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1. XMLD AS A NON-LINEAR PROBE OF MAGNETIZATION DYNAMICS ?

So far, only X-ray magnetic *circular* dichroism (XMCD) was used to *probe* locally the resonant precession of either orbital or spin magnetization components in response to a strong microwave field h_p oscillating along a direction which is orthogonal to the magnetic bias field H_0 in standard FMR experiments that are most often carried out in the so-called perpendicular pumping geometry^{1,2}. It was the aim of proposal HE-3139 to explore whether one could exploit X-ray Magnetic *Linear* Dichroism (XMLD) to probe new aspects of the dynamics of the resonant precession^{2,3}.



Whereas XMCD is time-reversal *odd*, XMLD is time-reversal *even*, *i.e.* its sign does not depend on the direction of the static bias field **H**₀. This implies that XMLD can only probe *non-linear* susceptibilities $\chi^{(0)}$ and $\chi^{(2)}(2\omega)$. Depending on the pumping geometry, one may probe a variety of non-linear processes of different nature: 2nd order effects involving the uniform precession mode (k=0) are best investigated in perpendicular

pumping whereas non-linear processes resulting in the direct excitation of spin-waves ($k \neq 0$) are most easily investigated in *parallel* pumping geometry^{2,4}. *Oblique* parallel pumping is a convenient way to combine both options since the corresponding non-linear processes contribute to well resolved distinct spectral branches. Owing to the weakness of the expected XDMR signal, the *transverse* detection geometry did look to us most favourable since we could hope to measure *cross-terms* in the series expansion :

 $\Delta \sigma_{\text{XMLD}} \propto [M_z]^2 + 2M_z \Delta M_z^{(0)} + 2M_z \delta M_z^{(2)} (2\omega) + [\Delta M_z^{(0)}]^2 + \dots$ (1) in which M_z refers to the equilibrium magnetization, $\Delta M_z^{(0)}$ being the time-invariant change of M_z probed with XMCD in longitudinal XDMR geometry, whereas $\delta M_z^{(2)}(2\omega)$ is another nonlinear term due to elliptic precession. The highest priority was thus given to experiments carried out in parallel pumping geometry on the same YIG/GGG thin film that was used for our previous CP-XDMR experiments. What made such experiments particularly challenging was the **very weak** intensity of the *static* XMLD signatures measured at the Fe K-edge in YIG given that σ_{XMLD} hardly exceeded 2.5x10⁻⁴ of the edge jump⁵.

2. RESULTS

In the two PSD spectra reproduced in Fig 3, one would expect the LP-XDMR signatures to show up again as AM side-bands of the X-ray macrobunch repetition frequency $F_0 = 355.043$ kHz. Unfortunately, the very low level (*ca.* –115.dB) of the detected sidebands makes the story rather inconclusive because, at high pumping power, we could never get rid of a weak modulation residual peaking at *ca.* –117 dB even when the X-rays were switched off.



Thus, XMLD does not appear to be a practical way to probe the resonant precession of local magnetization components, at least at the Fe K-edge of YIG thin films: the interference term $M_z \Delta M_z^{(0)}$ predicted by eqn (1) can contribute at best to a LP-XDMR signal lying *ca.* 30 dB below the level of the XDMR signal $\propto \Delta M_z^{(0)}$ measured in longitudinal detection geometry with CP X-rays under similar pumping conditions. This led us to conclude that the effective operator associated with XMLD at the Fe K-edge⁶, *i.e.* the *electric charge quadrupole* Q_{ZZ} component is very weakly affected by the precession of the local magnetization components.

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