ESRF	**Experiment title:** Intersite hopping mechanism of Mn-3*d* electrons in bilayer manganites: a Mn L_3 edge RIXS study	**Experiment number**: HE3543
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Report:

Recently, Weber *et al*. (Phys. Rev. B 82, 085105 (2010)) reported a high-resolution resonant inelastic hard x-ray scattering (Mn K edge RIXS) investigation of the bilayer manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with x = 0.36 and x = 0.50. They "observe a clear dispersion of up to 0.5 eV in the measured q range, which is direct evidence of the nonlocal character of *dd* excitation". We think that soft x-ray (Mn L_3 edge) RIXS would represent a more conclusive test on the dispersion of *dd* excitations in these compounds, since it directly probes Mn-3*d* electrons.

We mailny focused on samples of the type $La_{2-2x}Sr_{1+2x}Mn_2O_7$, with nominal doping x = 0.50.
Note that, given the Mn L_3 edge resonance (around 640 eV), the in-plane lattice parameter of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ compounds (≈ 3.87 Å) and the scattering angle set by the spectrometer (fixed at 2θ = 130°), the fraction of the Brillouin Zone that can be probed within this experiment is approximately 55%.

Figure 1 shows the dependence of the spectral features of the sample at half doping (x = 0.50) vs. the incoming photon energy both for linear horizontal (black) and vertical (red) polarization. Excitations energies are indicated in the absorption profile in the right panel.
Mn K edge RIXS measurement evidenced a "2 eV feature" (assigned to *dd* excitations), which is better seen here due to the relative higher energy resolution of the present experiment. Actually, we were able to resolve several features in the low-energy loss region of the spectra (1 to 3 eV), well separated from the quasi-elastic peak. They have a strong dependence on the incident photon energy: the relative intensities of the features at about 1.1 and 2.3 eV changes remarkably when moving across the absorption edge. In the pre-edge region (excitation energy A), both of them are dramatically suppressed. The width of these features is much larger than the experimental resolution indicating that multiple structures are likely to be present.
In the high-energy loss region, a feature at constant emitted photon energy is present (fluorescence). Possibly, a second one at intermediate energy loss can be recognized, moving in the Raman representation from about 2.4 eV at excitation energy B to 5.3 eV at excitation energy F.

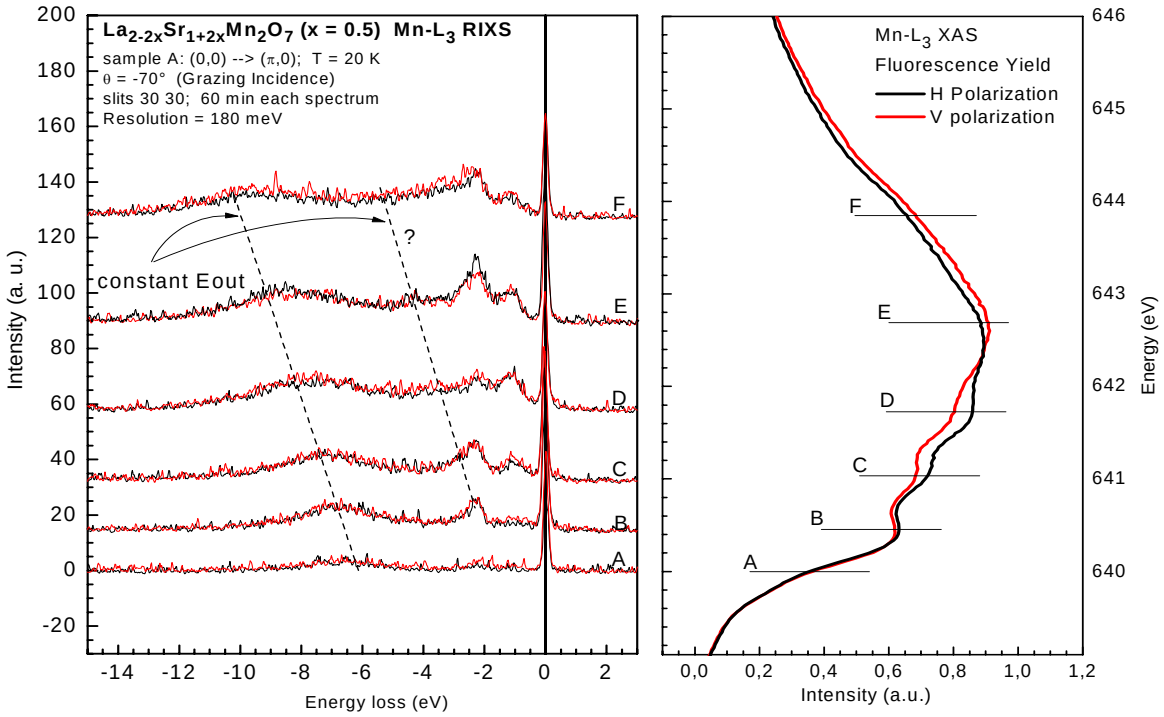

Figure 1: sample A (x = 0.50). Dependence of the spectral features vs. the incoming photon energy both for linear horizontal (black) and vertical (red) polarization. Excitations energies are indicated in the absorption profile on the right panel.

