ESRF	Experiment title: Orbital Waves in 2D systems	Experiment number: HC-3296
Beamline:	Date of experiment:	Date of report:
ID32	from: 27/01/2018 to: 04/02/2018	16/03/2018
Shifts:	Local contact(s):	Received at ESRF:
18	Davide Betto	

Names and affiliations of applicants (* indicates experimentalists):

R. Arpaia^{1,*}, L. Braicovich^{2,*}, D. Di Castro^{3,*}, R. Fumagalli^{2,*}, G. Ghiringhelli^{2,*}, M. Rossi^{2,*}

¹ Department of Microtechnology and Nanoscience, Chalmers University of Technology, SE-41296 Göteborg, Sweden

² Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo Da Vinci 32, I-20133 Milano, Italy

² CNR-SPIN and Dipartimento di Ingegneria Civile e Ingegneria Informatica, Università di Roma Tor Vergata, via del Politecnico 1, I-00133 Roma, Italy

Report:

Recently, experimental works have shown the existence of orbitons in quasi-1D cuprate [1,2,3] (e.g. Sr_2CuO_3) and in quasi-2D iridates [4]. Theories predict this effect in low dimensional systems: intraband excitations (called *dd* excitations) can split their spin and orbital components, giving rise to complex and intrigung dispersions in reciprocal space. In the case of correlated transition metal oxides, such as quasi-1D cuprates, the separation between the orbital and the spin degrees of freedom gives rise to different dispersion relations for orbiton and spinon, respectively. In the case of quasi-2D iridates it is not possible to excite a real orbiton but, instead, $a_{jeff}=3/2$ quasi-particle which carries both spin and orbital quantum numbers is present. Regarding specifically the cuprates, as already mentioned, *dd* excitations with sizeable dispersion were predicted and experimentally observed only in quasi-1D materials. In 2D cuprates, *dd* excitations (or magnons) display large energy dispersion thanks their collective nature. Despite this, our preliminary data (collected during LTP HC886) on CaCuO₂ (CCO) suggest that *dd* excitations may disperse also in 2D systems. It was suggested [5,6] that the orbiton excitation in a quasi-2D cuprate might still be dispersive, even though its dispersion relation should be different to that in a quasi-1D cuprate and it is due to the polaronic coupling between orbitons and magnons which is absent in 1D.

Since the theory model used in the 1D case cannot address what we have seen in CCO, we decided to perform a systematic measurement on both 1D Ca₂CuO₃ and 2D CaCuO₂ systems. In order to be able to obtain a detailed comparison of the 1D and 2D systems, we measured at fixed 2 θ angle (= 150°) RIXS spectra as a function of the transferred momentum tuning the incident x-rays energy at the Cu *L*₃-edge (931 eV) with both horizontal and vertical polarization of the incoming light. In particular, for the 2D CCO we collected spectra along the 10 direction, while for the 1D Ca₂CuO₃ we measured along the directions both parallel and perpendicular to the chains (01 and 10 directions, respectively). The latter didn't show any dispersing feature, so will not discuss that measurement.



Figure 1: RIXS intensity maps of CaCuO₂ (left) and Ca₂CuO₃ (right) measured with vertical incident polarization as a function of energy loss and transferred momentum.

In Fig. 1 we show RIXS intensity maps for the 2 systems. Spectra have been acquired with a combined energy resolution (beamline and spectrometer) of ~55 meV. First of all, we notice that in the low energy scale magnetic excitations show a different energy vs momentum dependence: Ca₂CuO₃ (right panel) has a twospinon continuum, characterized by 2π periodicity in reciprocal space, while CaCuO₂ (left panel) has a single dispersive peak (magnon) with π periodicity. In these maps, acquired with vertical polarization, we can also clearly distinguish at higher in-plane momentum transfer the bi-magnon peak (which disperses like the single magnon peak) and dispersionless phonons (close to the quasi-elastic line at 0 energy loss). Regarding *dd* excitations, they seem to be dispersing in both systems but with different symmetries. At first glance we clearly recognize that the energy positions of the *dd* excitations are quite different in the two systems, as are their angular dependences (especially for the z^2 peak at higher energy loss). Moreover, in the CCO we see an extra feature around -2.3 eV which seems not to be present in the 1D systems. To better understand the nature of this new dispersive feature we performed the polarization analysis of the scattered light using the unique capabilities of the ERIXS spectrometer and its polarimeter.

In order to characterize the polarization dependence of this feature, we performed systematic measurements for both 1D and 2D systems at three different in plane transferred momenta with both horizontal and vertical polarizations of the incoming light.



Figure 2: Polarization-resolved RIXS spectra of CaCuO₂ (left) and Ca₂CuO₃ (right) at (0.404; 0) and (0; 0.436) in plane transferred momentum, respectively. The spectra have been measured with parallel polarization of the incident light with respect to the scattering plane. In the inset at the top right of both figures we show a zoom-in of the low energy scale.

As an example, in Fig. 2 we show polarized RIXS spectra of both the systems under investigations. Spectra sketched in Fig. 2 were measured with horizontal incident polarization and with a combined energy resolution of ~65 meV for both the direct beam and the beam measured through the polarimeter. In the low energy scale we confirm the "crossed" nature of the magnetic excitations ($\pi\sigma$ channel), which results from a 90° rotation of the polarization in spin-flip excitations due to the conservation of angular momentum. *dd* excitations show a peculiar angular dependence which is quite different in the two systems. Moreover, the extra-feature seems to be present only in CaCuO₂.

After a detailed analysis of experimental data, which will include the study of the orbiton dispersions, comparison between different systems and polarization dependence of the different spectral features, we will write a manuscript including a theoretical model describing dd excitations and their dependence on the dimensionality of the system.

References

[1] J. Schlappa *et al.*, "Spin-orbital separation in the quasi-one-dimensional Mott insulator Sr₂CuO₃", Nature **485**, 82 (2012);

[2] V. Bisogni *et al.*, "Orbital control of effective dimensionality: from spin-orbital fractionalization to confinement in the anisotropic ladder system $CaCu_2O_3$ ", Phys. Rev. Lett. **114**, 096402 (2015);

[3] Kim B.J. et al., "Distinct spinon and holon dispersions in photoemission spectral functions from onedimensional $SrCuO_2$ ", Nature Phys. 2, 397–401 (2006);

[4] Kim, J. et al., "Excitonic quasiparticles in a spin–orbit Mott insulator", Nature Commun. 5, 4453 (2014);

[5] K. Wohlfeld *et al.*, "Intrinsic Coupling of Orbital Excitations to Spin Fluctuations in Mott Insulators", PRL **107**, 147201 (2011);