ESRF	**Experiment title:** Study of orbitons in the quantum antiferromagnets $KCuCl_3$ and $TlCuCl_3$	**Experiment number**: HC 2691
Beamline: ID28	**Date of experiment**: from: 01/02/2017 to: 07/02/2015	**Date of report**: 28/02/2017
Shifts: 21	**Local contact(s)**: Flora Yakhou	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Santiago Blanco Canosa

Justina Schlappa

Josu López-López

Natalia Gerasimova

Report:

In one dimensional systems, the electron breaks up into its three elementary quasiparticles, carrying the spin, charge and orbital degree of freedom. The finding of orbitons [1] in a strongly anisotropic 1D compound was surprising and revealed the importance of coupling between orbital and spin degrees of freedom. This coupling may lead to the confinement of orbital excitations in antiferromagnetic systems with higher dimensions [2].

In this beamtime, we have investigated if, despite the higher dimensionality of the magnetic correlations, the directed character of the orbital motion, combined with the small magnetic exchange energies favors a detectable dispersion of orbital excitations in these compounds.

We have measured the orbital excitations in the quantum antiferromagnet $TlCuCl_3$ at 20 K at the ERIXS spectrometer at ID32. We collected Resonant Inelastic X-ray scans for E_i=931 eV (Cu L_3-edge) and interval of loss energies between $-5 < \hbar\omega < 20$ eV at a number of reciprocal space positions, mainly focusing on the (100), (001) and (10-2) directions at 20 K. The total energy resolution of the setup was 60 meV. The achivable momentum transfer along the *a*-axis reached ±0.48rlu.

The cleaving plane reveals the *b*-axis perpendicular to the surface, thus, the (100) (010), (001) (010) and the (10-2) (010) scattering planes can be accessed by an azimuthal rotation around the *b*-direction. Figure 1

shows the XAS spectra as a function of the azimuthal rotation. We can see a strong dichroism of the fluorescence signal between 0 and 2π, with a maximum at π /2 that corresponds with a polarization of the light parallel to the (10-2) direction. This suggests that, in agreement with our expectations, the holes are mainly localized in the *3d* orbitals of local x^2-y^2 symmetry.

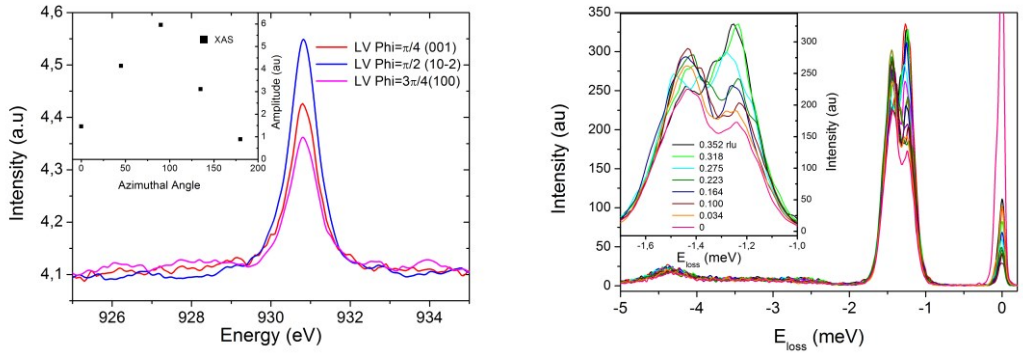

Fig. 1. ***(Left)*** X-Ray absorption spectra for the azimuthal dependences studied in TFY at θ=90º and 2θ=130º. LV accounts for vertical polarized light and each value of phi corresponds to the polarization of light along the direction specified in parenthesis. ***(Right)*** Momentum dependence of the RIXS spectra at T= 20 K along the (10-2) direction.

In figure 1 right, we show the RIXS spectra for Energy loss up to -5 eV. Centered at 0 Energy loss, the elastic line is observed. The broad band observed between -4 and -5 eV arises from charge transfer excitations due to the highly insulating character of $TlCuCl_3$. Between -1 and -2 eV, orbital excitations are observed. They are composed of two main bands with several shoulders. The two main bands show a clear dispersion, which demonstrates that the small exchange coupling energies and the directed character of the orbital motion along the chains will favor the dispersion of orbital excitations in these compounds, as we pointed out in the proposal. Similar set of scans were taken transferring momentum along the *a*- and *c*-directions. Prof. Jeroen van der Brink and Dr. K. Wohlfeld are currently performing quantum mechanical calculations based on the *t-J* model in order to model the spectra.

Unfortunately, there was not enough time to carry over the study of the orbital excitations in $KCuCl_3$. Although isostructural, the 3D spin correlations are weaker in this system than in $TlCuCl_3$ [3]. It is of primary importance to complete the study in $KCuCl_3$ in order to quantify the influence of magnetic coupling strength on the orbiton bandwidth in this model system. For this, we are kindly resubmitting the proposal and request another 18 shifts of beamtime.

References.

[1] J. Schlappa et al., Nature 485, 82 (2012).
[2] V. Bisogni et al., Phys. Rev. Lett. 114, 096402 (2015).
[3] N. Cavadini et al., Phys. Rev. B 63, 172414 (2001).