# Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF**.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

## Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

## Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

| ESRF | **Experiment title:** X-ray natural circular dichroism in a charge-ordered prototype cuprate superconductor La1.875Ba0.125CuO4 | **Experiment number**: 32299 |
|---|---|---|
| **Beamline**: ID12 | **Date of experiment**: from: Oct. 30, 2013 to: Nov. 5, 2014 | **Date of report**: Feb. 28, 2014 |
| **Shifts:** 18 | **Local contact(s)**: WILHELM Fabrice | *Received at ESRF:* |


**Names and affiliations of applicants** (* indicates experimentalists):

JUNFENG HE, Boston College (BC), K. SATO, Tohoku, K. TANAKA, Osaka, X.N. SUN, CNRS, I. KENJI, SPring-8, S.I. FUJIMORI, JAEA, F. WILHELM, ESRF, S. MOTOHIRO, N. KAWAMURA, SPring-8, M. FERNANDEZ, G. GU, Brookhaven, A. ROGALEV, ESRF, M. FUJITA, Tohoku, RUI-HUA HE, BC (All coauthors are experimentalists.)


## Report:

During the beamtime, we paid particular attention to the possible contamination of elastic scattering/ diffraction signals in the XAS and dichroic spectra. As these signals are highly angle dependent, they usually showed up in individual detectors mounted at different angles with respect to the sample (but not all of them), and can be removed from the interested energy range by tweaking the sample polar angle θ (the angle between the X-ray wavevector and the c-axis of the crystal) only slightly (within ~1 degree).

We first observed a dichroic signal which is largely temperature independent (Fig. 1a) and whose overall magnitude increases with the increase of the incidence angle of light away from normal incidence (Fig. 1b). We ascribed this signal to the x-ray natural linear dichroism (XNLD), which reflects the crystallographic asymmetry between the a-axis and the c-axis that was probed upon switching the residual linear polarization components of the incident light with ~90% circular polarization.

Interestingly, near normal incidence where XNLD is small, we observed an additional dichroic signal emerging on top of the XNLD signal upon decreasing temperature (Fig. 1c). Its strength showed an abrupt onset across ~42 K (Fig. 1d). After removal of the XNLD background, this low-temperature signal features a distinct peak at ~8.99 keV and a few sign changes right below the edge (Fig. 1e).

As LBCO-1/8 is in the low-temperature tetragonal (LTT) structural phase below 54 K which is uniaxial, no XNLD signal is expected from any ab-plane crystallographic asymmetry, although one would need to study the sample azimuthal rotation angle φ (about the c-axis) dependence of the low-temperature signal to firmly exclude its XNLD nature; for which XNLD varies as cos2φ while XCD should be φ independent [1]. Alternative, the signal strength of a XCD signal should follow the magnitude of the circular polarization of the incident light that we can adjust and compare the results.

Therefore, the observed signal is likely due to XCD, whether magnetic (XMCD) or natural (XNCD). Indeed, a spin stripe order develops around 42 K. Canted magnetic moments in an antiferromagnetic order can in principle give rise to weak ferromagnetism that can manifest in XMCD. Nevertheless, this type of XMCD signal appears unlikely to be relevant here. First, magnetic moments on Cu predominantly involve 3d electronic states. Their XMCD signal should peak at ~8.983 keV corresponding to the (dipole-forbidden) 1s-3d transition, different from the experiment. Second, unlike in a homogeneous antiferromagnetic order in the LTT phase, spin canting in the spin stripe ordered phase (<42 K) produces zero net canted moment on each $CuO_2$ plane at low field [2]. Third, with the spin canting angle set by the Dzyaloshinsky–Moriya superexchange interaction that arises from the $CuO_6$ octahedral tilting which saturates below 54 K, the temperature dependence of the weak ferromagnetism should follow that of the order parameter of the spin stripe order. The latter has been measured by neutron diffraction on the same material [3] (Fig. 1d), which shows a much more gradual onset behaviour than our observed XCD signal strength.

XMCD can also be caused by some other non-trivial forms of broken time-reversal symmetry, e.g., in the orbital current loop order [4], nevertheless, the ordering temperatures reported so far are at least many times higher than 42 K [5] and the onsets found are anything but abrupt. Therefore, what we newly observed below 42 K is very likely a XNCD signal. That being said, independent experimental differentiation between the XMCD and XNCD nature of the observed signal would be very helpful, which can be achieved by means of a polar angle θ dependence study; in which XNCD varies as $3\cos^2\theta-1$ while XMCD as $\cos\theta$ [6]. Attempts were made to study the θ dependence at 29 K in experiment (results not shown), but because of the increasing strength of XNLD background signal with θ, the result obtained was ambiguous and incomplete. A better approach is to perform for each θ measurements at two temperatures, e.g., 35 K and 45 K (with and w/o the XCD signal), and then subtract off the XNLD background to obtain the XNCD spectrum (as shown in Fig. 1e).

To the extent that the observed signal is XNCD, its onset at 42 K coincides with the crossover of the charge stripe order from 2D (above) to 3D (below), which was observed by x-ray diffraction measured with photon energies near the Cu K-edge [7]. A good correlation of the charge order along the c-axis ensures the principle axes of the local chiral orders to be aligned (with the c-axis), which is a prerequisite for the detection of a non-zero XNCD signal (because XNCD probes chirality through the pseudo-deviator part of the optical activity tensor). Kerr effect which probes the pseudo-scalar part does not require such alignment, and can in principle be sensitive to the nascent chiral order which sets in at ~50 K when it is still poorly correlated along the c-axis. Though plausible, this scenario is not sufficient to explain the apparent abruptness of the XNCD onset we observed, whose implication requires more thinking.

