	Experiment title: ATOMIC-SCALE DESIGN OF MAGNETIZATION PROFILES IN MANGANITE "DEAD LAYERS"	**Experiment number**: HE 3110
Beamline: ID08	**Date of experiment**: from: 9/2/2010 to: 18/2/2010	**Date of report**: 1/3/2010
Shifts: 18	**Local contact(s)**: F. Yakhou-Harris	*Received at ESRF:*

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Report:

During beamtime HE3110 at ID08 (completed 2 weeks ago) we successfully measured a complete set of XRMS and XMCD spectra on four $La_{1-x}Sr_xMnO_3$ (LSMO) (001), x=0.35 samples at four temperatures between 20-300K. The data is sufficient for analysis of the temperature evolution of the magnetization profiles at the interfaces, as described in Refs. [1] and [2] and as outlined in proposal HE 3110. Preliminary analysis indicates surprising results, as described below.

The samples were prepared at the Oxide MBE chamber at TASC Lab specifically for this beamtime, each with different atomic stacking sequences and doping levels at the interfaces [3]; sample 182 was a capped thick film (80 unit cells (uc), i.e., effectively "infinite" thickness for the measurements reported here), and samples 183, 184 and 185 were capped ultrathin films (~9 unit cells). The cap material was identical to the substrate, $SrTiO_3$, so that the "dead layer" here exists both at the manganite/titanate substrate/film and film/cap interfaces. The "atomic-scale design" imposed at the interfaces adjusts the doping level of the interfacial MnO_2 plane, by surrounding this BO_2 plane by two identical AO layers with a doping level different than the bulk x=0.35 (optimal "bulk" doping). The doping levels chosen at the interface ranged from x=0 ("underdoped") to x=0.5 ("overdoped"), and were previously characterized by EELS measurements (Ref. [3]) so that the effect of the doping profiles on Mn and Ti valences across the interface are known. The goal of HE3110 was to measure the magnetization profiles at the interfaces; they are expected to depend strongly on the interfacial doping profile, since T_{Curie} for ultrathin (~10uc) layers depends strongly on the interfacial doping, as well the measured differences in the XRMS spectra versus incidence angle seen in preliminary data taken at Elettra (Trieste).

The success of this beamtime was due to the high photon flux of the Dragon undulator/monochromator optics, so that we were able to measure reflectivity with excellent signal-to-noise ratio up to an incidence angle of θ=45º. The large range of incidence angle is necessary to allow unique determination of the magnetization profile, as described by our previous modeling of XRMS spectra (see in Fig. 3 of proposal HE3110 and Ref. [2]). This modeling shows that the depth of the midpoint of the magnetization profile (i.e., "dead layer" thickness) can be inferred from the first inversion angle of the XRMS, while any gradient to the profile can only be determined by fitting the XRMS at high angles (between the first, second and third inversion angles). Based on the modeling results, from the angular dependence of the integrated XRMS signal measured on sample 182 we can make an immediate first interpretation of the temperature evolution of the "dead layer" in this sample. Fig. 1 shows the XRMS signal near the inversion angle for 182 at four different temperatures. The inversion angle in temperatures moves from θ≈7º at 300K to larger angles as the temperature is decreased (θ≈9º at 200K and θ≈11º at 100K) and then, surprisingly, back to lower angles as the temperature is further reduced (θ≈8º at 20K). This implies that the dead layer thickness is large at 300K (~4uc =16Å), and first shrinks as the temperature is decreased (~2uc at 100K) but below 100K the dead layer starts to grow again (~3uc at 20K). Such behavior is quite unexpected, but it is unmistakable in the angular dependence of the XRMS – the only aspect of the XRMS geometry that can influence substantially the first inversion angle is the thickness of the dead layer, and it does so inversely (larger inversion angle implies smaller dead layer thickness).

