Experimental report

12/09/2022

Proposal: 5-54-375 **Council:** 4/2021

Title: Engineering of long range couplingin manganate superlattices by design of spacer layer.

Research area: Materials

This proposal is a continuation of 5-54-269

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Experimental team: Laura GUASCO
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Samples: La0.2Sr0.8MnO3-x
La0.5Sr0.5MnO3
La0.6Sr0.4MnO3

Instrument	Requested days	Allocated days	From	To
SUPERADAM	7	5	08/07/2021	13/07/2021

Abstract:

Interfaces made of strongly correlated oxides have shown unexpected physical properties, such as the exchange bias, proximity effects, charge transfer, spontaneous mangetic reversal, exchange springs and orbital reconstruction. Given the complexity of its structural and magnetic phase diagram, LSMO offers a wide range of tunable properties that we can stack into heterostructures. We synthesized all manganate superlattices with a large gradient in hole doping by tuning the Sr-doping layer-by-layer. In previous PNR experiments, we observed a doubled non-collinear magnetic structure at intermediate spacer thickness, temperature and fields. We believe that this non-collinear magnetic phase is linked to the presence of highly canted spins at the interfaces. The aim of this proposal is to understand the magnetic nature of the spacer by a comparative study of SLs and single layer LSMO of different Sr-doping. This study will help to shed light on the role of the spacer magnetic ordering in the formation of the global doubled magnetic structure.

Experimental report on proposal 5-54-375

Dates: 8.07.2021-13.07.2021

Experimental team: L. Guasco (MPI FKF), Y. Khaydukov (MPI FKF).

Single crystal manganite thin films of nominal composition [130 ML $La_{0.5}Sr_{0.5}MnO_3$ (x=0.5)] +Pd (5nm) (Fig.1a) were deposited on LSAT substrates with ozone assisted molecular beam epitaxy (MBE) of MPI-FKF of Stuttgart. The layer by layer deposition was controlled in situ by RHEED measurement, confirming the stack of epitaxial layers. Samples of this composition were grown to mimic the intermediate effective hole doping expected for depth dependent Sr-doped heterostructures with the nominal composition $10x[9xLa_{0.6}Sr_{0.4}MnO_3+5xLa_{0.2}Sr_{0.8}MnO_3]$, previously studied elsewhere.

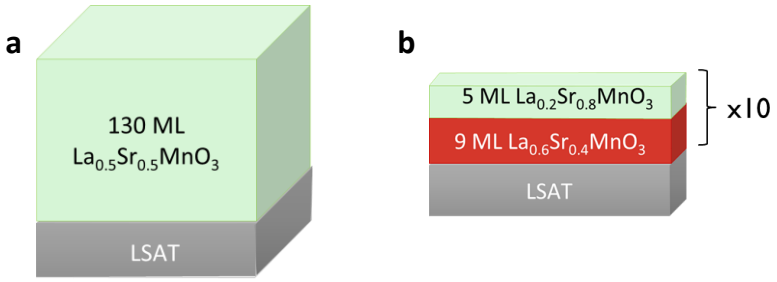

Figure 1: a. Sketch of single layer film of composition $La_{0.5}Sr_{0.5}MnO_3$; **b.** sketch of the previously studied $10x[9xLa_{0.6}Sr_{0.4}MnO_3+5xLa_{0.2}Sr_{0.8}MnO_3]$ heterostructure.

According to previous literature, a Sr-doping x=0.5 shows canted A-type antiferromagnetic (AF) ordering with Neel temperature above 200 K. The aim of this experiment was to confirm the A-type AF with combined polarized neutron diffraction (PND) and reflectometry (PNR).

The sample was measured in full polarized mode in a Q range between 0.005 and 0.1 $Å^{-1}$ for the PNR mode, and up to 0.86 $Å^{-1}$ for the PND mode in a range of temperatures between 5 and 295 K. The sample was first magnetized in the maximum field of 5 kOe and cooled in such field down to 5K. Successively the temperature was gradually increased and the field released, in order to study the field dependent magnetic properties.