No evident dichroism is present when switching from one polarization to another. This fact is better documented in Figure 2, in which spectra with higher statistics at two selected angles and at one incident photon energy (excitation energy D of Figure 1) are shown. The two angles θ = 0° and -50° typically represent two interesting cases (θ is the angle of the incoming photon beam with respect to the sample normal), that is normal incidence and normal emission conditions, respectively. However, just a small increase in the counting rate is seen when moving from linear horizontal to vertical polarization, with no appreciable change in the shape of the overall spectrum.

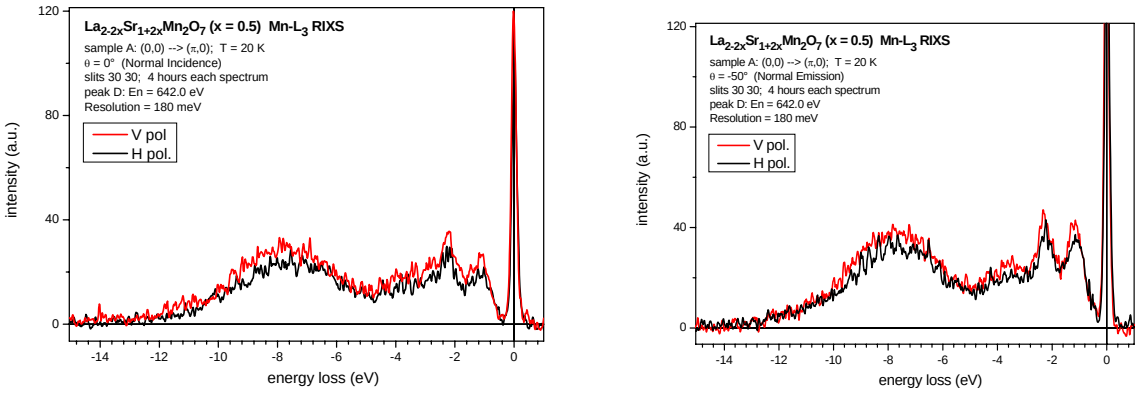

Figure 2: sample A (x = 0.50). Polarization (linear horizontal and vertical) dependence of the spectra at two selected angles (θ = 0° on the left and -50° on the right panel) at excitation energy D.

We thus decided to switch to circularly polarized light (being the flux provided by the undulators twice as much that in the case of linear polarization, for technical reasons) to study the momentum dependence of the Raman features at 1.1 and 2.3 eV. The study was carried out along two directions of the Brillouin Zone, that is the (0,0) → (π,0) and the (0,0) → (π,π) on sample A and B, respectively. The projection of the transferred

momentum in the *ab*-plane is changed by rotating the sample by an angle $\delta = \theta + 25°$ ($\mathbf{q}_\parallel \sim \sin \delta$). $\delta = 0°$ represent the specular-geometry case, for which $\mathbf{q}_\parallel \equiv 0$, i.e. the $\Gamma = (0,0)$ point is sampled. As one can see in Figure 3 a clear dispersion has not been seen, but

- the shape of the features changes, and
- their intensity evolves as well.

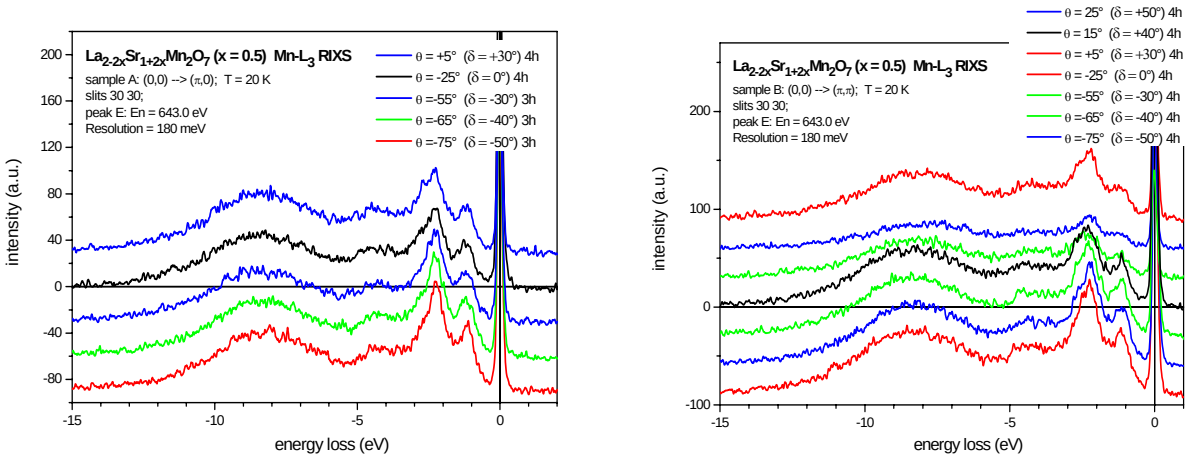

Figure 3: sample A and B (x = 0.50). Momentum dependence of the spectra along two direction in the Brillouin space ((0,0) → (π,0) on the left and (0,0) → (π,π) on the right panel) at excitation energy E for circularly polarized light.