

# Experimental report

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**Proposal:** CRG-2656

**Council:** 4/2019

**Title:** Investigating high-TC spiral orders in  $\text{YBa}(\text{Cu}_{1-x}\text{Co}_x)\text{FeO}_5$  multiferroics with enhanced anisotropy and spin-orbit coupling

**Research area:**

This proposal is a new proposal

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**Experimental team:** Arnau ROMAGUERA CAMPS  
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**Samples:**  $\text{YBa}(\text{Cu}_{1-x}\text{Co}_x)\text{FeO}_5$

Instrument	Requested days	Allocated days	From	To
D1B	3	3	17/01/2020	20/01/2020

**Abstract:**

# Experimental report

**CRG-2656**

**17<sup>th</sup>-20<sup>th</sup> January 2020**

**Experiment title:**

Investigating high-*tc* spiral orders in YBa(Cu<sub>1-x</sub>Co<sub>x</sub>)FeO<sub>5</sub> multiferroics with enhanced anisotropy and spin-orbit coupling

**Participants:** Xiaodong Zhang, Arnau Romaguera Camps, Jose Luis Garcia Muñoz, Oscar Fabelo

## Abstract

The search of magnetoelectric multiferroic (MF) materials, where magnetic order combines with ferroelectricity (FE) and both are coupled to each other is an issue of keen interest in condensed matter physics and in spin-related emerging communication technologies. However, their low-magnetic ordering temperatures (typically < 100K) critically restrict the potential uses of these materials for spintronics and low-power magnetoelectric devices. YBaCuFeO<sub>5</sub> (YBCFO) displays magnetism-driven ferroelectricity at unexpectedly high temperatures (above RT), being one of the best candidates to switchable, magnetism-driven ferroelectricity at zero field. The stability range of its spiral phase can be extended far beyond room temperature by manipulating the Cu/Fe chemical disorder in the bipyramids and by chemical pressure. As an alternative strategy to upgrade its multiferroic properties, the substitution of Cu<sup>2+</sup> by Co<sup>2+</sup> is proving to be one of the most interesting. The substitution increases the magnetic anisotropy in the system and the spin-lattice coupling thanks to the significant orbital moment contribution from Co<sup>2+</sup> ions, with direct incidence on the stability of the spiral and the magnetoelectric coupling. All of it preserving the strong and key Fe<sup>3+</sup>-Fe<sup>3+</sup> magnetic interaction in the system and the oxidation state of the different atoms.

## Experimental part

There were no problems with the beam, cryofurnace, diffractometer during the measurements, and neutron patterns could be collected within the desired temperature range, between 10 and 500K.

In this experiment, we measured 7 samples of YBaCu<sub>1-x</sub>Co<sub>x</sub>FeO<sub>5</sub> with *x*= 6.5%, 7.5%, 8%, 9%, 11%, 12% and 25%. All samples have been prepared using solid-state reaction method and applying the same cooling method. In general, all samples were measured in dynamic mode by means of temperature ramps with heating rates between 1.4 and 3K/min, from 10-500K using wavelength  $\lambda=2.52\text{\AA}$ . In addition, all samples were measured with longer counting times at different fixed selected temperatures. In most cases at T=10K, 300K and 500K with the same wavelength. The last temperature T=500K corresponds to the paramagnetic state.

According to the experimental data, as shown in Fig.1, we have detected 3 different phases in YBaCu<sub>1-x</sub>Co<sub>x</sub>FeO<sub>5</sub> which *x*= 6.5%, 8%, 11% and 12% within the temperature range of our measurements (10-500K): (i) the commensurate (CM) AF1 collinear phase which is characterized by the magnetic propagation vector  $k = (1/2 \ 1/2 \ 1/2)$ ; (ii) the commensurate (CM) AF1' collinear phase which is

characterized by the magnetic propagation vector  $k = (1/2 \ 1/2 \ 0)$ ; (iii) the paramagnetic phase (see Fig.1 (b)). In some compositions (e.g.  $x=7.5\%$ ) an additional different phase was detected, which is the incommensurate (ICM) phase characterized by the magnetic propagation vector  $k = (1/2 \ 1/2 \ 0 \pm q)$  (see Fig.1 (a)). Different behavior was observed in sample  $x=25\%$ , where only 2 phases were detected: (i) the commensurate (CM) AF1' collinear phase which is characterized by the magnetic propagation vector  $k = (1/2 \ 1/2 \ 0)$ ; (ii) the paramagnetic phase (see Fig.1 (c)).

