# Experimental report

13/02/2019

**Proposal:** 5-41-920 **Council:** 4/2017

**Title:** Single crystal neutron diffractioninvestigation of multiferroic Lu2CoMnO6

**Research area:** Physics

**This proposal is a new proposal**


**Main proposer:** Tapan CHATTERJI

**Experimental team:** Tapan CHATTERJI

**Local contacts:** Bachir OULADDIAF


**Samples:** Lu2CoMnO6

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| D10 | 7 | 7 | 31/05/2018 | 07/06/2018 |


## Abstract:

Double perovskites are a fascinating material [1-5] class with a lot of functional properties. Multiferroicity, magneotcapacitance, magnetoresistance, different magnetic ground states including spin-glass and three dimensional quantum magnetism have been either experimentally observed or theoretically proposed for these materials. Lu2CoMnO6 (LCMO) crystallizes in a monoclinic P 21/n double-perovskite structure with a unit cell of a = 0.516 nm,b = 0.554 nm, and c = 0.742 nm. Co2+ and Mn4+ ions are alternatingly located in corner-shared octahedral environments. It has drawn an interestdue to its newly found multiferroicity in the previous workpolycrystalline materials. We propose single crystal neutron diffraction investigation on these samples. The investigation can lead to the microscopic origin of anisotropic behavior of the multiferroic properties of Lu2CoMnO6. We are interested to determine the magnetic ordering and phase transition and also ferroelectric properties and therefore we need to measure nuclear and magnetic reflections very accurately. We request 7 days of neutron beam time on D10 for the proposed experiment.

# Single crystal neutron diffraction study of the magnetic structure of multiferroic $Lu_2CoMnO_6$

**Introduction:**

Double perovskites are a fascinating material [1-5] class with a lot of functional properties. Multiferroicity, magneotcapacitance, magnetoresistance, different magnetic ground states including spin-glass and three dimensional quantum magnetism have been either experimentally observed or theoretically proposed for these materials. One of the famous double perovskite is the $Sr_2FeMoO_6$ discovered, where high magnetoresistance (MR) was reported. A strong correlation between the observed MR and the inherent crystallographic *antisite* disorder was proposed in that compound. The *antisite* disorder – where the transition metal atoms occupy the crystallographic positions interchangeably – is found to be an ubiquitous presence in most of the double perovskite compounds and play an important role in the magnetism of these materials. Theoretically proposed (using the Goodenough-Kanamori rules) to behave as long-range ordered magnets, the double perovskites often display strong signs of disordered magnetism or short-range magnetic order primarily due to the presence of *antisite* disorder.

Here we report the results of our single crystal neutron diffraction investigation on the double perovskite $Lu_2CoMnO_6$ (LCMO). $Lu_2CoMnO_6$ crystallizes in a monoclinic P $2_1/n$ double-perovskite structure with a unit cell of a = 5.16 Å, b = 5.54 Å, = 742 Å and b = 90.08 deg. $Co^{2+}$ and $Mn^{4+}$ ions are alternatingly located in corner-shared octahedral environments.

**Experimental results:**

The monochromatic single crystal diffraction experiments have been performed on the four-circle triple axis spectrometer D10 at the ILL using a pyrolitic graphite monochromator with a wavelength of 2.36 Å and with $\lambda$=1.26 Å using the Cu monochromator. A set of magnetic and nuclear reflections was measured at 2K. The propagation vector of magnetic structure was determined to be k= [0.06, 0.02, 0.5] by the Laue diffraction experiment. This was also confirmed by the monochromatic single crystal diffraction performed on D10, leading to broad unresolved magnetic peaks corresponding to the contribution from different stars of the propagation vectors (different magnetic k-domains).

