ESRF	**Experiment title:** **Resonant inelastic x-ray scattering study of collective many-body excitations in $La_{1-x}Sr_{1+x}MnO_4$**	**Experiment number**: HS 3766
Beamline: ID26	**Date of experiment**: from: 29.10.2008 to: 4.11.2008	**Date of report**: 31.08.2009
Shifts: 18	**Local contact(s)**: Pieter Glatzel	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

*Dr. GECK Jochen / IFW Dresden, Helmholtzstr. 20, 01069 Dresden, Germany

*Dr. HERRERO MARTIN Javier / E.S.R.F., 6 rue Jules Horowitz, B.P 220, F-38043 Grenoble Cedex, France

*Dr. GLATZEL Pieter / E.S.R.F., 6 rue Jules Horowitz, B.P 220, F-38043 Grenoble Cedex, France

Dr. GARCIA RUIZ Joaquín / Instituto de Ciencia de Materiales de Aragón, C.S.I.C-Facultad de Ciencias, Universidad de Zaragoza, Plaza San Francisco n.9, E-50009 Zaragoza - SPAIN

MIRONE Alessandro / E.S.R.F., 6 rue Jules Horowitz, B.P 220, F-38043 Grenoble Cedex, France

Report:

The doped perovskite manganites are famous for their complex electronic behaviors [1]. Most notably this class of materials exhibits colossal magneto resistance, electronic superlattices or multiferroic properties. The richness of electronic phases results from an intimate coupling between lattice, charge, orbital and spin degrees of freedom, which interact strongly on various energy scales. In this experiment we studied the collective charge density excitations of $La_{1-x}Sr_{1+x}MnO_4$ as a function of hole-doping x. Well characterized single-crystals were studied with doping levels x=0, 0.125, 0.3 and 0.5.

The RIXS spectra were recorded at beamline ID26 of the ESRF. The incident X- rays were selected by means of a pair of cryogenically cooled Si (311) crystals. The total flux on the samples was 5×10^{12} photons/second. The emission spectrometer, equipped with five spherically bent (R=2 m) Si crystals in (440) reflection, was arranged with sample and detector (avalanche photodiode) in a horizontal Rowland geometry at 90 degrees scattering angle. The beam size was 0.3×1 mm^2 (hor.×vert.) and a slit with 1.5 mm horizontal opening was placed in front of the detector. The energy bandwidth of the spectrometer was set to 0.5 eV at 6490 eV. In this experiment we focused on the incident energy dependence of the RIXS intensity. Note that the momentum resolution in the present setup was significantly relaxed: the measured intensity was integrated over a finite but large volume in momentum space around the zone center.

No significant RIXS-intensity was found in the Mn K pre-edge region. Only around the main Mn K-edge RIXS-intensity was observed. As can be seen in Fig. 1, the characteristic y-shape of the intensity distribution within the RIXS-plane changes systematically as a function of x. The main change being that the splitting of the excitations along the incident energy (E_i) axis, observed for 6.550 keV $\leq$ Ei $\leq$ 6.557 keV, gets more and more reduced upon increasing x. In other words, the intensity distribution in this region of the RIXS-plane evolves from a bended to a straight line.

