ESRF	**Experiment title:** Disentangling charge and spin excitations in Cu L3 RIXS spectra of YBCO	**Experiment number**: HE3753
Beamline: ID08	**Date of experiment**: from: 19/06/2012 to: 26/06/2012	**Date of report**: 27/08/2012
Shifts: 18	**Local contact(s)**: Nick Brookes	*Received at ESRF:*

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Report:

Depending on the scattering geometry (scattering angle and incident polarization), the relative weight of the charge and spin excitations in the RIXS response of doped cuprates varies strongly. Being able to separate charge and spin contribution to the RIXS responses is a crucial prerequisite to give an absolute value of the spin susceptibility using this technique.
This was the purpose of the proposed experiment, using the polarization analysis set-up of the scattered photons designed (Figure 1 and ref. [1]) and built by the Milano group, in collaboration with ESRF staff.

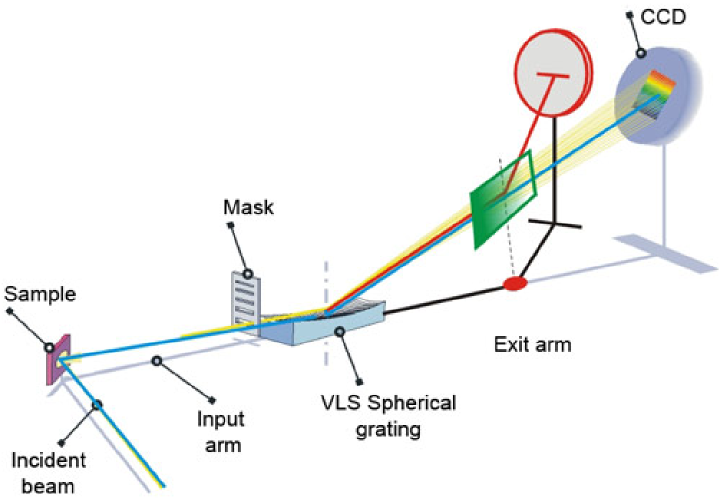

Figure 1: Optical layout of the AXES spectrometer with the multilayer mirror mounted between the grating analyzer and the CCD detector. The "traditional" optical path is in blue. When the multilayer is inserted, the beam gets deflected by 40 degrees (red line) and the detector is moved accordingly. The multilayer rotates around the axis normal to the output arm.

The polarimeter used here is an evolution of the initial version presented in ref. [1], with more advanced optics (variable spacing multilayer) and increased luminosity. It has been installed and commissioned on the AXES RIXS spectrometer at beamline ID08 two weeks prior to the present experiment. Due to the limited beam availability during this initial commissioning, the last phase of the commissioning, which consisted in a mapping of the diffraction from the multilayer as function of the incident energy, has been inserted in the program of the present experiment (first two days)

Figure 2 shows typical RIXS spectra on undoped and doped $NdBa_2Cu_3O_{6+x}$ with different incident polarizations [2] , without analysis of the scattered light polarization. The chosen momentum transfer is $q_{//}$ = 0.37 r.l.u. As seen in fig. 2, adapted from ref.[3] and based on a model developed in refs. [4,5] for undoped compounds, theoretically, at this momentum in π scattering geometry one mostly probes spin flip excitations, whereas in the σ scattering geometry charge excitations (which also includes even spin excitation such as bi-magnons) are dominant. Our central finding in the work reported in ref.[2] is that, at high $q_{//}$, in the π geometry, where magnetic excitations are largely dominating the RIXS response, the spectral weight of these excitations is essentially doping independent (Figure 4).
At lower $q_{//}$, this spectral weight is even found to increase with doping. This is likely due to the fact that in this case, instead of pure magnetic excitations, a mix of charge and spin excitations is seen, and that the relative weight of the charge excitations increases as doping increases. This can also be seen in Figure 2, where the relative weight of the continuum seen in the σ geometry (in red) increases strongly when going from the undoped (top) to the doped (bottom).

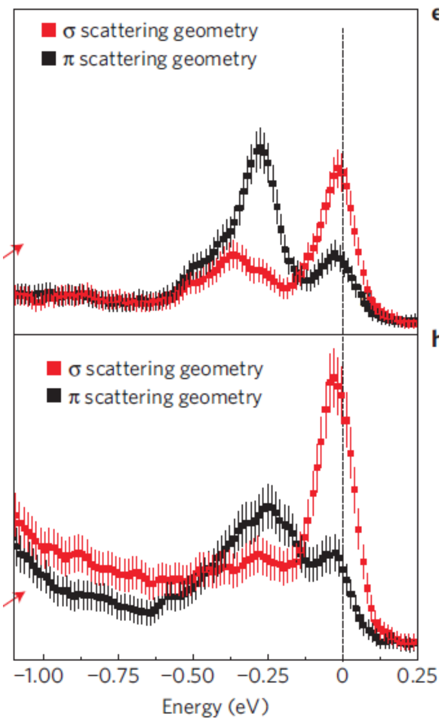

Figure 2: Typical Cu-L_3 RIXS spectra of undoped (upper panel) and doped superconducting cuprate (lower panel) measured with π (black) and σ (red) incident polarizations. Adapted from ref. [2]