In the samples, phase transitions involving the appearance of new Bragg reflections are clearly observable at  $T_{N1}$  and  $T_{N1'}$ , as shown in Fig. 1 (a) for one of the compositions. The successive magnetic transition temperatures were determined for each sample and is plotted in Fig. 2. Some results from previous preliminary measurements were used here in order to show the evolution. It indicates that  $T_{N1}$  decreases with increasing Co content, while  $T_{N1'}$  shows opposite trend and increases with increasing Co content. A strong tuning of  $T_{N1}$  and  $T_{N1'}$  is possible by introducing Co to the material.

The main magnetic peaks of varying Co content were identified, and their evolution with increasing Co content have been map out and explained. By means of Rietveld refinements the temperature evolution of the neutron patterns for each composition has been fully analyzed and the evolution of the main magnetic parameters has been extracted. In addition, main structural information has been extracted from the neutron patterns collected at 500K in paramagnetic state. The results of these analyses will be conveniently published.

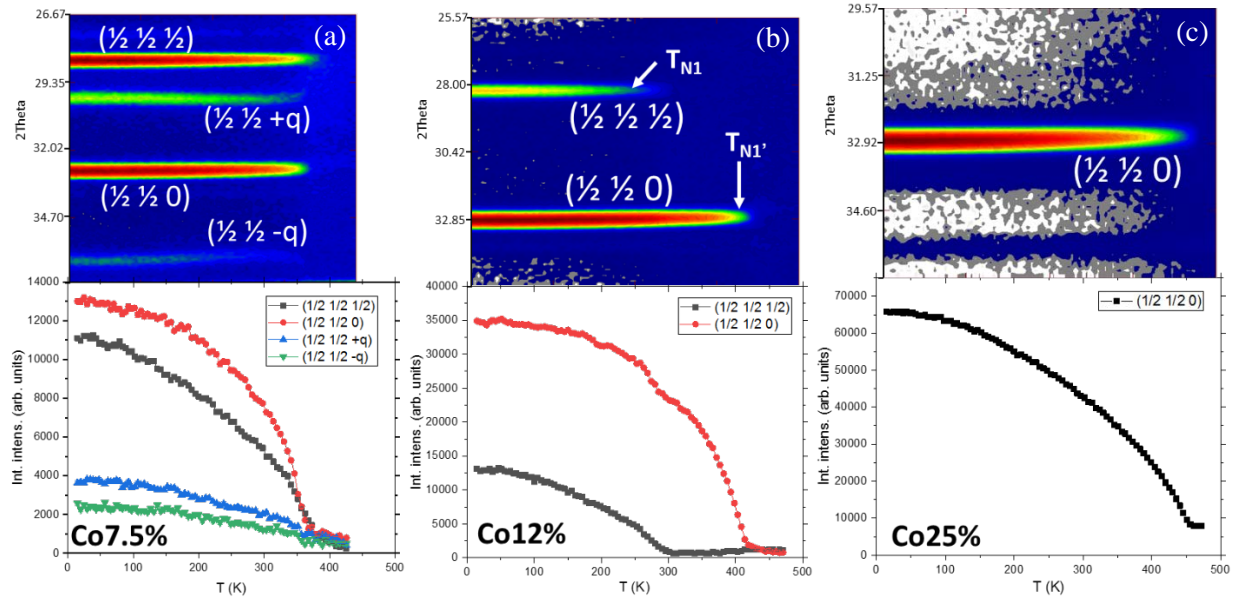


Fig. 1. Contour map and integrated intensity showing the temperature dependence of the position and the intensities of the magnetic Bragg reflection for YBaCu<sub>1-x</sub>Co<sub>x</sub>FeO<sub>5</sub> with  $x = 7.5\%$ ,  $12\%$  and  $25\%$ .

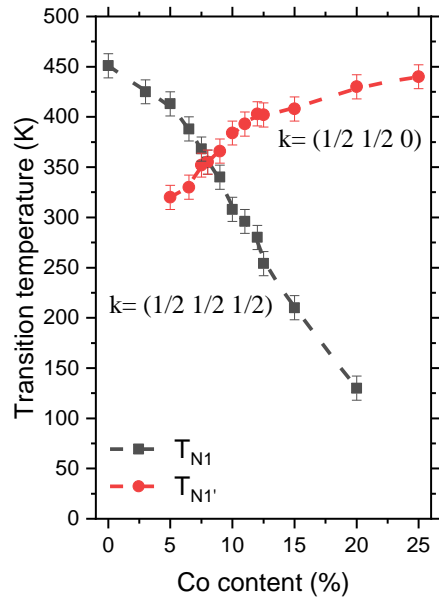


Fig. 2. The ordering temperatures  $T_{N1}$  and  $T_{N1'}$  as a function of Co content.