ESRF	**Experiment title:** The hidden chirality in high Tc cuprate superconductors studied by non-magnetic x-ray circular dichroism	**Experiment number**: HC-1275
Beamline: ID32	**Date of experiment**: from: 26/11/2014 to: 2/12/2014	**Date of report**: 27/2/2015
Shifts: 18	**Local contact(s)**: N.B. Brookes	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Giacomo Ghiringhelli * Politecnico di Milano, Italy

Lucio Braicovich * Politecnico di Milano, Italy
Bernhard Keimer Max Planck Institute for Solid State Physics, Stuttgart, Germany
Marco Salluzzo CNR/SPIN – Napoli - Italy
Nicholas Brookes * ESRF
Greta Dellea * Politecnico di Milano, Italy
Mathieu Le Tacon Max Planck Institute for Solid State Research, Stuttgart, Germany
Gabriella M. De Luca * CNR/SPIN – Napoli – Italy
Matteo Minola * Max Planck Institute for Solid State Research, Stuttgart, Germany
Yingying Peng * Politecnico di Milano, Italy
Cassiano Langini * Politecnico di Milano, Italy
Nunzia Coppola * CNR/SPIN – Napoli – Italy

Report:

This experiment aimed at detecting non-magnetic circular dichroism in the soft x-ray absorption spectra of high Tc cuprate superconductors (HTS). The motivation was the recent theoretical proposal that the Kerr rotation observed in several cuprates [1] after exposure to intense magnetic fields could be due to the chiral arrangement of charge modulation patterns, usually named as “charge order” or “charge density waves”. Charge order can be detected by scattering experiments, where the incommensurate ordering wave-vector lies in the CuO_2 planes. Along the *c*-axis, perpendicular to the superconducting CuO_2 planes, charge density waves might organize in some sort of chiral pattern, which could be at the origin of the Kerr rotation of the light [2]. However another possible explanation could be that the Kerr rotation is due to time reversal symmetry breaking, *i.e.* some sort of latent magnetic order, initially set by a high external magnetic field [3]. In fact the Kerr rotation sign can be reversed by “training” the sample in a several tesla field at room T, followed by zero-field cooling. Finally other explanations of the Kerr rotation are based on the so-called Varma currents, sort of local intra-plaquette loops whose arrangement is supposed to lead to magnetic anapoles [4].

Cu $L_{2,3}$ and O K edge XAS is usually an ideal tool for the study of symmetry breaking in 3*d* transition metal compounds: circular dichroism can detect magnetization, and linear dichroism can detect magnetic and orbital anisotropy. In layered cuprates we have already used XMCD to measure weak (ferro)magnetism in undoped and superconducting samples [5,6], and we have tried here to use circular dichroism to detect possible chiral order and/or anapoles. The former should give rise to X-ray Natural Circular Dichroism (XNCD), that is a difference in the XAS measured with left- and right-circularly polarized photons, while averaging over the magnetization state of the sample (*i.e.* when averaging the spectra over the two signs of the magnetic field). The latter should manifest itself as X-ray Magneto-Chiral Dichroism (XMχD), that is a difference in the XAS measured with opposite magnetization but with unpolarized radiation [7].

We have performed a complete XMCD-XNCD-XMχD experiment on Nd1+xBa$_{2-x}$Cu$_3$O$_{7-\delta}$ (NBCO) films grown on STO, at T=10K, 50K, 90K and 300K and at low (±0.03T) and high (±8T) magnetic field along the incident beam. We have studied one antiferromagnetic film, undoped and thus without charge order and/or Varma currents; one underdoped (T_c = 49 K) and one optimally doped (T_c = 90 K) samples, both presenting charge order, although more stable and stronger in the former than in the latter. We have also measured, at T=10K, two La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) films, x = 0.15 for the optimally doped (T_c = 30 K) and x = 0.05 for the underdoped (insulating) one.

