



	Experiment title: NON-RECIPROCAL X-RAY OPTICAL ACTIVITY IN ANTIFERROMAGNETIC SOLIDS	Experiment number: HE-968
Beamline: ID-12	Date of experiment: from: 25-AVR-2001 to: 02-MAY-2001	Date of report: 01-MAR-2002
Shifts: 21	Local contact(s): A. ROGALEV and F. WILHELM	<i>Received at ESRF:</i>
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Unlike magneto-optical spectroscopies, *e.g.* Faraday rotation, Magnetic Circular or Linear Dichroisms (MCD,MLD), which are all consistent with the usual electric dipole approximation, Optical Activity (OA) mixes multipole moments of opposite parity and thus requires **odd space parity**. Typically, OA of X-rays is caused by electric dipole-electric quadrupole E1.E2 interference terms¹. It has long been argued that OA effects could be either *even* or *odd* with respect to **time-reversal**: time-reversal even OA properties are called *natural* whereas time-reversal odd properties are called *non-reciprocal*. A non-reciprocal X-ray magnetic linear dichroism (nr-XMLD) has recently been measured² at the ESRF. We report below another non-reciprocal effect in the X-ray range which we called *X-ray Magneto-chiral Dichroism* (XM χ D). We stress that XM χ D, unlike X-ray magnetic circular dichroism (XMCD), does not require any polarized beam since it is a property of the Stokes component S_0 .

Magnetolectric (ME) solids are good candidates to detect XM χ D because magnetolectric properties are odd with respect to Parity (P) and time reversal (Θ) but are invariant in the product $P\Theta$. The generic example of ME crystals is Cr_2O_3 : it has the centrosymmetric corundum space group ($R\bar{3}c$) but it belongs to the non-centrosymmetric $\bar{3}'m'$ space-time group below the Néel temperature. The spin moments can order in either one of two 180° domains: one can grow such single domains by **magnetolectric annealing**: it consists in heating the crystal in the paramagnetic phase and in applying simultaneously along the **c** axis a modest electric field E (5 kV/cm) plus a weak magnetic field H ($\pm 0.5\text{T}$). All spectra were recorded in the fluorescence excitation mode using the most convenient backscattering configuration. We produced artificially unpolarized light by incoherent superposition of fluorescence excitation spectra recorded with Right and Left circularly polarized incident photons: $F_0 = F[\text{Rcp}] + F[\text{Lcp}]$. We have reproduced in **Figure 1** the XM χ D spectrum measured at $T=50\text{K}$ of a (001) Cr_2O_3 single crystal with the **c** axis parallel to the wavevector **k**.

It is shown that the signal can be as large as 1.6% due to the strong contribution of the E1E2 interference terms in the X-ray regime. It also appears from **Figure 1** that the same XM χ D spectrum can be obtained using a *powdered pellet* of Cr₂O₃ : the price to be paid is, however, a reduction (1:6) of the amplitude of the signal whereas the theory of XM χ D let us expect a slightly smaller reduction (1:5). Whereas X-ray natural circular dichroism (XNCD) can only be detected in single crystals, it is quite remarkable that XM χ D can be measured in a powder due to the fact that the orientational isotropy is broken by the ME order.

