ESRF	**Experiment title:** Interface properties of correlated oxide multilayers studied by standing-wave resonant inelastic x-ray scattering	**Experiment number**: HC 2391
Beamline: ID32	**Date of experiment**: from: 06/22/2016 to: 06/28/2016	**Date of report**: 09/01/2016
Shifts: 18	**Local contact(s)**: Dr. Nicholas BROOKES (email: brookes@esrf.fr)	*Received at ESRF:*

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Report:

In this experiment, we have for the first time demonstrated the feasibility of standing-wave RIXS (SW-RIXS) as a probe of interface-specific excitations. The standing wave (SW) was created by Bragg reflecting the incident x-ray from a multilayer heterostructure, tuning the photon energy to a maximum of reflectivity and thus SW amplitude, and then scanning the incidence angle through the Bragg angle to move the SW through the relevant interface(s). We carried out successful measurements on two types of multilayer samples: true multilayer samples with the form $[(La_{1.85}Sr_{0.15}CuO_4)_{2\ \text{unit cells}}/(La_{0.66}Sr_{0.33}MnO_3)_{7\ \text{unit cells}}]_{20}$ (LSCO/LSMO) and $[(CaCuO_2)_c/(SrTiO_3)_t]_m$ (CCO/STO), with c, t and m variable, and heterostructures grown on top of suitable multilayer SW generators, as bilayer $(SrTiO_3)_5/(La_{1.85}Sr_{0.15}CuO_4)_2$ grown on $[(SrTiO_3)_8/(La_{0.66}Sr_{0.33}MnO_3)_8]_{20}$. For the LSCO/LSMO multilayers, samples with two atomic stacking sequences were studied, as grown on TiO_2- and SrO-terminated $SrTiO_3$ substrates. We will focus on these two samples in this brief report.

The strength of the standing wave modulation is proportional to the square root of reflectivity of the multilayer, which varies as a function of incidence angle and photon energy. By means of the unique sample manipulator and spectrometer motions of the ERIXS facility, we were able to survey the reflectivity of multilayers for maximizing the standing wave effect in an LSCO/LSMO sample and scanning energy through the Cu L_3 absorption edge. The comparsion of a maximum reflectivity cut and the Cu L_3 x-ray absorption spectrum (XAS) lead us to use a photon energy 0.2 eV below the XAS for the SW-RIXS measurements on the two LSCO/LSMO samples.

Figure 1(a) shows a full RIXS spectrum typical of both LSCO/LSMO samples, which includes a quasi-elastic peak, magnetic excitations (magnon and bimagnon), dd excitations (xy, xz/yz, and z^2-types), and charge-transfer excitations [1]. Figs. 1(b) and (c) show zoomed portions of this spectrum, with peak fitting to derive the intensities of different components. In order to translate the standing wave in the direction perpendicular to the sample surface, the incidence angle was varied ±1.0° around the Bragg angle, producing “rocking curves” (RCs) of the intensities of different excitations. Figure 1(d) shows RCs of quasi-elastic and magnetic excitations, and Fig. (e) of dd excitations, for the TiO_2-terminated sample, with Figures

(f) and (g) showing analogous results for the SrO-terminated sample. The shapes of the RCs of bimagnon and charge excitations for both SrO- and TiO_2-terminated LSCO/LSMO samples are significantly different from those of the quasi-elastic excitations and the magnons, indicating that they have different depth distributions. By contrast, the RCs in Fig 1(g) for the three dd excitations of the SrO-terminated sample, are very close to each other, suggesting a nearly identical origin in depth in this sample, while the RC for z^2 excitations is much different from those for xy and xz/yz excitations for the TiO_2-terminated sample, as shown in Figure 1(e).

To gain a first-pass qualitative understanding of the depth sensitivity in these SW-RIXS results, we compared the RCs with simulations of the depth-dependent SW field, generated using an x-ray optical simulation program developed by Yang *et. al.* [2]. In order to estimate what the depth-resolved RCs will look like, the top LSCO layer was divided into three layers (surface, bulk, and interface), as shown in Fig.1(h), and the corresponding x-ray intensity profiles from these layers were generated (Fig.1(i)). By comparing these to the RCs in Figs. 1(d) and 1(f), we can tentatively conclude that the bimagnon and charge excitations are more interface-like. In addition, our data suggest that, for the dd excitations of the TiO_2-terminated sample, the z^2 orbital excitations have their origin more from the interface, while the xy and xz/yz orbital excitations are more from the bulk of the LSCO layer.

