ESRF	**Experiment title:** Interface properties of correlated oxide multilayers studied by standing-wave resonant inelastic x-ray scattering	**Experiment number**: HC 3006
Beamline: ID32	**Date of experiment**: from: 06/28/2017 to: 07/04/2017	**Date of report**: 08/20/2019
Shifts: 18	**Local contact(s)**: Dr. Nicholas BROOKES (email: brookes@esrf.fr)	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Cheng-Tai KUO*, Charles FADLEY*, Shih Chieh LIN*, *Lawrence Berkeley National Laboratory, Materials Science Division, USA*

Giuseppe BALESTRINO, Daniele DI CASTRO*, *CNR-SPIN & University of Rome, Tor Vergata, Italy*
Gabriella maria DE LUCA*, *CNR-SPIN & University of Napoli, Physics Department, Italy*
Lucio BRAICOVICH, Giacomo GHIRINGHELLI*, Yingying PENG*, *Politecnico di Milano, Physics Department, Italy*
Marc HUIJBEN, *University of Twente, Low Temperature Division, Netherlands*

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 interfaces. We carried out successful measurements on two types of multilayer samples: true multilayer samples with the form $[(La_{1.85}Sr_{0.15}CuO_4)_2/(La_{0.66}Sr_{0.33}MnO_3)_7]_{20}$ (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 the SrO-terminated sample 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 of 931.2 eV for the SW-RIXS measurements on the LSCO/LSMO samples.

Figure 1(a) shows a full RIXS spectrum typical of both LSCO/LSMO samples, which includes a quasielastic peak, magnetic excitations (magnon and bimagnon), dd excitations (d_{xy}, d_{xz}/d_{yz}, and d_{z2}-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 the RCs of the *dd* excitations. All the experimental and theoretical RCs are normalized to a maximum of unity and are offset vertically for readability. The intensity of all *dd* excitations is modulated by 15-20%, meaning that the SW has a clear influence on the RIXS process: this is the experimental demonstration that SW-RIXS is feasible. We note also that these three *dd* RCs show a very similar shape, with intensity minima at ~8.2°, thus indicating a very similar depth distribution.

As the RC intensity modulation is significant, we now try to relate these experimental RCs to an approximate depth distribution of the loss processes involved, by simulating the RIXS process using the YXRO program developed by Yang *et. al.* [2]. Comparing these simulations to the experimental data, we find that the experimental *dd* RCs match the average of the RCs from the whole LSCO layer, as shown by the solid curves in Fig. 1(d). We can thus conclude that all three *dd* excitations show very similar behavior, with profiles suggesting that this part of the RIXS spectrum is quite independent from the position inside the LSCO layer, as indicated by the inset in Fig. 1(d). We now consider the RCs of the quasielastic and magnetic excitations, as shown in Fig. 1(e). The excitations in this range are more complex to analyze since the magnetic excitations lie very close to the phonon peaks and the elastic zero-loss line, and are also relatively weak. Again, we compare the experimental RC of magnetic excitations to the simulations for determining its depth distribution, and this yields the conclusion of a depth distribution peaked at the LSMO-top/LSCO-bottom and LSCO-bottom/LSMO-top interfaces, as shown in Fig. 1(f). At last, the experimental RC of quasielastic excitations was used to determine it depth profile. The quasi-elastic excitation is found to show contributions from the full LSCO layer, although weighted away from the top interface (see Fig. 1(f)).

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. Our article describing this first proof-of-principle demonstration of SW-RIXS has been published in the Physical Review B [3]. SW-RIXS measurements of correlated-oxide and other multilayer heterostructures should provide unique layer-resolved insights concerning their orbital and magnetic excitations, as well as a challenge for RIXS theory to specifically deal with interface effects.

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.* Phys. Rev. B **98**, 235146 (2018).

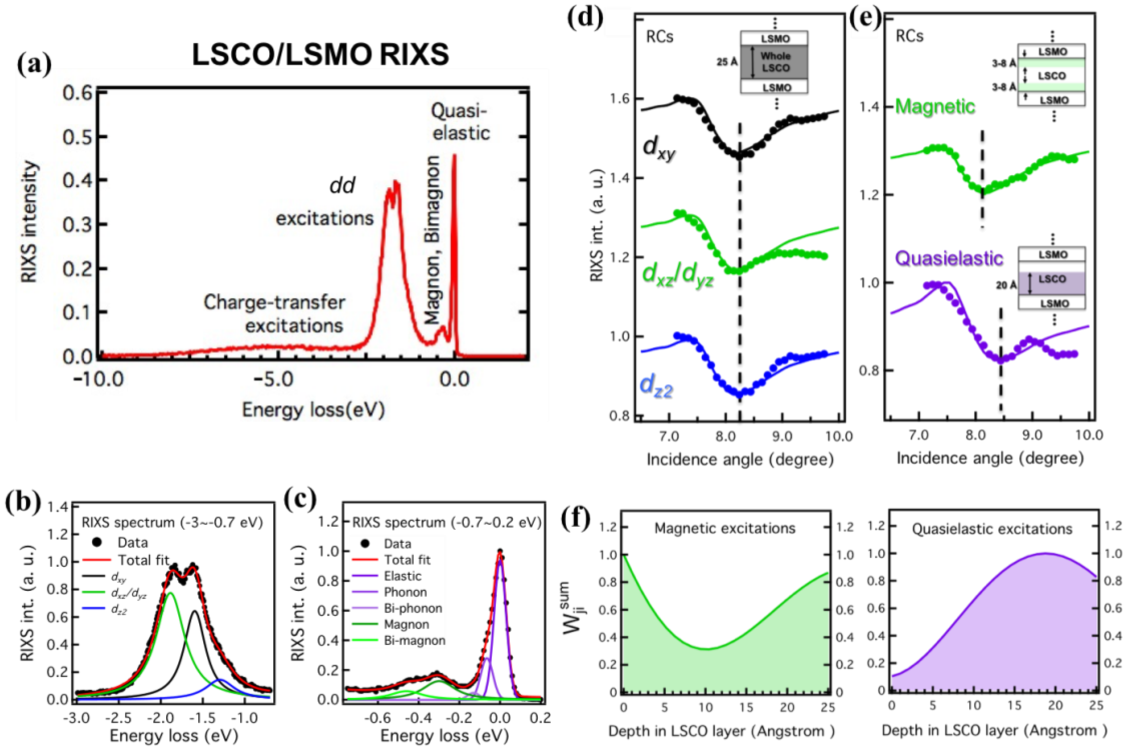

FIGURE 1 SW-RIXS measurements for a LSCO/LSMO multilayer. (a) Its RIXS spectrum, together with zoomed and fitted regions in (b) and (c), and the corresponding rocking curves for (d) *dd* excitations and (e) quasielastic and magnetic excitations. In (d) and (e) the experimental data are solid points and the simulated data are curves. (f) The determined depth profiles for the magnetic and quasielastic excitations. [3]