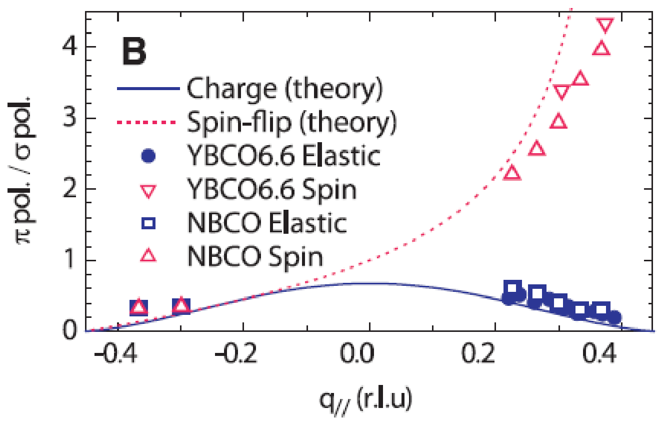

Figure 3: Calculated relative intensity of charge and spin excitations as function of momentum in π and σ channels (adapted from ref. [3])

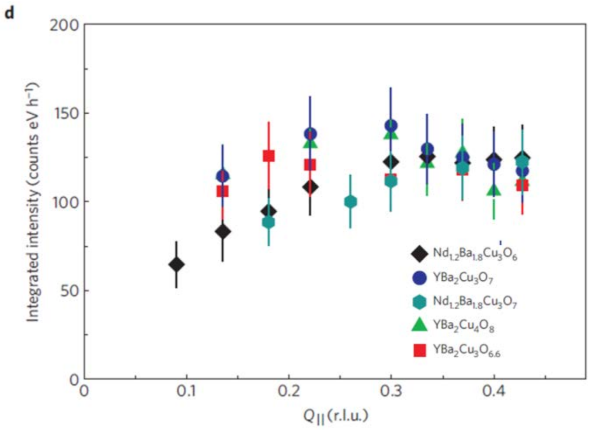

Figure 4: Integrated intensity of the magnetic excitations as function of momentum for samples with different doping levels.

In order to try to separate experimentally the charge from the spin contribution in the data, we have used polarization analysis of the scattered light. To maximize the effect of the polarimeter, we have chosen to limit ourselves to two samples: one with very low doping ($YBa_2Cu_3O_{6.35}$) in which we expect low charge signal, and one with much higher doping ($Y_{0.85}Ca_{0.15}Ba_2Cu_3O_7$) in which the charge signal is much higher.

We present in this report the first systematic analysis of the data taken during the beamtime (from 19/06 to 26/06) on the latter sample, i.e. the overdoped one. Analysis of the measurements on the other sample is still ongoing. As a matter of fact, we demonstrate that:

1) The polarimeter is working properly
2) The identification of charge and spin contributions can be done experimentally at all angles