A set of 210 magnetic and 270 nuclear reflections have been measured at 2K. The refinement of the crystal structure using the $P2_1/n$ space group reported in reference [1] where the 2c site is occupied by 91%of Co and 9% of Mn atoms and the 2d site is occupied by 94 % of Mn and 6 % of Co atoms, leads to a very bad agreement between observation and calculations giving an RF2 factor of 21 %. However, the refinement is much improved when refining the occupancy factors of both sites. The RF2 factor reaches then 7.6% with 53(2) % of Co atoms and 47(2) % of Mn atoms are located at the 2c site and 56(2) % of Mn atoms and 43(2) % of Co atoms occupy the 2d site which indicates an almost random distibution between Mn and Co atoms within the two sites. Figure 1 shows the calculated and observed integrated intensities of nuclear reflections.

The magnetic reflections being not resolved, the integrated intensity of each magnetic reflection used in the refinement, is considered as a cluster of four magnetic satellites generated by the propagation vector ±k and its star. Thus 210 clusters of magnetic reflections have been used in the refinement of the magnetic structure. The magnetic sites 2c and 2d split into two orbits under the propagation vector k leading to four magnetic sublattices without any symmetry constrains. Several magnetic configurations have been used in the refinements. Only amplitude modulation arrangement with moments along the c-axis gives good agreement between observation and calculation. The reliability factor RF2 factor reaches 8.9 %. The refined magnetic moments are 1.7(1) $\mu_B$ and 1.3(1) $\mu_B$ on the 2c and the 2d sites respectively leading to a saturation moment of 6 $\mu_B$/ f.u. in excellent agreement with the deduced saturated

moment of 6 $\mu_B$/ f.u. at 6T [2,5]. Figure 2 shows the calculated and observed integrated intensities of magnetic reflections.

