ESRF	Experiment title: NON-RECIPROCAL X-RAY GYROTROPY IN ANTIFERROMAGNETIC SINGLE CRYSTALS	Experiment number: HE-628
Beamline: ID12-A	Date of experiment: November 14 - November 22, 1999	Date of report: 1rst Sept 2000
Shifts: 24	Local contact(s): J. GOULON and A. ROGALEV	*Received at ESRF:*

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Non-reciprocal X-ray Linear Dichroism (nr-XLD) is a new spectroscopy which refers to a differential absorption of the (σ,π) components of linearly polarized X-ray photons in magnetic *gyrotropic* media, *i.e.* in systems in which the time-space inversion symmetry is broken as a consequence of magnetic ordering. Typically, this dichroism is expected to be observable in antiferromagnetic materials that are magneto-electric. There is remarkable link between nr-XLD and X-ray Natural Circular Dichroism (XNCD), *i.e.* the differential absorption of left- and right-handed circularly polarized X-ray photons in Optically Active crystals: this link is the complex *gyrotropy tensor* ζ^* which, in the X-ray range, is dominated by electric dipole-electric quadrupole (E1.E2) interference terms in Fermi's golden rule. While XNCD refers to its *imaginary* part ζ' which is required to have an *even* time-reversal symmetry to induce differential light absorption, nr-XLD is related to the *real* part which has been recognized to have *odd* time reversal symmetry. Near resonance, the transverse dichroism is then expected to be the sum of three contributions:

$$\sigma^{\pi} - \sigma^{\sigma} \propto \left[\alpha^{\pi}_{yy} - \alpha^{\sigma}_{xx}\right] + \left[\varsigma^{\pi}_{yyz} - \varsigma^{\sigma}_{xxz}\right] + \left[Q^{\pi}_{yzy} - Q^{\sigma}_{xzx}\right]$$

where $\alpha_{\alpha\beta}$ and $Q_{\alpha\gamma\beta}$ are the time-reversal *even*, *real* parts of the electric dipole and electric quadrupole polarizability tensors. Only the second term, *i.e.* the gyrotropy contribution, is odd with respect to time-reversal symmetry. In the experiment HE-628, we have produced the unambiguous evidence that, in antiferromagnetic crystals of appropriate space-time symmetry, the contribution of the second term can be quite significant and can largely exceed the two other contributions[1]. This successful experiment has been performed at the Vanadium K-edge in the antiferromagnetic insulating (AFI) phase of a Cr doped V_2O_3 Mott crystal in which a single antiferromagnetic domain was grown by magneto-electric annealing. This was achieved by applying simultaneously a modest electric field (5.10^3V/cm) plus a weak Magnetic field (±0.5T)

both oriented along the trigonal c axis while the crystal was heated up in the paramagnetic phase (T=300K). The crystal was then slowly cooled down well below the phase transition temperature (T_N =181K) and the electric field was switched off during the XLD measurements. For these experiments, we used the helical undulator source HELIOS-II in combination with a diamond Quarter-Wave Plate (QWP) inserted after the Si(111) double crystal monochromator: the QWP was used to convert circularly polarized photons into (σ,π) linearly polarized photons and to switch quickly from one linear polarization to the orthogonal. The crystal being cleaved perpendicular to the c axis, the incident X-ray beam was strictly parallel to the crystal c axis and the fluorescence detected dichroism spectra were recorded in the backscattering mode. As illustrated by Fig. 1, in the paramagnetic trigonal phase $R\bar{3}c$, *i.e. **before*** annealing, we measured only a very weak XLD signal ($<3.10^{-4}$) in the preedge region -possibly due to a small misalignment of the crystal-.

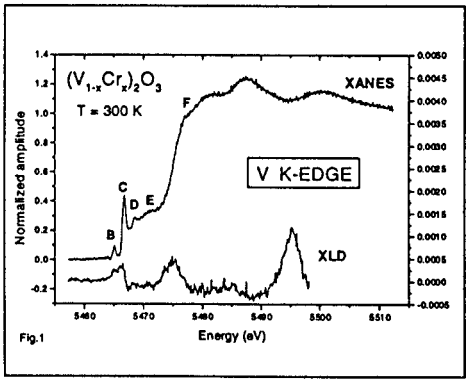

Fig.1

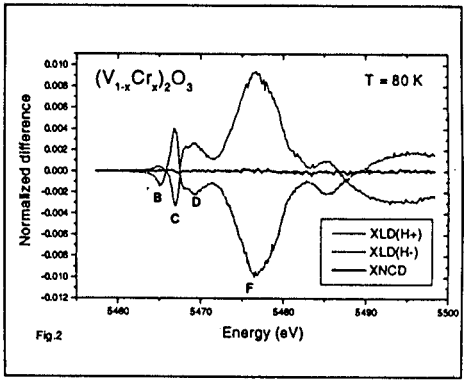

Fig.2

The XLD spectra recorded in the AFI phase ***after*** magneto-electric annealing (Fig.2) show a spectacular increase in the maximum intensity (*ca.* 1%). As expected for a time-reversal *odd* effect, the sign of dichroism is nicely inverted when the relative direction of the magnetic field is inverted with respect to the electric field during the magnetoelectric annealing process. Recall that such an inversion is not observed for conventional X-ray Magnetic Linear Dichroism (XMLD). Although the local point group C3 at each metal center is chiral, the space group I2/a belongs to a centrosymmetric class for which no XNCD should be observed. In practice, we measured a very weak residual XNCD signal ($<1.510^{-4}$). The non-reciprocal effect displayed in Fig.2 can thus only be explained if the inversion symmetry is broken by the magnetic order as expected in the space-time magneto-electric group A2 of V_2O_3.

There are 58 non-centrosymmetrical magnetic crystal classes compatible with nr-XLD but not all geometries are suitable to detect the effect. We failed to measure nr-XLD with a Cr_2O_3 crystal cleaved perpendicular to the c axis. Below the Néel temperature, this crystal has the magneto-electric space-time group $\underline{\bar{3}m}$ but since the easy magnetization is along the trigonal axis, there cannot be any strong transverse gyrotropic anisotropy. In a preliminary experiment, we detected a significant irreversible dichroism which we interpreted as resulting from spin-flop effects since we performed the annealing under too strong magnetic fields.

Reference:

[1]J. Goulon, A. Rogalev, C.Goulon-Ginet, G. Benayoun, L. Paolasini, C. Brouder, C. Malgrange and P.A. Metcalf, revised manuscript submitted to *Phys. Rev. Lett.*