ESRF	**Experiment title:** Study of the three-dimensional magnetic field induced charge-density wave order in the cuprate superconductors	**Experiment number**: HC 2887
Beamline: ID 24	**Date of experiment**: from: 28 September 2016 to: 05 October 2016	**Date of report**:
Shifts: 18	**Local contact(s)**: Olivier Mathon, Sakura Pascarelli	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Wojciech Tabis*, I. Biało*, Faculty of Physics and Applied Computer Science, AGH UST, Kraków, Poland

Fabienne Duc*, Naveen Kumar* Paul Frings , LNCMI, Toulouse, France

Emilio Lorenzo*, CNRS - UJF - Institut Neel, Grenoble, France

Matthieu Le Tacon, MPI für Festkörperforschung Heisenbergstrasse, Stuttgard, Germany

The ordered states within the pseudogap phase of the cuprate superconductors have attracted attention as the fluctuations of the order parameter can be responsibe for the superconductivity in these materials [1]. Although the CDW order has been found to be present in all the families of cuprates and its universality in the underdoped region of the phase diagram is well documented, its role in the pseudogap formation and the superconducting pairing is not unambiguously established. While the two-dimensional (2D) charge density wave order has been established to be universal property of the cuprates [2], an additional correlations along the *c*-axis, perpendicular to the CuO_2 planes, have been observed in $YBa_2Cu_3O_{6+\delta}$ (YBCO) at fields exceeding 18 T [3]. It has been suggested that the additional three-dimentional (3D) order does not simply evolve from the short-range 2D correlations, but rather that the latter coexist with the former [4]. Whether the 2D or the 3D CDW order is responsible for the changes of the Fermi surface topology in the underdoped cuprates, observed via quantum oscillations in high-magnetic-fields, is a central question. It is thus of a great intrest to determine the mechanism of the formation of the magnetic-field-induced 3D CDW order, and thus, to establish whether the high-magnetic- field state is the property of the copper-oxygen plane or if other building blocks contribute to its formation.

We addressed this question by performing X-ray absorption spectroscopy (XAS) in high magnetic fields (up to 30 T) in high quality of single crystals of YBCO. YBCO features two nonequivalent Cu positions: Cu(1) in the Cu-O_d chains and Cu(2) in the CuO_2 planes. While the Cu-O_d chains host both Cu^+ and Cu^{2+}, the CuO_2 layer is occupied exclusively by Cu^{2+}. X-ray absorption spectroscopy, as a site selective probe, allows to determine which of the Cu atoms contribue in the formation of the magnetic field induced 3D CDW order, i.e., in another words, if the charge correlations are related exclusively to the CuO_2 planes or the Cu-O_d chains contribute in its formation. These nonequivalent Cu^+ and Cu^{2+} atoms give rise to the two distinct pre-peaks in the X-ray absorption spectra at the Cu *K*-resonance (see Fig. 1 and Fig. 2) We have used this fact, and observed the evolution of the two peaks as a function of magnetic field.

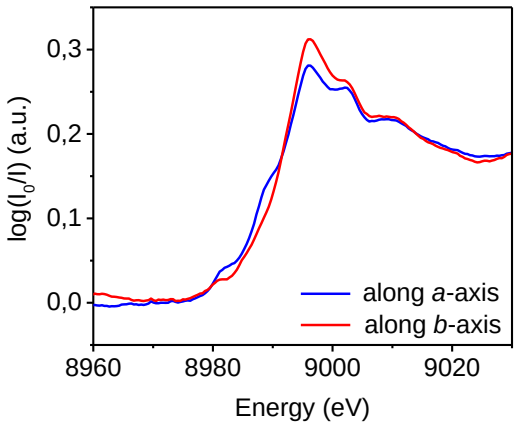

Fig. 1 X-ray absorption spectra of YBCO with the polarization along *a*-axis (perpendicular to the Cu-O chains) and *b*-axis (parallel to them) at 300 K and without magnetic field.

The single crystals of underdoped YBCO (δ=0.55 and δ=0.67) of ca 1×1 mm^2 had its *a*-*b* plane parallel to the surface and are 10-12 μm thick (We have also performed test measurements without magnetic field on single crystal of underdoped $HgBa_2CuO_{4+d}$). Cu K-edge spectra were collected in two polarizations (along *a* and *b* crystallographic axis) at energy range from 8 to 10 keV. The beam was polarized linearly, but sample was

rotated by 90 degrees to obtain required orientation. Fig. 1 compares the spectra obtained in two polarizations. The apparent changes are observed at the pre-edge peaks (~8985 eV) when the X-ray beam is polarised either perpendicular or parrallel to Cu-chains, respectively. The measurements were performed below and above the various characteristic temperatures (at 10 K, 20 K and 40 K, i.e. at $T < T_C$, at 60 K, i.e. $T \sim T_C$, at 150 K, i.e. $T \sim T_{CDW}$ and at 290 K, i.e. at $T > T^*$).

