EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

# Experiment Report Form

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| ESRF | **Experiment title:** RIXS study of novel Cu-based density wave orders and low-energy excitations in cuprate/manganite multilayers | **Experiment number:** 84331 HC-4174 |
| **Beamline:** ID32 | **Date of experiment:** from: 11.02.2021 to: 22.02.2021 | **Date of report:** 28.02.2021 *Received at ESRF:* |
| **Shifts:** 16 | **Local contact(s):** Dr. Davide Betto | |


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We performed RIXS measurements on $YBa_2Cu_3O_7/Nd_x(Ca_{1-y}Sr_y)_{1-x}MnO_3$ (YBCO/NCSMO) multilayers for which different kinds of Cu-based density wave orders can be induced via the hole doping, x, and the Sr:Ca ratio, y, (or the tolerance factor, t) of the manganite layer. We measured high resolution RIXS at the Cu $L_3$-edge at grazing incidence geometry to maximize the signal of the charge order and minimize the influence of the magnons. Oppositely, by measuring at grazing exit geometry, we could also investigate the presence and nature of the magnons present in the multilayers. In these samples, the YBCO thickness being 7 nm, the measurement with polarimeter took aprox 20 hours per spectrum, with a resolution of 35 meV. We measured three heterostructures with (1) x=0.5, y=0.25 and (2) x=0.35, y=0.3 and (3) x=0.42, y=0.3. Some of the analysis observed in these samples are described in the following:

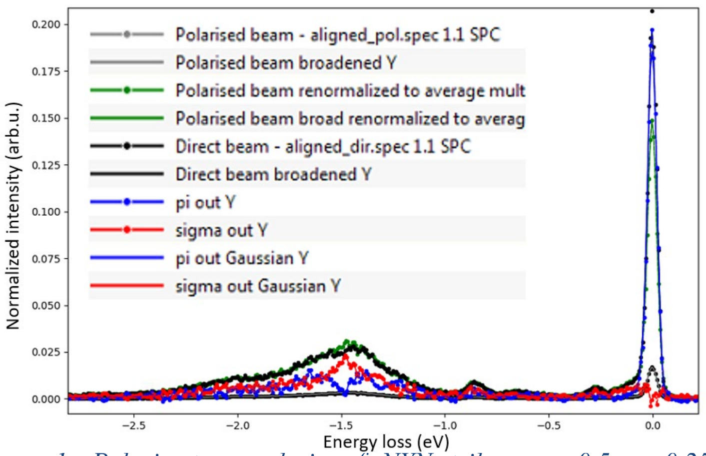


*Figure 1. Polarimetry analyzis of NYN trilayer x=0.5, y=0.25 indicating the absence of any elastic spin flip scattering for $Q_{||}$≈0.1 rlu.*

**(1) x=0.5, y=0.25:**

Very recently we found an entirely new kind of Cu-based density wave order with a periodicity of about 10 unit cells in this NCSMO/YBCO/NCSMO trilayer, for which NCSMO exhibits a long-range CE-type antiferromagnetic (AFM) and charge/orbital order **[1]**. We reconfirmed the presence of this charge order by performing a q-scan as described in **[1]**. Our intention was to understand the origin of this order by performing a polarimetric analysis at 20K at a few momentum positions along ($\pi$,0) and especially at $Q_{||}$ ≈0.1 rlu at LH polarisation and interface Cu-resonance energy (0.5 eV smaller than the bulk Cu-$L_3$ resonance observed in XAS (TEY)). The analysis of the polarimetry data shows that the $\pi$→σ chanel does not contribute to the elastic signal and thus confirms that the Bragg peak is due to a charge order without any significant magnetic contribution (Figure 1). This is a very important result.

We then focussed our attention to the magnon contributions in the inelastic part of the spectrum. We performed RIXS measurements in grazing exit geometry at the interface Cu-resonace energy of Cu (at $Q_{||}$ = 0.1, 0.25 and 0.4 r.l.u, along (π,0)) and bulk Cu-L3 edge (at $Q_{||}$ = 0.1, 0.2, 0.3 and 0.4 r.l.u along (π,0) and at $Q_x$= $Q_y$=0.21 r.l.u). The spectra at interface Cu-resonance energy indicated sharp inelastic features besides broad magnetic, phonon, elastic and crystal field contributions, which were absent when measured in bulk Cu-L3 resonance energy. This implies their origin as the interfacial states. We performed polarimetric analysis for one spectra which indicated that these features do not arise from spin-flip scattering. However, polarimetric analysis at $Q_x$= $Q_y$=0.21 r.l.u at bulk Cu-L3 edge clearly indicated two well resolved magnetic contributions (Fig. 2).

