ESRF	**Experiment title:** SKEW POLARISATION OF X-RAYS IN GYROTROPIC CRYSTALS	**Experiment number:** **HE-969**
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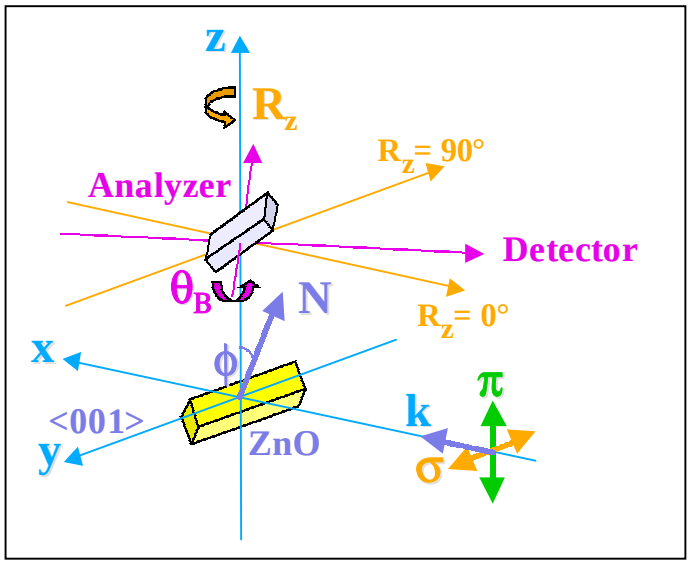
1. POLARIZED HIGH RESOLUTION X-RAY EMISSION SPECTROSCOPY : GOALS

Our 1rst goal was to check how far the *pseudo* reduction of the core hole life-time[1] in XANES spectra recorded using a high resolution crystal analyzer would result in enhanced linear (or circular) dichroism signatures. This led us to concentrate our efforts on a uniaxial single crystal of ZnO (Würtzite type of structure: $P6_3mc$) which exhibits a large linear dichroism (LD) respectively along the ***a*** and ***c*** axes. Another objective was to try to look for anisotropy in the Resonant Inelastic X-ray Scattering (RIXS) channels by analyzing the polarization of the photons emitted along a well-defined direction.



The geometry of the experiment is illustrated by **Fig. 1**. A diamond Quarter-Wave-Plate (QWP) was used to convert the circularly polarized (CP) X-rays emitted by the helical undulator HU38 into a (σ) or (π) linearly polarized beam with the capability to flip rapidly the polarization direction. The (001) ZnO crystal was slightly inclined by $\phi \approx 5°$. The emitted fluorescence (or Raman scattered) photons were analyzed along the vertical axis (z) using a spherically bent Si (440) crystal analyzer. The incident X-ray beam was focused down to a spot size of 30x100 μm^2 at the sample location in order to preserve the energy resolution. Since the Bragg angle θ_B was of the order of 48.4° , the analyzer was acting as a linear polarimeter selecting the polarization component normal to the diffraction plane. A rotation R_z of the whole analyzer from 0° to 90° allowed us to explore the anisotropy of the emission channel around the vertical axis.

Since ZnO belongs to the 6mm class of non-centrosymmetric crystals, it should exhibit a *skew polarization effect*[2] associated with the irreducible representation of natural Optical Activity that transforms as a *polar vector*[3] with respect to the rotation group SO_3. It was our ultimate goal to investigate whether such a *skew polarization* would induce some additional anisotropy in the polarized RIXS spectra.

2. RESULTS

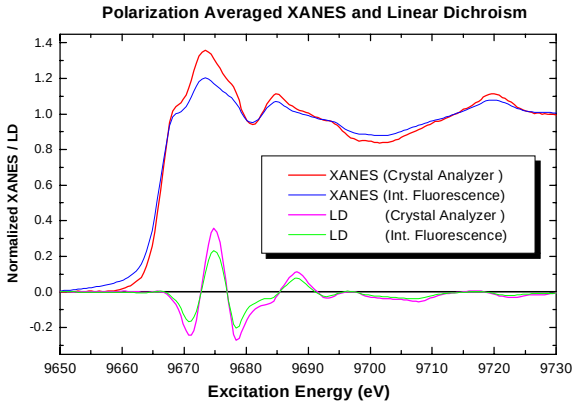


Fig. 2 *Polarization averaged* (σ,π) XANES and LD spectra recorded simultaneously with the analyzer and 8 photodiodes collecting the integrated fluorescence in a conventional backscattering geometry.

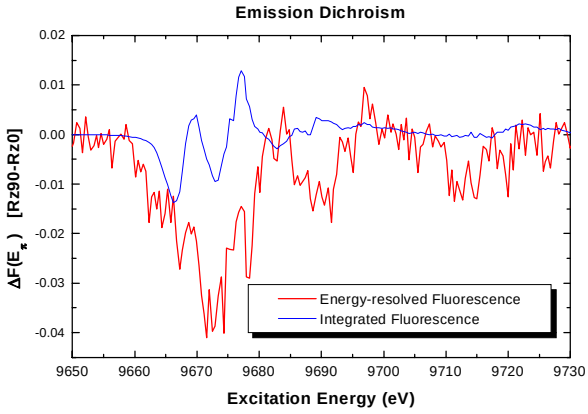


Fig. 3 Anisotropy of the emission channel under excitation by a π–Polarized incident X-ray beam. It exceeds the effects due to the beam instabilities monitored by the photodiodes in a backscattering geometry.

As illustrated by Fig. 2, LD dichroism signatures recorded with our high resolution emission spectrometer are quite significantly enhanced with respect to LD spectra recorded by collecting the integrated fluorescence in the common backscattering geometry. The same conclusion holds true for the shape resonances of the (σ,π) polarization averaged, high resolution XANES spectra which also exhibit much sharper edges as already noted by F. de Groot for Platinum at the L_3 edge[1]. Figure 3 is interesting since it shows that there is only a *very small polarization anisotropy* of the radiation scattered inelastically in the vertical direction for a π-polarized excitation whereas there is a strong crystal anisotropy parallel and perpendicular to the optical axis. This would suggest that there is very little angular correlation left between the relative orientations of the absorbing and emitting dipoles[4]: one is led therefore to the conclusion that the emission process could be coupled to a long-living excited state but the orientational correlations would be lost due to very fast many-body interactions. We believe that the signal reproduced in Fig.3 is real since it is not correlated with beam instabilities. It seems to be also fairly different from a pure E1E1 emission dichroism. Note that our experimental configuration was *a priori* propitious to the detection of the vector part of optical activity associated with E1E2 interference terms because the optical axis **c** was oriented perpendicular to the plane of scattering while $\mathbf{k_{in}}$ and $\mathbf{k_{out}}$ were strictly orthogonal. Unfortunately, as revealed by the spectra recorded with the photodiodes in the backscattering geometry, angular drifts of the incident beam in the horizontal plane have clearly contaminated the data recorded with a σ-polarized excitation and no definitive interpretation can be given for the spectrum displayed in Fig. 3.

The excitation spectra reproduced in Figs 2 and 3 were recorded at the fixed energy of the $K\alpha_1$ line. We noted, however, a small but quite significant dispersion in energy of the emission spectra for excitation energies close to the white line. Preliminary tests have been made to record LD spectra in a quasi constant transferred energy mode. Unfortunately, the quality of these data was insufficient to draw reliable conclusions due to, again, beam instabilities.

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