The full-energy spectra were collected as a function of the magnetic field using the 30 T pulsed-magnet setup. For a single field ~~im~~pulse, duration of 23 ms, spectra were collected every 1 ms. Each dataset included more than fourty XAS spectra corresponding to the sequent field value. The high sensitivity of the setup and high stability of the magnet installation allowed to obtain high quality data in fields up to 30 T and observe the 2D to 3D CDW transition.

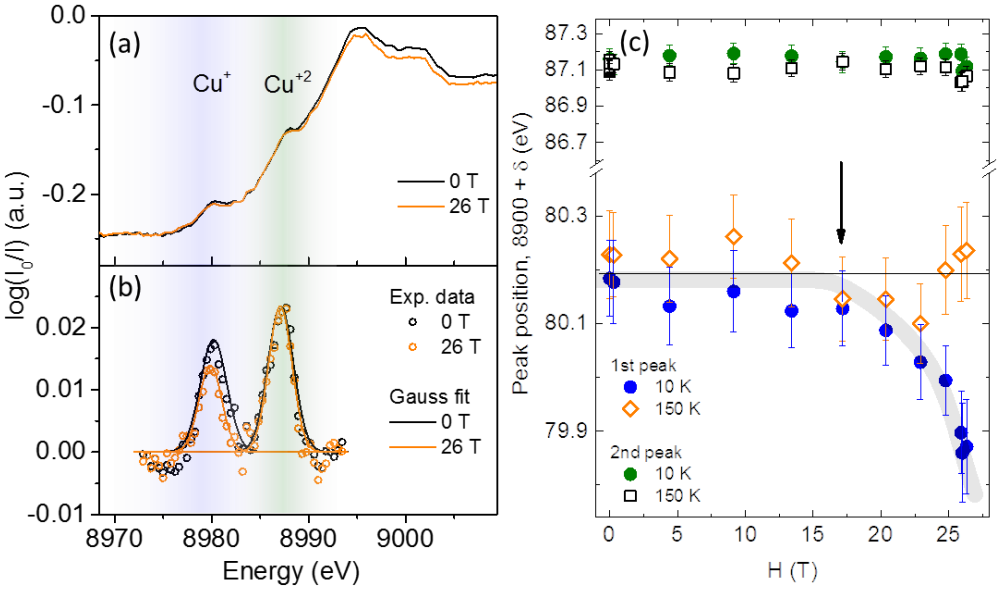

Fig. 2 a. X-ray absorption spectra of YBCO with the polarization along *a* axis at 10 K (i.e. also perpendicular to the Cu-O_d chains). The absorption edge features two distinct peaks originated from two inequivalent Cu valences, Cu^+ and Cu^{2+} at 8983 and 8988 eV, respectively. **b.** Background subtracted spectra at the Cu K-edge. The two pre-peaks are prominent. **c.** Magnetic field dependence of the pre-peak energy at 150 K and 10 K. Above approximately 18 T the Shift of the lower-energy (Cu^+) peak, corresponding to the copper atoms occupying exclusively the Cu-Od chains, is observed at 10K, i.e. below the onset temperature of the CDW order. The black arrow marks the value of the magnetic field at which the field-induced charge order was observed by X-ray diffraction [3].

Figure 2 summarizes the most important results. While the magnetic field has no effect on the absorption spectra at the temperature above the CDW order (T_{CDW}), the lower-energy peak changes as the magnetic field is applied; the peak's position, in the energy scale, shifts towards lower values. The onset of the shift is coincident with the onset of the field-induced CDW order observed in X-ray diffraction experiments [3], and thus we ascribe this shift as due to the appearance of the field-induced charge correlations. The observation of the lower-energy peak shift (associated to Cu^+ ions within the Cu-O_d chains) indicates that the Cu-O_d chains, characteristic for YBCO, contribute in the formation of the high magnetic field induced 3D CDW order. This initial result points towards the possibility that the magnetic field induced state may be specific to YBCO due to the underlying crystal structure (e.g. the existence of the Cu-O_d chains, orthorhombic distortion).

The data are under detailed analysis accompanied by FDMNS calculations of the XAS spectra, in search for the possible electronic/atomic changes giving rise to the field induced CDW order. Those preliminary experimental results presented above are very important for further modeling of the high-magnetic field state of the cuprates but requires further study, also in other cuprates. The experiment demonstrates the feasibility of pulsed field-XAS technique in studies of CDW order and the continuation of this research is needed.

References:

[1] S. Caprara *et al.*, arXiv:1604.07852 (2016)
[2] see for example W. Tabis *et al.*, Nat. Commun. **5** 5875 (2014)
[3] S. Gerber *et al.*, Science 350, 949 (2015)
[4] T. Wu *et al.*, Nat. Commun. **6**, 6438 (2015)