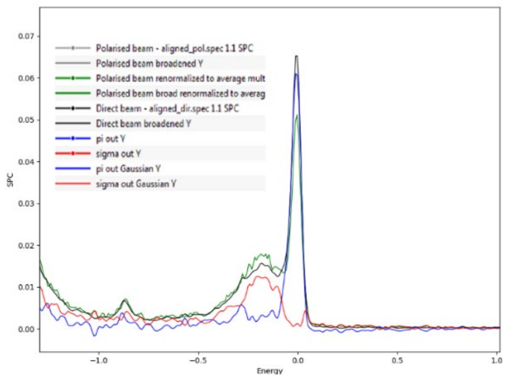


*Figure 2. Polarimetric analysis at $Q_x$= $Q_y$=0.21 r.l.u at bulk Cu-L3 edge clearly indicated two well resolved magnetic contributions (red line)*

The high flux, superior resolution of ID32 was instrumental to resolve these fine structures in the spectra even in a sample with 7 nm thick YBCO, which are otherwise convoluted to a much broader feature when measured in facilities like ADRESS. On the other hand, the ability to perform polarization selective scans helped us in identifying their possible origin.

**(2) x=0.35, y=0.3:**

For a multilayer with *x=0.35*, we performed a polarimetric analysis to learn whether the low-energy excitations between about 100 and 500 meV are of entirely magnetic origin. Our preliminary results from a previous beamtime at Diamond Light Source along the (π,0), (π,π) and (π,0)→(π,π) directions had revealed two such low-energy modes. This raised the question whether both of these modes are of magnetic origin which would be a strong indication for an anomalous magnon dispersion behavior in these multilayers. At ID 32, we performed a spin-polarimetric analysis at three $Q$ points along (π,π): $Q_x$= $Q_y$= 0.14, 0.21 and 0.28 r.l.u.. As shown in Fig. 1, our study revealed that the two inelastic modes between 100 and 500 meV are indeed of magnetic origin, since they occur mainly in the π-σ scattering channel. To our surprise, we observed an additional elastic signal in the π-σ scattering channel that is indicative of a static magnetic order near $Q_x$= $Q_y$= 0.21 r.l.u. (see panel c). Such a magnetic order would be new to YBCO and the other cuprates (the Bragg peaks of the spin-stripe-order in LSCO occur at ($Q_x$=0.5±δ, $Q_y$= 0.5∓δ) and are not accessible with RIXS at the Cu L-edge) and seems to arise from the coupling with the manganite layer and its CE-type antiferromagnetic and charge/orbital order.

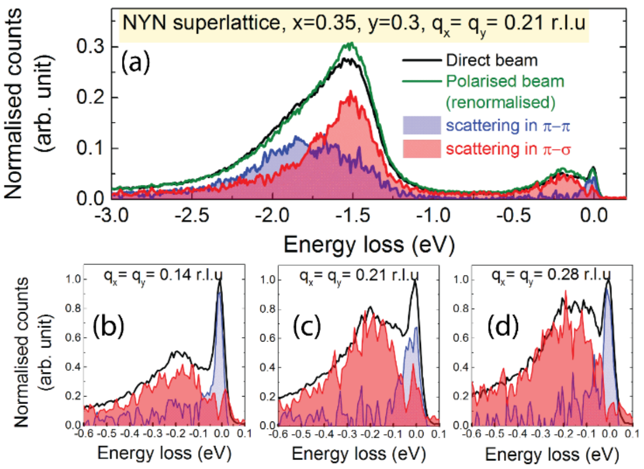


*Figure 3. RIXS-polarimetry of a NYN superlattice with x=0.3 and y=0.3: (a) Spectra at Qx=Qy=0.21 r.l.u indicating the signals without and with the polarization analysis of the scattered beam, as well as the deduced signals for the π→π and π→σ channels. (b) to (d): Dispersion along (π,π) of the low-energy π→π (blue) and π-σ (red) signals that are normalized to the maximum of the unpolarized elastic peak. Most notably is the signifciant spin-flip (π→σ) contribution to the elastic peak at $Q_x$ = $Q_y$ = 0.21 r.l.u.*

**(3) x=0.42, y=0.3:**

In the end, we performed a q-scan on this sample with a much bigger energy resolution (ΔE = 100 meV) to identify the Cu-charge order in this sample. Interstingly, we observed both the $Q_{||}$ =0.1 r.l.u (Cu-interface energy, LH polarization) order observed in samples with (x=0.5, y=0.3) **[1]** as well as $Q_{||}$ =0.3 r.l.u (buk Cu-resonance energy, LV polarization) order as observed in samples with (x=0.35) **[2]**.


**References:** **[1]** R.Gaina *et al.*, npj QM, **6**, 12 (2021) ; **[2]** E.Perret *et al.*, Comm.Phys **1**, 45 (2018)