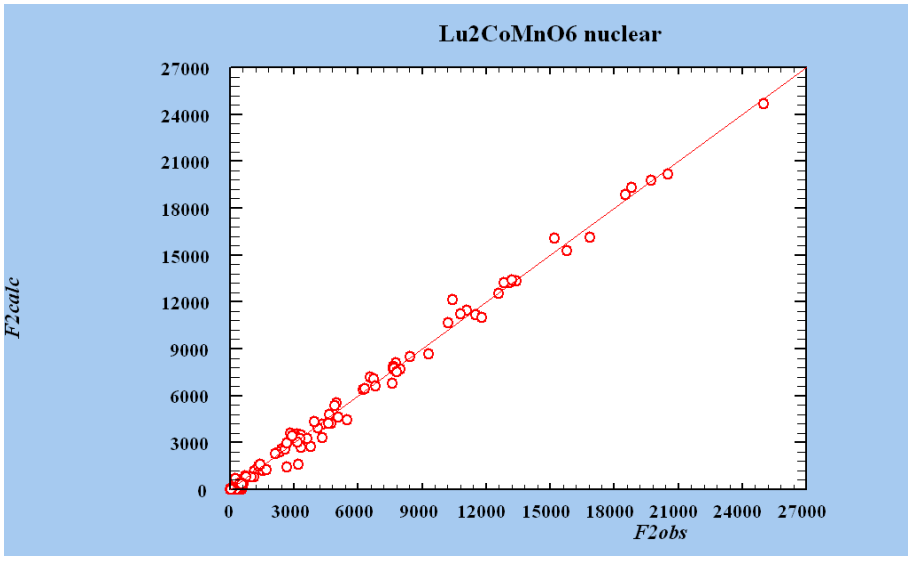


Figure 1: Observed and calculated nuclear intensities

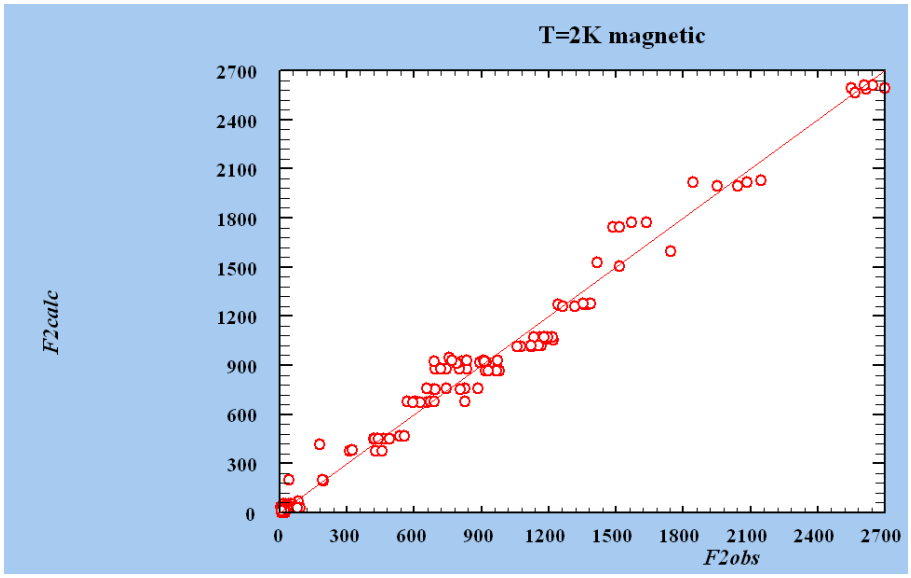


Figure 2: Observed and calculated magnetic intensities

The magnetic arrangement can be considered as four spin waves with long periods about 130 Å and 550 Å along a and b axis, respectively. However the unit cell being doubled along the c-axis, the ++-- spin arrangement is then obtained along the c-axis as proposed in [1]. Figure 3 shows the deduced magnetic structure.

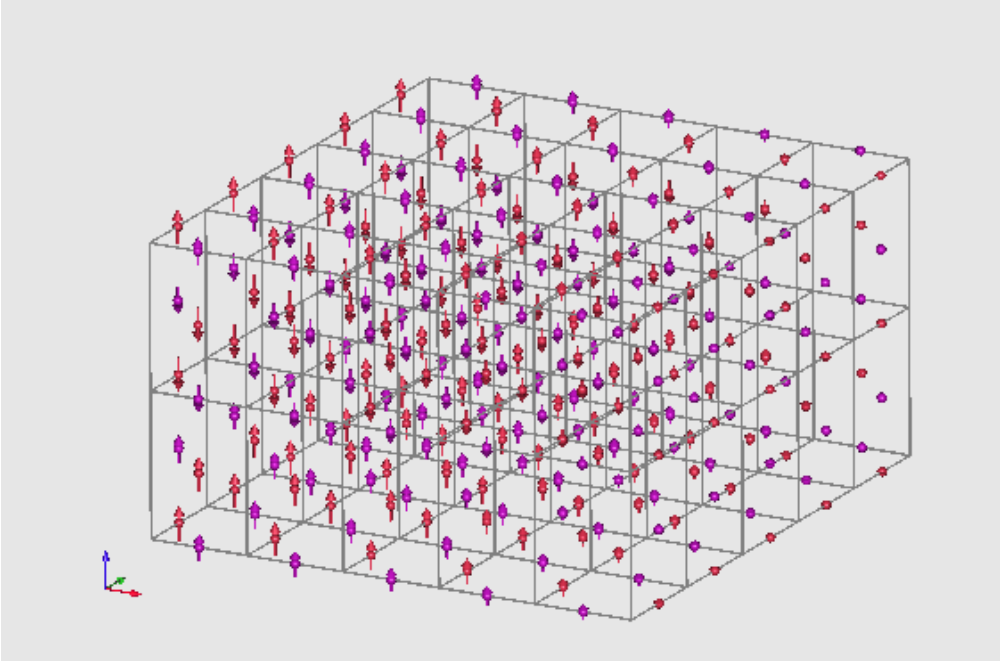

Figure 3: Long periods magnetic structure at 2K deduced from the neutron diffraction experiments.

**References:**


[1] S. Yanez-Vilar et al. Phys. Rev. B 84, 134427 (2011).
[2] M.K. Kim et al., J. Phys.: Condens. Matter 27, 426002 (2015)
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[5] N. Lee et al., Appl. Phys. Lett. 104, 112907 (2014)