We also compared the potential XNCD spectrum (black circles in Fig. 1e) with the theoretical calculation [8] done for a related but different cuprate material (whose crystal structural is also achiral) based on different configurations of the chiral order. We found a reasonable agreement with the results for a particular configuration (red dashed curve in Fig. 1e), in terms of the spectral line shape, positions of features and their absolute magnitudes. We are currently working on a similar theoretical calculation for LBCO-1/8 in order to determine the actual configuration of the chiral order.

**References** **[1]** S. Di Matteo, M. R. Norman, Phys. Rev. B 76, 014510 (2007). [**2**] M. Hücker, Phys. Rev. B 79, 104523 (2009). [**3**] M. Hücker et al., Phys. Rev. B 83, 104506 (2011). [**4**] V. Aji et al., arXiv:1211.1391 (2013); S. S. Pershoguba et al., Phys. Rev. Lett. 111, 047005 (2013). [**5**] V. Baledent et al., Phys. Rev. Lett. 105, 027004 (2010); Y. Sidis, P. Bourges, J Phys Conf Ser. 449, 012012 (2013). [**6**] J. Goulon et al., J. Synchrotron Rad. 7, 182 (2000). [**7**] Y.-J. Kim et al., Phys. Rev. B 77, 064520 (2008). [**8**] M. R. Norman, Phys. Rev. B 87, 180506(R) (2013). [**9**] H. Karapetyan et al., Phys. Rev. Lett. 112, 047003 (2014).

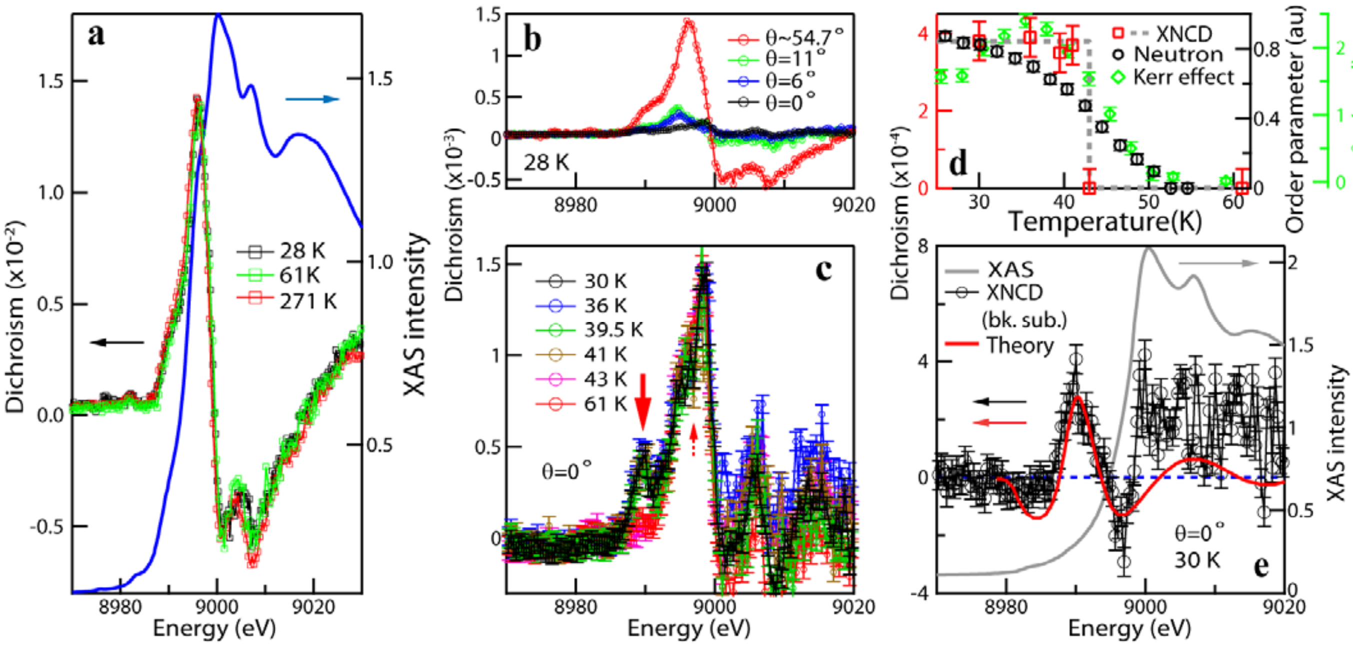


Fig. 1 X-ray absorption (XAS) and dichroic spectra measured at (a) selected temperatures and (b) θ. (c) Dichroic spectra at low temperatures and θ=0˚, showing the appearance of additional features. (d) Intensity of the dichroic feature marked by the big red arrow in c), compared with the square root of the spin ordering

neutron diffraction intensity [3] and the Kerr rotation [9] on LBCO-1/8. e) XAS and background-subtracted dichroic spectrum at θ=0˚, compared with the calculated XNCD. Note the common y-scale for dichroism.