Experimental report

20/11/2019

Proposal: **CRG-2655** **Council:** 4/2019

Title: Neutron study of selective Mn substitution in Fe pyramids of high-TC YBaCu(Fe1-xMnx)O5 spiral muiltiferroic

Research area:

This proposal is a new proposal

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Samples: YBaCu(Fe1-xMnx)O5+d

Instrument	Requested days	Allocated days	From	To
D1B	3	3	13/09/2019	16/09/2019

Abstract:

Experimental report

CRG-2655

13th-16th September 2019

Experimental title:

Neutron study of selective Mn substitution in Fe pyramids of high-TC $YBaCu(Fe_{1-x}Mn_x)O_5$ spiral multifferoic

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Abstract

The low-magnetic ordering temperatures (typically <100 K) critically restrict the potential uses of magnetoelectric multiferroics for spintronics and low-power magnetoelectric devices [1-2]. $YBaCuFeO_5$ (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 [3-4]. 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 [4-5]. As an alternative strategy to upgrade its multiferroic properties, the selective partial substitution of very symmetric Fe^{3+} by Jahn-Teller (JT) Mn^{3+} ions in Fe-pyramids offers very interesting possibilities.

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 aimed to measure two sets of samples. The first set of samples included six different compositions of $YBaCu(Fe_{1-x}Mn_x)O_5$ with x=0%, 1%, 5%, 10%, 15% and 20% of Mn. All samples have been synthesized following the same cooling condition (C1). The other set of samples are $YBaCuFe_{1-x}Mn_xO_5$ (x=0.10) which were cooled with three different cooling rate conditions (C1, C2 and C3). 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 λ=2.52Å. In addition, all the 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 corresponds to the paramagnetic state.

Three different types of phases were detected within the measurement temperature range (10-500K): (i) the incommensurate (ICM) AF2 spiral phase associated to the generation of electrical polarization and the appearance of ferroelectricity, characterized by the magnetic propagation vector k = (1/2 1/2 1/2 ± q); (ii) the commensurate (CM) AF1 collinear phase which is characterized by the magnetic propagation vector k = (1/2 1/2 1/2); (iii) the paramagnetic phase.

In the samples, two phase transitions involving the appearance of new Bragg reflections are clearly observable at T_{N2} and T_{N1}, as shown in Fig. 1 for one of the compositions. Table 1 shows the ordering temperatures of $YBaCu(Fe_{1-x}Mn_x)O_5$ (x=0.10) prepared by three different cooling approaches. T_{N2} increases approximately 20K from C1 to C2. Chemical disorder is supposed to be at the origin of this increasing transition temperature.

From full range temperature dependent neutron data, the positions and the intensities of the CM and ICM satellites for the $YBaCuFe_{1-x}Mn_xO_5$ series samples were explored in detail. First, we confirmed that the spiral phase is rather stable under substitution of Fe by Mn. The successive magnetic transition temperatures were determined the for each sample (see Fig. 2.). It shows that both T_{N1} and T_{N2} decrease with increasing Mn content, but they do with very different slopes. A strong tuning of T_{N2} is possible by introducing Mn.

Fig. 3 plots the main magnetic peaks of each sample cooled with C1 (Fig. 3.). Their evolution with increasing Mn content is shown. The temperature evolution of the neutron patterns for each composition is being fully analyzed in order to get the evolution of the main magnetic parameters. Main structural information is being extracted from the neutron patterns collected at 500K in the paramagnetic state. Of importance it is the variation of the chemical disorder (Fe/Cu distribution respect to the ordered structure). The results of these analyses will be conveniently published once they are finished.

The integrated intensity of magnetic collinear peak (see Fig. 4.) for this series samples was depicted as well, which indicate that they have different chemical disorder due to distinct cooling approaches.

References

[1] S.-W. Cheong and M. Mostovoy, Nat. Mater. 6, 13 (2007)
[2] T. Kimura et al. Nat. Mater. 7, 291 (2008)
[3] M. Morin et al. Phys. Rev. B 91, 064408 (2015)
[4] M. Morin et al, Nature Communications 7, 13758 (2016)
[5] Shang et al., Science Adv. 4(10), eaau6386 (2018)

Table 1. The ordering temperatures of Mn10% doped YBCFO samples prepared by 3 different cooling procedures.

sample	TN1 (K)	TN2 (K)
Mn10%-YBCFO_C1	400 (6)	156 (6)
Mn10%-YBCFO_C2	400 (6)	165 (6)
Mn10%-YBCFO_C3	396 (6)	172 (6)