We note another trend in the data on sample 182 that is clarified in Fig. 2. Here we show the XRMS at the four different temperatures and fixed incidence θ=5º, in both remanence (Fig. 2(a)) and saturation (H_{appl}~350 Oe, Fig. 2(b)). First, we see that in remanence the amplitude of the dichroism grows as the temperature decreases from 300K to 100K, but then decreases upon further cooling. This decrease could be due to a rotation of the magnetization towards the (110) axis due to magnetocrystalline anisotropy that appears in thicker films at low temperature (Ref. [4]). Note that the scattering plane was always along the (100) direction during our measurements. We will check this as a possible explanation of the decrease in amplitude of the dichroism in a conventional magnetometer in Trieste. But we note that this

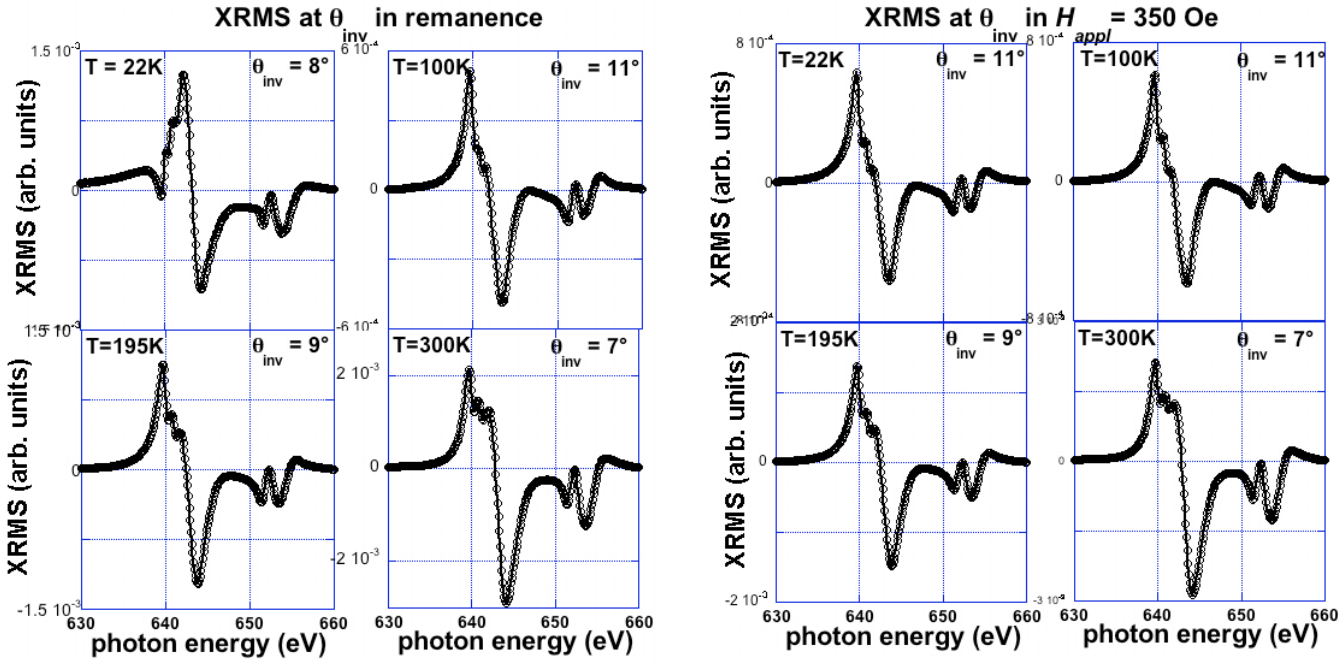

Fig. 1. XRMS curves ($R^+ - R^-$) at the inversion angle θ_{inv} at different T (a) in remanence; (b) in applied field. θ_{inv} is the angle at which the L3 integral equals 0, i.e., it is half positive and half negative. Note in (b) the shift of θ_{inv} to higher angles (indicating smaller dead layer) as T decreases, and below 100K the dead layer does not change thickness. By contrast, in (a) at 22K, θ_{inv} shifts to lower angle, 7°, as compared to 11° at 100K, indicating a regrowth of the dead layer thickness at low T.

effect could only cause a scaling of the intensity of the XRMS as a function of angle going from 100K to 20K; it cannot explain a shift in the inversion angle upon decreasing the temperature. These two trends (amplitude of XRMS versus temperature for fixed angle, integral of XRMS versus incidence angle for fixed temperature) are clearly separable in our data.