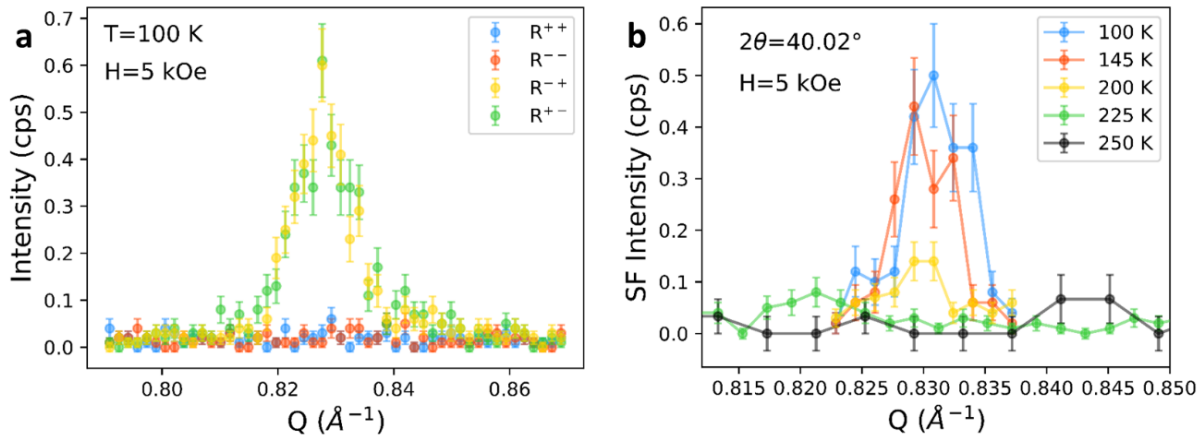

Figure 2: a. PND measurements at 100 K and external field 5 kOe, showing the emergence of the magnetic peak at ½ of the structural Bragg peak. **b.** Temperature dependence of SF scattering in PND mode.

Starting from the PND, we found a peak corresponding to the (00½) magnetic reflection at 0.83 $Å^{-1}$ in the SF channel exclusively, as shown in Figure 2a. The temperature dependence of this

feature was studied by measuring spin flip scattering while varying the temperature scanning around the Q position corresponding to the structural Bragg peak, as shown in Fig.2b.

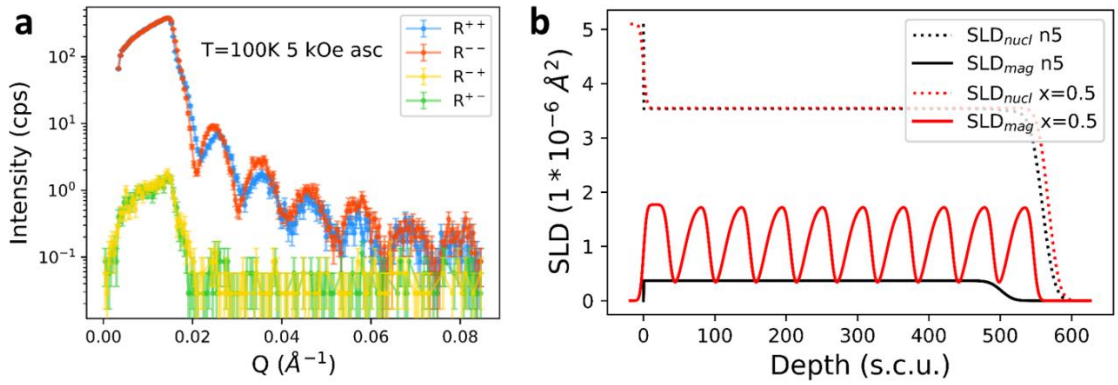

Figure 3: a. PNR measurements at 100 K and external field 5 kOe, showing the splitting between the two non-spin flip channels. **b.** Nuclear (dotted line) and magnetic (solid line) SLD profiles for the single film x=0.5, corresponding to the sketch in Figure 1a (black), and to the superlattice represented in Figure 1b (red).

In order to confirm the canting of spins we measured in the same temperature and field conditions the PNR curve of such single layer. Fig.3a shows the full polarization analysis performed at 100 K and 5 kOe, showing clear spin asymmetry between the R++ and R— channels. This spin asymmetry corresponds to a net magnetic moment of 0.7 μ_B/Mn atom, obtained from fitting of the experimental curves. The corresponding depth profile of the nuclear and magnetic scattering length density (SLD) are shown as black lines in Figure 3b.

The comparison between the single layer depth profile (black lines) and the heterostructure one (red lines) obtained in previous experiments from the sample schematized in Figure 1b, allowed to confirm the presence of canted AF in our superlattices.

The temperature dependence study of canted AF phase allowed to highlight the crucial role of the interlayer magnetism in the realization of long range non collinear magnetic structure observed in the heterostructure.

This work was recently published in the following work:

Guasco, L., Khaydukov, Y., Kim, G., Keller, T., Vorobiev, A., Devishvili, A., Wochner, P., Christiani, G., Logvenov, G., Keimer, B., Emergent Magnetic Fan Structures in Manganite Homojunction Arrays. *Adv. Mater.* 2022, 34, 2202971.
https://doi.org/10.1002/adma.202202971