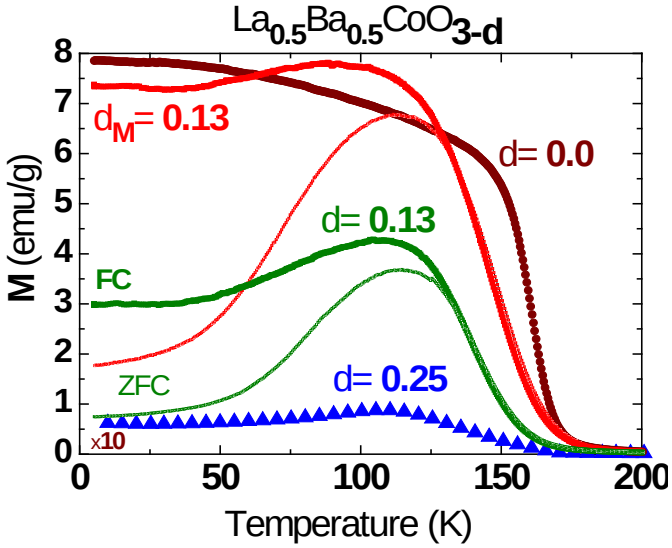

Fig. 3. M vs T magnetization for $La_{0.5}Ba_{0.5}CoO_{3-d}$ (d = 0.0; 0.13 and 0.25) in comparison with $La_{0.5}Ba_{0.5}CoO_{2.87}$ modified by pulsed magnetic field 40 Tl.