We note that currently there are no alternative spectroscopic techniques that can give the information we report here. Analysis of the XRMS spectra of 10uc films is significantly more complicated than that of thick films because of the presence of two closely spaced dead layers, but it is possible with the magnetooptic multilayer code.

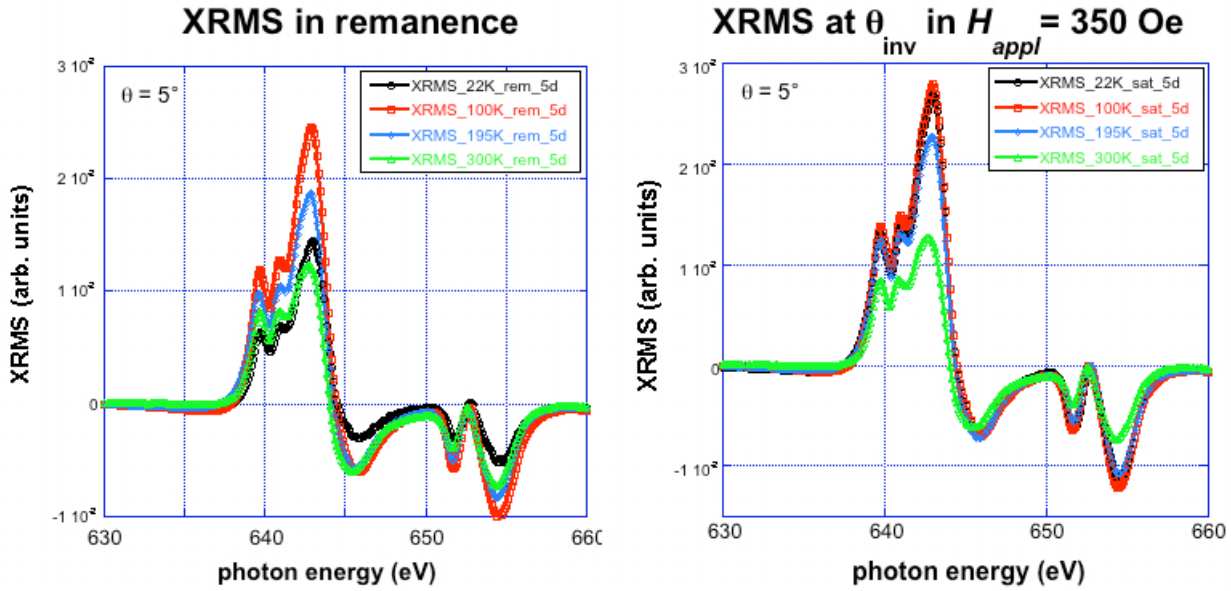

Fig. 2. XRMS curves ($R^+ - R^-$) at fixed incidence angle θ=5° at different T (a) in remanence; (b) in applied field. Note in (b) (in applied field) that the amplitude of the L3 peak increases monotonically for decreasing T, while in (a) (in remanence) the amplitude of the L3 peak increases going 300→100K, and then decreases going 100→22K.

References

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[3] B. A. Davidson et al., "*Atomic design of complex oxide heterostructures by molecular beam epitaxy*", submitted to Adv. Mater. (2010).

[4] M. Mathews et al., Appl. Phys. Lett. **87**, 242507 (2005).