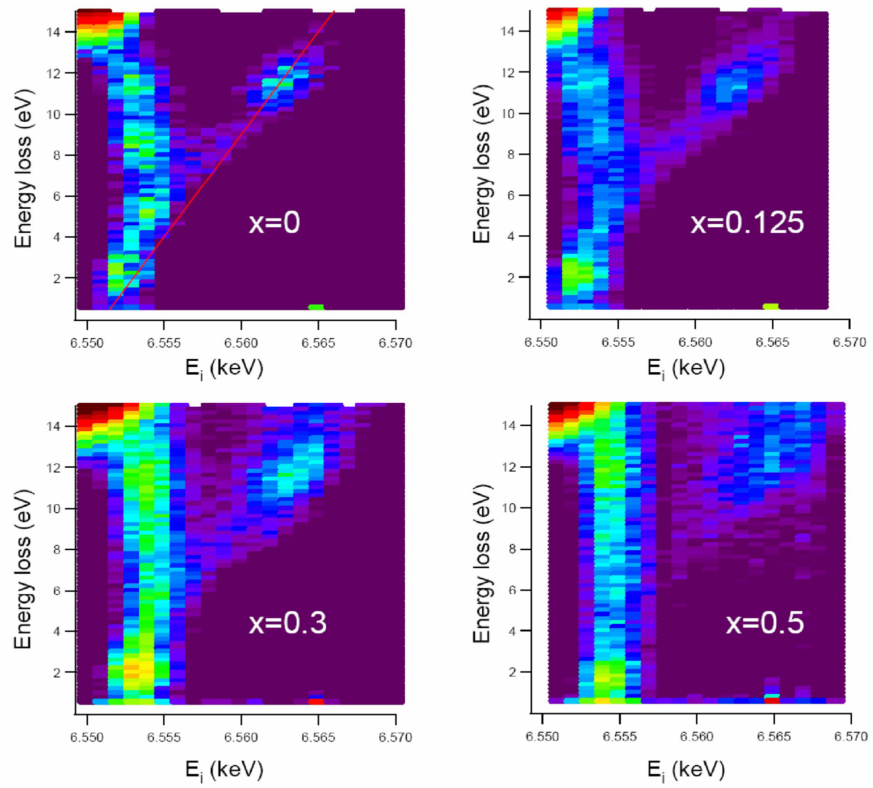

Figure 1: (color) Room temperature RIXS planes of $La_{1-x}Sr_{1+x}MnO_4$ single crystals with x=0, 0.125, 0.3 and 0.5. The data exhibits systematic changes as a function of x, i.e., y-shape of the intensity distributions changes. The red line shown for x=0 indicates the so-called out-going photon resonance [2].

For x=0, there are 5 broad features centered at 2eV, 3.8eV, 5.2eV, 8.6eV, 11.4eV energy loss, which resonate at slightly different incident energies. Upon doping, these excitations seem to become less pronounced, smeared out and the resonances get closer in E_i. These changes of the resonance energies seem to track the reduction of the local tetragonal distortion of the MnO_6-octahedra. This indicates that the different electronic excitations resonate with different intermediate states of different symmetries, which in turn, correspond to different excitation energies E_i

It has been proposed recently that, within the ultra-short core-hole lifetime (UCL) approximation, the RIXS intensity can be related to the dynamical structure factor $S(\mathbf{q},\omega)$ [2]. In Fig.2 we show a first comparison between UCL calculation and the experimental RIXS plane. The $S(\mathbf{q},\omega)$ used for the calculation was determined from electron energy-loss spectroscopy (EELS) measurements. It can be observed that the y-shape is reproduced qualitatively within the UCL. However, there are important quantitative discrepancies. For instance, the excitations observed around 11.4 eV in experiment, are not reproduced by the UCL. Instead a strong excitation around 13.5 eV is obtained in the calculation. One reason for this disagreement seems to lie in the different sensitivities of EELS and RIXS. However, this issue needs to be studied in more detail.

In addition XES-data, also obtained during this experiment, together with extended cluster calculations implies that non-local many-body effects lead to a anomalous stabilization of in-plane $3dx^2$-y^2 states [3].

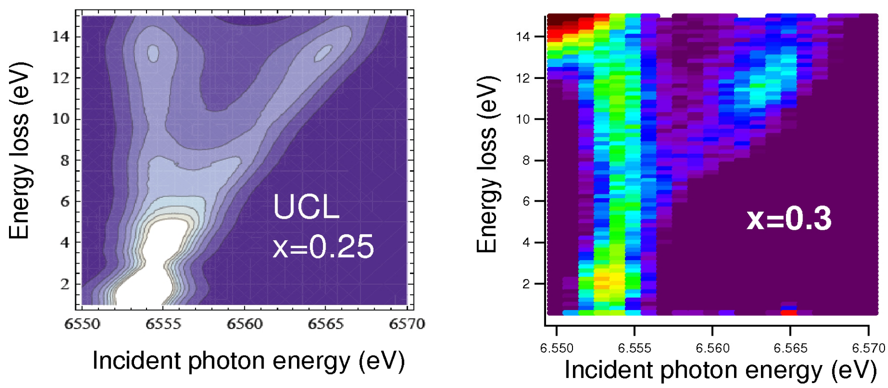

Figure 2: Comparison between the calculated UCL (left) and measured RIXS plane (right). The $S(\mathbf{q},\omega)$ used for the UCL calculation was determined by EELS.

References

[1] Focus on Orbital Physics, New J. Phys. 6 (2006)
[2] L.J. P. Ament et al., Phys. Rev. B 75, 115118 (2007)
[3] J. Herrero-Martin et al., arXiv:0906.5474 [cond-mat.str-el]