ESRF	**Experiment title:** Low-energy excitations in strongly correlated materials using high-resolution RIXS	**Experiment number:** HE-2569
Beamline: ID16	**Date of experiment:** from: 12.09.2007 to: 18.09.2007	**Date of report:** 1-Sep-2009
Shifts: 18	**Local contact(s):** S. Huotari	*Received at ESRF:*

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Report:

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The purpose of the experiment was to study the low-energy (0.5–5 eV) electronic excitations in a prototype Mott insulator nickel monoxide (NiO), using resonant inelastic x-ray scattering (RIXS) at the Ni K edge (8.33 keV). In the energy range of the experiment, typical excitations observable are excitations of the d-d (also called crystal field) type and charge-transfer type. NiO is a charge-transfer insulator with a gap of 4 eV, and the excitation spectra were expected to comprise a charge-transfer excitation starting from 4 eV, and within that gap of possible d-d and perhaps even magnon excitations. From optical, soft-xray RIXS and energy-loss spectroscopy studies [1] we knew that d-d excitations exist at energies of 1, 1.6, and 3 eV. Earlier hard x-ray RIXS studies [2] had seen hints of excitations at 1.6 and 3, but nothing clearly, especially nothing at all at 1 eV which was intriguing. We decided to set up an experiment at Ni K edge using our new technology based on diced analyser crystal and a position-sensitive detector.

The experiment was performed at the beamline ID16. The incident beam was monochromatised to a bandwidth of 70 meV at 8.3 keV. The beam was focused using a toroidal mirror. The spectrometer was based on a diced Si(551) scanning analyser crystal, working on a vertical Rowland circle with a diameter of 1 m, and a pixelated Medipix2 detector. The achieved total energy resolution was 0.2 eV.

Fig. 1 (left) shows the absorption cross section with different sample orientations with respect to the linear polarisation vector when changing it between [100] and [010]. A quadrupolar prepeak is visible at 8331 eV, maximised in intensity when the polarisation vector is oriented along [110]. The right panel shows examples of scattered-photon spectra measured at a few such sample orientations. The spectra consist of three lines (d-d excitations), at 1.05, 1.6 and 2.95 eV. The charge-transfer spectrum starts at 3.5 eV. The relative intensities of the d-d excitations is seen to be sensitive to the sample orientation, reflecting the symmetry of the related excitons. A measurement of the two-dimensional RIXS map as a function of incident photon energy E_1 and energy transfer E_2 around the quadrupolar absorption prepeak and the d-d excitations reveals the spectrum depicted in Fig. 1.

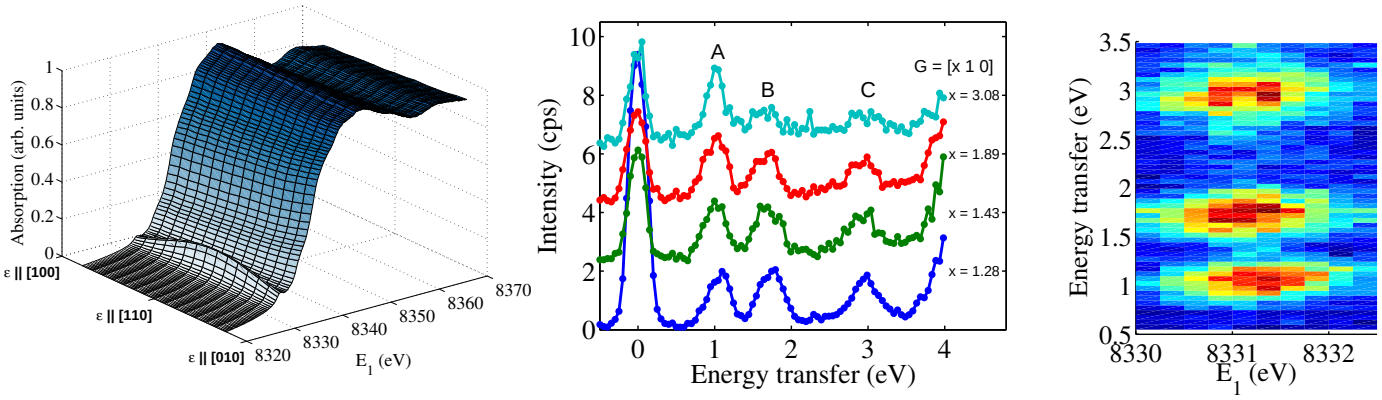

Figure 1. Left: Absorption cross section measured using a total fluorescence yield method. Middle: Examples of energy-loss spectra taken at the prepeak with different sample orientations. Right: Experimental 2D RIXS plane.

From Fig. 1 it can be seen that while the experimental result shows all three *dd* excitations being centered almost around the same resonance energy E_1, the 1.05-eV excitation has a slightly shifted E_1. The difference in resonances can only be due to nondegeneracy of the empty e_g orbitals. The lifting of the degeneracy in atomic picture could be due to lattice distortions or exchange interaction. It has also been evidenced in the band structure calculations by Kuneš et al. [3] and Zhang et al [4] which show k-dependent nondegeneracy. Tentatively the band structure picture seems to be correct.

[1] S. G. Chiuzbaian et al. PRL 95, 197402 (2005);
[2] E. Collart. PhD Thesis (University of Paris VI (Dir.Abhay Shukla).
[3] J. Kuneš et al. Phys. Rev. Lett. 99, 156404 (2007)
[4] W.-B. Zhang et al. Phys. Rev. B 74, 054421 (2006)