In November 2014, this was the very first official experiment of the new ID32 beam line, which had seen the first beam through the monochromator of the XMCD branch a just few weeks earlier. Despite the partly unsettled working conditions and some incomplete commissioning, the measured XAS spectra were of excellent quality. The electronic noise was extremely small and the main limitations came from some energy jitter and drifts that we managed to correct almost perfectly in the data processing. It was ultimately more difficult to account for a residual non-linearity in the I_0 measurement that was different for the two photon polarizations (LCP and RCP) and somehow sensitive also to the sign of the magnetic field set in the high field electro-magnet. This non-linearity has introduced a systematic spurious contribution in the XNCD and XMχD spectra that we have not been able to completely correct in the data processing, leaving a residual artificial signal of less than 1% (in some cases as low as 0.2%), nevertheless totally dominant with respect to any possible genuine signal. In fact the chiral and magneto chiral effects are expected to be extremely small in the soft x-ray XAS, and they have never been detected before to our knowledge. This is due to the fact that, whereas XMCD is proportional to the electric dipole operator squared (E1^2) for the given core-to-valence transitions, XNCD and XMχD are proportional to the E1E2 and E1M1 operators, where E1 (E2) is the electric dipole (quadrupole) and M1 is the magnetic dipole operator [7,8]. In our case the spectra are totally dominated by the 2p$\rightarrow$3d and 1$s\rightarrow$2p E1-allowed transitions; on the other hand the E2 and M1 allowed transitions are 2$p\rightarrow$4p for Cu L_3 and 1$s\rightarrow$3s or 1$s\rightarrow$3d for O K edge. As a consequence, a *p-d* mixing of Cu states and *s-p* or *d-p* mixing of oxygen states are required in order to obtain non-zero XNCD and XMχD signals. The signatures of these orbital hybridizations can be extremely small. This experiment was an explorative attempt to detect this kind of effects in the soft x-ray regime, in a system where XMCD is known to be detectable. As mentioned above, despite the excellent quality of the XMCD spectra, the XNCD and XMχD spectra were affected by residual instrumental asymmetries hindering the detection of genuine effects. Therefore hereafter we summarize only the XMCD results.

At the Cu L_3 edge, in a strong field, a paramagnetic XMCD has been found in LSCO and NBCO at different dopings and at various temperatures (Figure 1). With respect to reference [5] we have found here a non-zero XMCD up to room T (see Figure 2), thanks to the improved experimental conditions. Moreover we find that the two NBCO samples have comparable integrated XMCD (~0.2%) at 90 K and 300 K, but they behave differently at lower temperature.

The other original result is the first detection (to the best of our knowledge) of O K edge XMCD in HTS. In LSCO, at 10 K and 8 T the effect is extremely small, although visible ($<5\times10^{-4}$). In NBCO, probably due to the presence of the Nd magnetic ion in the structure, the effect is much larger (Figure 3). These results demonstrate that weak magnetism, driven by the Dzyaloshinskii–Moriya interaction, is present up to room temperature in NBCO, where a magnetic ion is present in the charge reservoir layer. It is still unclear whether this high T magnetism, clearly mediated by the oxygen states, is partly or completely localized in the Cu(1) sites of NBCO, *i.e.* outside the CuO$_2$ superconducting planes. On the other hand, further attempts to measure XNCD and XMχD signals should be carried out in the future, once the beam line commissioning will have significantly reduced the residual instrumental asymmetries. For the moment the questions about the possible chirality in the charge order of HTS remain unanswered by XAS dichroism in the soft x-ray regime.

[1] Jing Xiaet al. Phys. Rev. Lett **100** 127002 (2008)
[2] P. Hosur et al Phys Rev B **87** 115116 (2012)
[3] P. Norman, Phys. Rev. B **87** 180506 (2013)

[4] Pershoguba et al., Phys. Rev. Lett. **111** 047005 (2013)
[5] G.M. De Luca et al, Phys. Rev. B **82**, 214504 (2010)
[6] G.M. De Luca et al Nat. Comm **5** 5626 (2014)|
[7] Goulon, J. et al. Phys. Rev. Lett. **88**, 237401 (2002).
[8] R. Sessoli et al, Nat. Phys. **11**, 69 (2014)

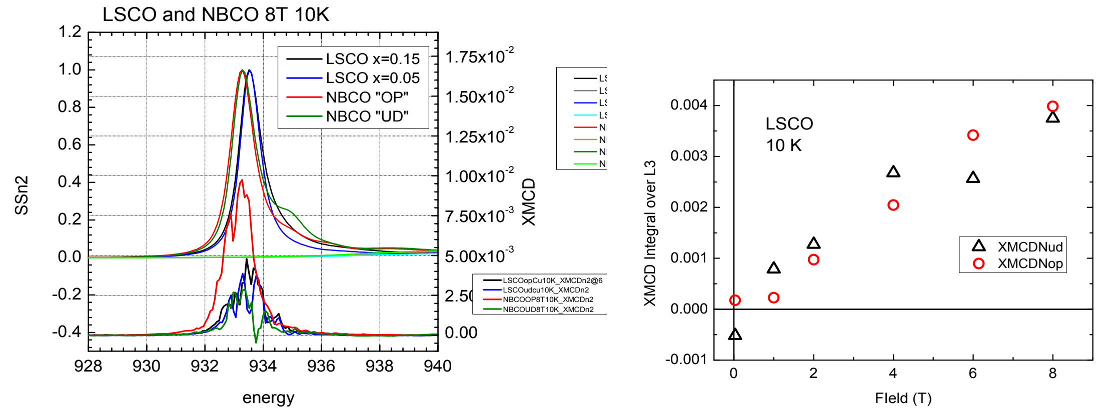

Figure 1. Comparing Cu L_3 MCD of LSCO and NBCO. Right panel: the field depencenc in LSCO. The NBCO sample labelled “OP” is underdoped,(Tc=49K), the NBCO sample labelled “UD” is optimally doped (Tc=90K).