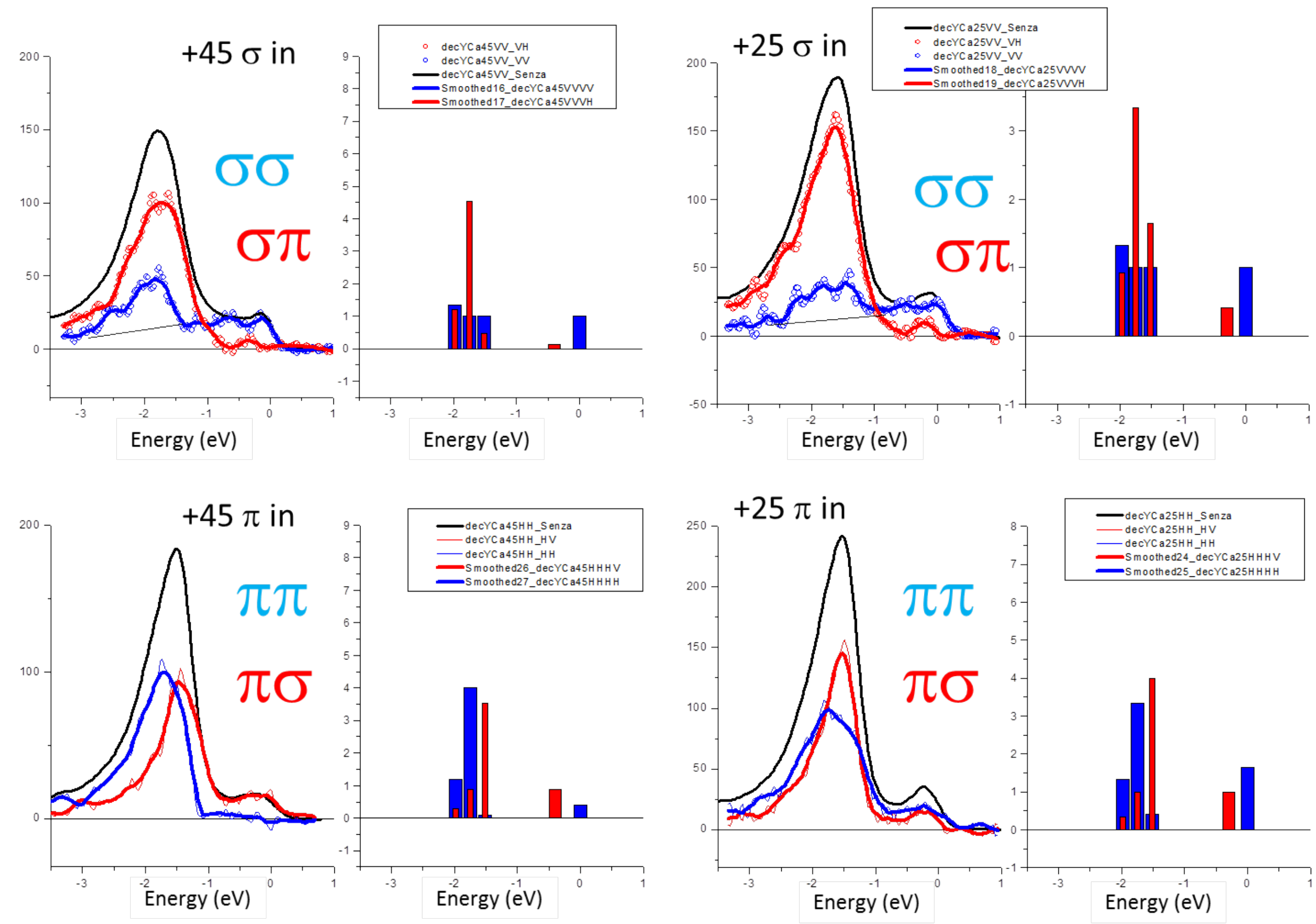

Figure 5: in each panel is plotted the data taken without polarimeter (black line) and the decomposition if the scattered light from our analysis. In the right part of each panel we shows calculated intensity of elastic line, single magnon signal and various dd excitations calculated after the theoretical model developed in ref.[4]and ref.[5]

Inserting the multilayer on the scattered photons path allows changing the relative weight of π and σ components of the scattered light, by an amount determined independently during the experiment.
Confrontation of the measurements performed with and without the polarimeter therefore allows reconstructing without assumption the scattered intensity in all for channels ($\sigma\sigma$, $\sigma\pi$, $\pi\pi$, $\pi\sigma$) at all angles.
We did it for two angles, +45° and +25°, corresponding respectively to large $q_{//}$ (mostly charge excitations expected in s, mostly spin in p) and smaller $q_{//}$ (mixing of charge and spin in both channels).

Each panel of the figure took 6 hours plus 1 hour without the polarimeter and 2 hours in various tests.

The main outcomes are the following:

1) The component separation at 45° (large $q_{//}$) confirms empirically the theoretical prediction (never tested directly) that in the mid-infrared, the whole intensity with π incidence comes from magnetic excitations (and seen as expected in the crossed polarization ($\pi\sigma$) channel). This validates *a posteriori* the conclusion of ref. [2] that the spectral weight of the short range magnetic fluctuations is doping independent.
2) At 45° (large q) large angle with σ incidence, the signal is dominated by the non crossed channel (charge excitations), with only a tiny spin flip component, which increases slightly when going towards lower $q_{//}$.
3) At low q, with p incident polarization, only about one half of the intensity seen without polarimeter actually comes from pure spin flip excitations.

The main conclusion is therefore that we can separated odd spin excitations, even spin excitations and charge contributions from our data.
There is a qualitative general agreement of experiment with the theory developed for undoped systems, although, not surprisingly, a more complete treatment is needed to discuss the details.
As a byproduct of the analysis, the subcomponents of the dd excitations can be obtained experimentally, which is impossible without polarimeter in the overdoped systems, even with higher resolution.
The limit to the sensitivity comes from the oscillations of the background of the detector which will be changed in the medium term. The dd decomposition is extremely sensitive to the normalization and a complete scrutiny of the results to get the best normalization is still going on; on the other hand the analysis in the low energy scale is almost final.

References:

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3. Ghiringhelli, G., et al., Science, **337**, 821-825 (2012).
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5. Moretti Sala, M., et al., New Journal of Physics, **13**, 043026 (2011).