In summary, we have successfully demonstrated the first SW-RIXS measurements, in particular for correlated oxide multilayers. The differences in the rocking curves of different excitations, such as the magnetic and dd excitations of the LSCO/LSMO multilayers, indicate different average depths of origin. A much more quantitative analysis of these data is in progress, using the aforementioned code [2]. A manuscript describing this first proof-of-principle demonstration of SW-RIXS is in preparation [3].

References:

[1] M. Moretti Sala, *et. al.* N. J. Phys. **13**, 043026 (2011).

[2] S. H. Yang, *et. al.* J. Appl. Phys. **113**, 073513 (2013).

[3] C.-T. Kuo, *et. al.* in preparation.

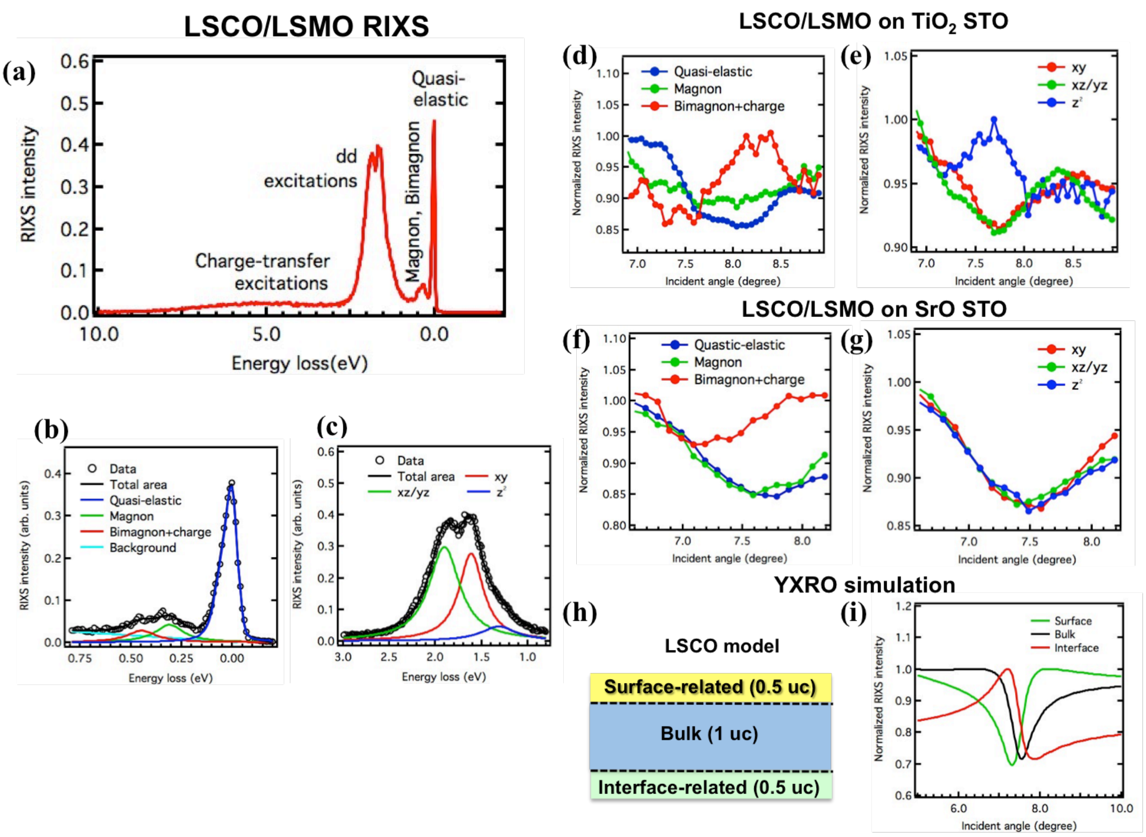

FIGURE 1 SW-RIXS measurements for two LSCO/LSMO multilayers. (a) RIXS spectrum of a TiO_2-terminated sample, together with zoomed and fitted regions in (b) and (c), and the corresponding rocking curves for (d) quasi-elastic and magnetic excitations and (e) dd excitations. Analogous rocking curves of an SrO-terminated sample for (f) quasi-elastic and magnetic excitations and (g) dd excitations. (h) Schematic drawing of the division of the top LSCO layer into surface (0.5 unit cell), bulk (1 unit cell), and interface (0.5 unit cell) using x-ray optical calculations and (i) the corresponding SW intensity profiles.