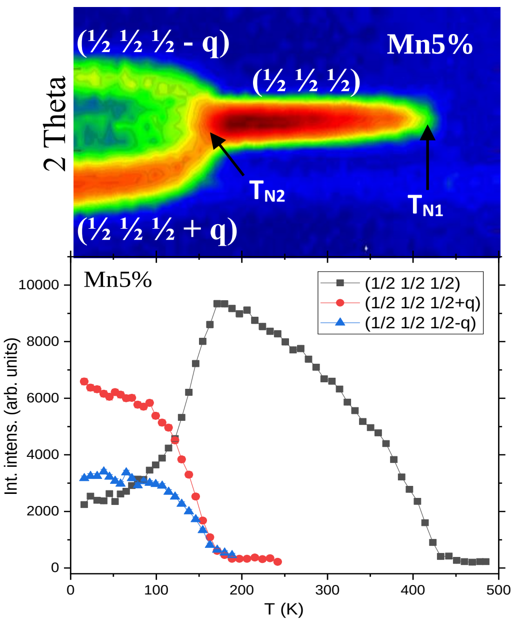

Fig. 1. Contour map and integrated intensity showing the temperature dependence of the position and the intensities of the magnetic Bragg reflection (½ ½ ½) associated to the high-temperature collinear antiferromagnetic phase, and the (½ ½ ½ ± q) satellites of the low-temperature magnetic spiral phase for $YBaCu(Fe_{1-x}Mn_x)O_5$ (x=0.05).

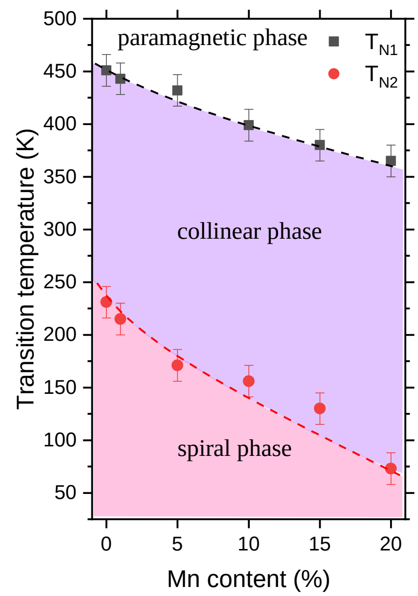

Fig. 2. The phase diagram showing the ordering temperatures T_{N1} and T_{N2} as a function of Mn content.

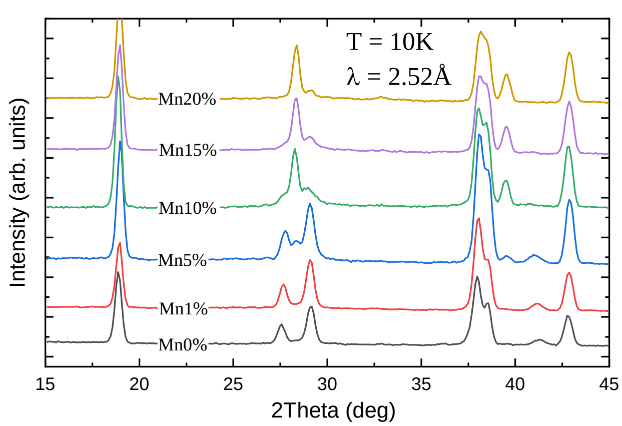

Fig. 3. The ground state showing main magnetic peaks evolution for Mn samples. Data collected at T=10K.

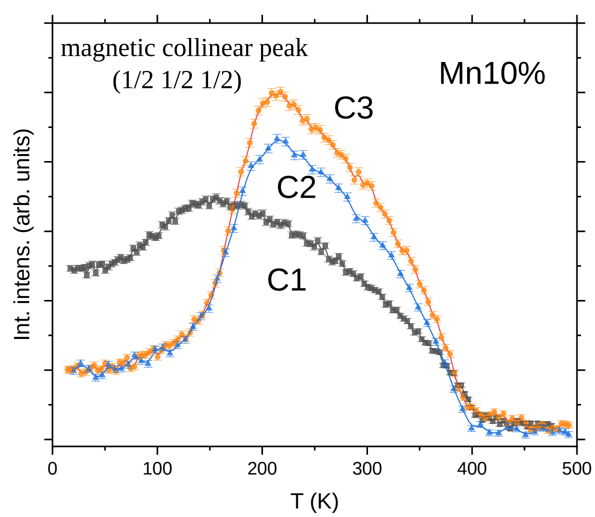

Fig. 4. The integrated intensity of (½ ½ ½) magnetic peak (collinear order) for $YBaCu(Fe_{1-x}Mn_x)O_5$ (x= 0.10) prepared with 3 different cooling methods.