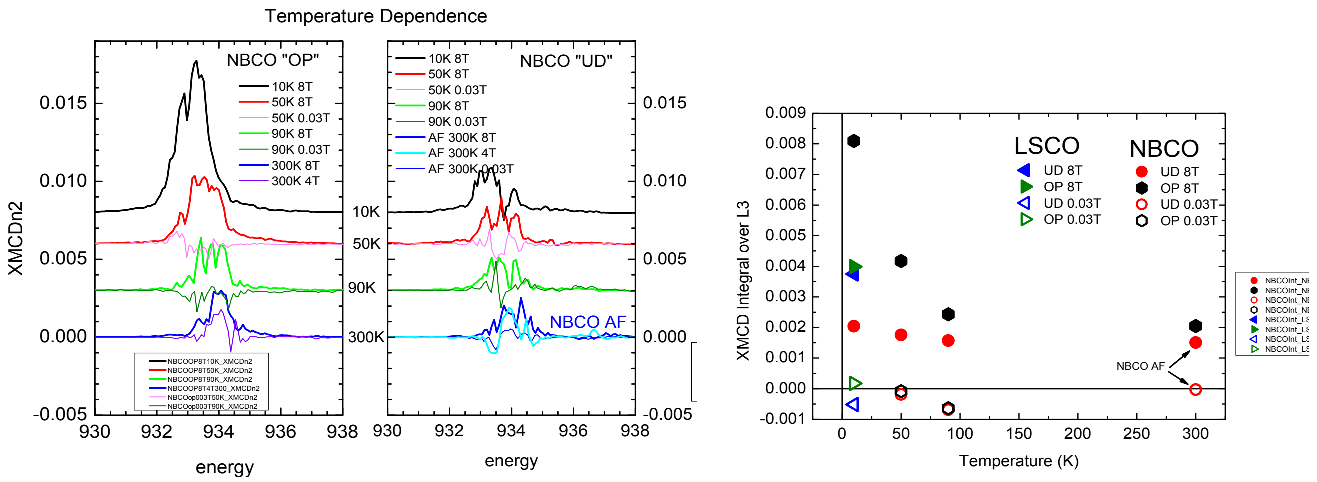

Figure 2. T dependence of the Cu L_3 XMCD in NBCO. The thick lines are for field 8 T, the thin lines are for 0.03 T measurements, shown to provide the error bar of the experiment. The shift in the peak position is due to a drift of the beam line monochromator on a timescale of several hours. The results are summarized in the right hand panel, showing a different behavior for the two doping of NBCO.

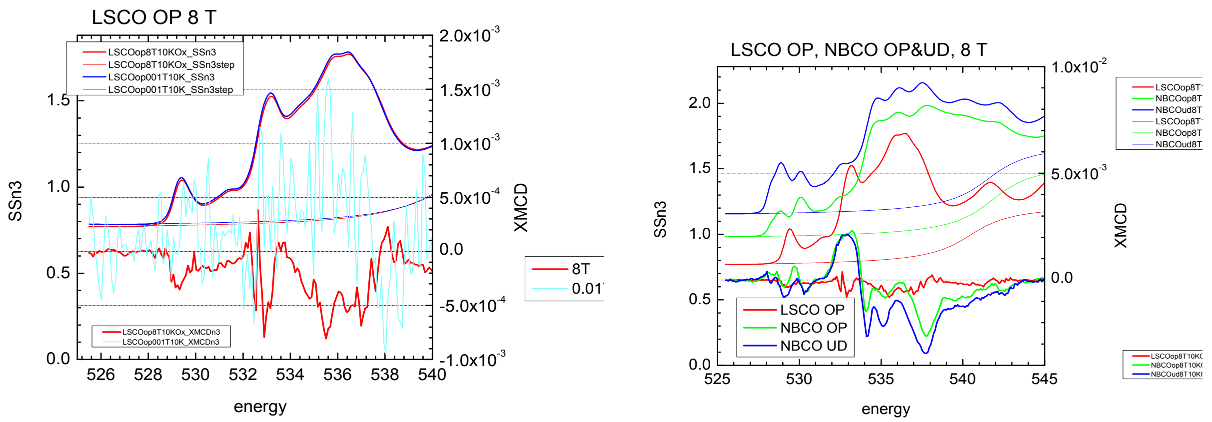

Figure 3. The O K edge XMCD of LSCO and NBCO at 10 K. The dichroic spectra in NBCO are dominated by peaks at 533 eV and 538 eV, corresponding to O 2p - Nd 4p hybrid final states several eV above the fermi level.