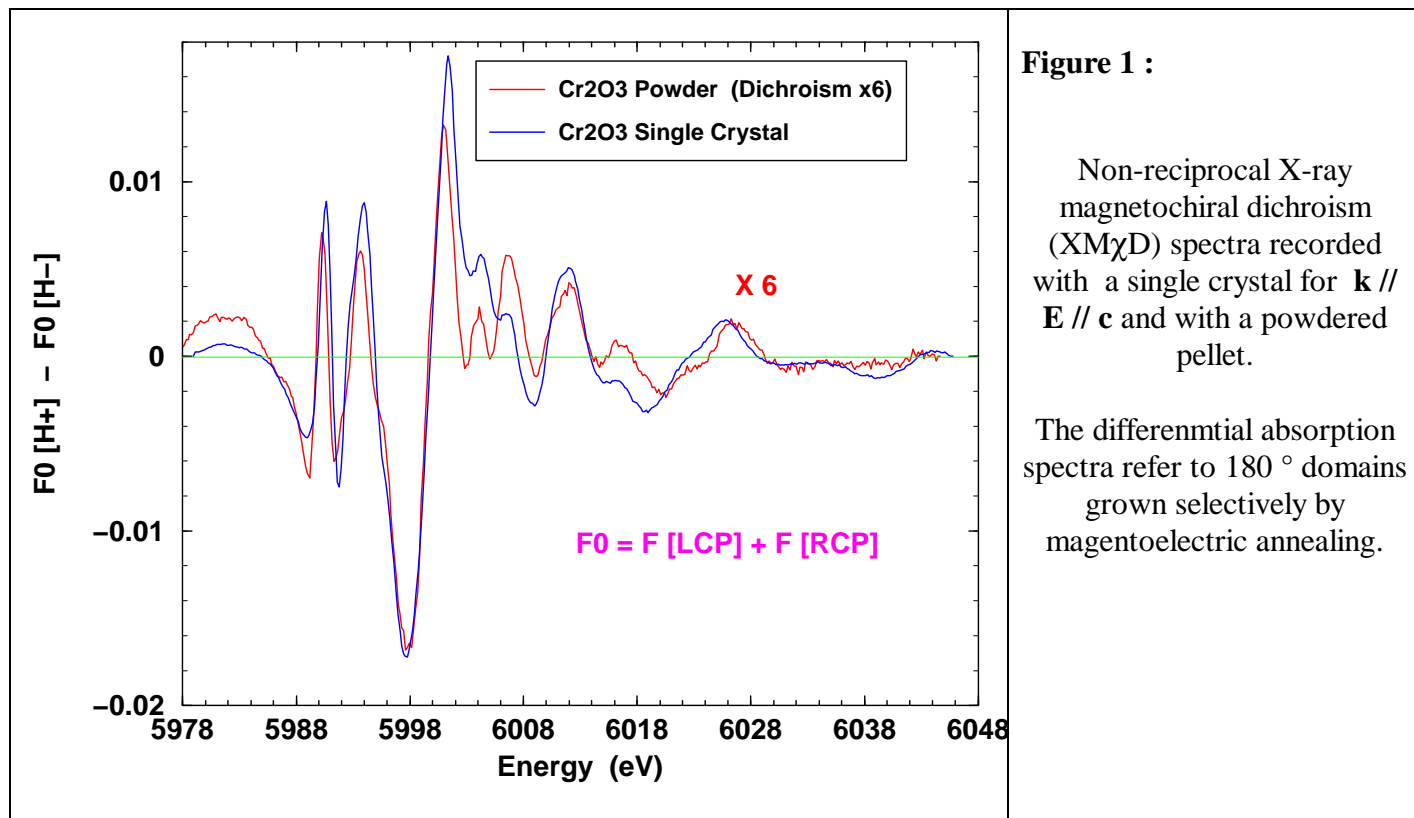


Figure 1 :

Non-reciprocal X-ray magnetochiral dichroism (XM χ D) spectra recorded with a single crystal for $\mathbf{k} // \mathbf{E} // \mathbf{c}$ and with a powdered pellet.

The differential absorption spectra refer to 180 ° domains grown selectively by magnetoelectric annealing.

At this stage, there is no existing *ab initio* computer program which would allow one to simulate properly our XM χ D spectra but, nevertheless, some valuable information can yet be extracted from the edge selective OA sum rules derived recently by Carra and co-workers. The effective operator Ω_z^- which describes the mixing in the ground state g of p et d atomic orbitals at the Cr absorbing site was identified with the *orbital anapole*. According to group theory, the universally cited magnetic group $\bar{3}'m'$ of Cr₂O₃ is *not* compatible with an invariant anapole moment. There is, however, no deep contradiction with our experiment : our XM χ D spectra reveal that this magnetic group may be suitable to describe the *spin* configuration but not the overall magnetic symmetry including orbital moments and currents. In other words, the spin anapole moment of Cr₂O₃ certainly vanishes but it is our interpretation³ that the true space-time symmetry is most probably only $\bar{3}'$ since this group admits the anapole moment Ω_z^- as invariant.

References:

1. J. Goulon, C. Goulon-Ginet, A. Rogalev, V. Gotte, C. Malgrange, Ch. Brouder and C.R. Natoli, *J. Chem. Phys.* **108**, 6394-6403 (1998)
2. J. Goulon, A. Rogalev, C. Goulon-Ginet, G. Benayoun, L. Paolasini, Ch. Brouder, C. Malgrange and P.A. Metcalf, *Physical Review Letters*, **85**, 4385-8 (2000)
3. J. Goulon, A. Rogalev, F. Wilhelm, C. Goulon-Ginet, P. Carra, D. Cabaret and Ch. Brouder, *Physical Review Letters* (2002) *submitted*