ESRF	**Experiment title:** ***ADVANCES IN XDMR : AMPLITUDE, FREQUENCY & PHASE MODULATON OF MICROWAVES***	**Experiment number:** **HE-2710**
Beamline: **ID-12**	**Date of experiment:** from: 10-JUN-2008 to: 17-JUN-2008	**Date of report:** 18-FEB-2009
Shifts: **18**	**Local contact(s):** **A. ROGALEV and F. WILHELM**	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

José GOULON[1*], *Andrei* ROGALEV[1*], *Fabrice* WILHELM[1*] and *Gérard* GOUJON[1*]

[1] European Synchrotron Radiation Facility (ESRF), B.P. 220, F-38043 Grenoble Cedex

1. MOTIVATIONS OF THE PROJECT

In 2007-2008, the ID12 team has invested much time and effort in designing and in commissionning a new, fully modular XDMR spectrometer exploiting amplitude (AM), frequency (FM) and phase modulation (PM) of the incident microwaves. Indeed, we expected a significant gain in detection sensitivity but also a far better immunity against artefacts, especially at very high pumping power. Moreover, there are attractive opportunities with modulation techniques to get a *direct* access to *longitudinal* and *transverse* relaxation times: the longitudinal relaxation time T_1 would tell us how fast energy is locally transferred to the lattice, whereas the transverse relaxation times T_2 would describe how fast precession coherence could be lost due to magnon-magnon scattering processes. Extracting relaxation times from measurements of the precession angle (ϑ_0) is not a trivial task even though, according to the Bloch-Bloembergen eqn. of motion[1,2]:

$\Delta M_z/M_s = -1/2\ [\tan \vartheta_0]^2 \approx -1/4\ \ (\gamma h_{cp})^2\ T_1 T_2$ in *longitudinal* geometry;

$\sin \vartheta_0 \approx \gamma h_{cp} T_2$ in *transverse* geometry.

Difficulties are threefold: (i) we failed so far to establish a reliable calibration method of the amplitude of the circularly polarized microwave field h_{cp} inside the resonant cavity; (ii) one should combine XDMR measurements in both longitudinal and transverse geometries in order to disentangle T_1 and T_2 contributions with the handicap that those measurements can hardly be performed under the same experimental conditions; (iii) *absolute* determinations of the opening angle of precession in transverse geometry would rely on still questionable approximations regarding the electron bunch length in the ESRF storage ring. In this respect, modulation techniques appear much more promising and could potentially be exploited to measure spin-lattice relaxation times as short as 100 ps.

The underlying concept, however, requires us to analyze as *complex vectors* every modulation satellite measured in the frequency domain: in other terms, we need accurate measurements of both the phase and amplitude of the output signals. In this respect, it is mandatory to operate the Agilent vector spectrum analyzer (VSA) in the *time-average* data acquisation mode. For example, measurements carried out with either amplitude (AM) or frequency (FM) modulation in the longitudinal geometry would yield the following phase shifts at resonance:

$$\tan \Phi_{AM} = -\omega_{AM} T_1 \{(1 - \xi^2 T_1 T_2)[1+½ (T_2/T_1)] + ½ (\omega_{AM})^2 T_1 T_2 [1+(T_2/T_1)] + ...\} \quad (1)$$

$$\tan \Phi_{FM} = -2\omega_{FM} T_2 \{1 + ¼ (T_1/T_2) + ½ (\omega_{FM})^2 T_1 T_2 [1+(T_1/T_2)] + ½ \Delta^2 T_1 T_2 [1-(T_2/T_1)] - ½ \xi^2 T_1^2 ...\} \quad (2)$$

in which: $\omega_{AM} = 2\pi F_{AM}$ and $\omega_{FM} = 2\pi F_{FM}$ whereas $\xi^2 = ¼ (\gamma h_{cp})^2$ and $\Delta = 2\pi[F_{MW} - F_{MW0}]$. Here, F_{AM} and F_{FM} are modulation frequencies whereas F_{MW} and F_{MW0} refer to the actual microwave frequency and its value at resonance. Note that in eqn (1), the microwave power now appears only as a correction since $\xi^2 T_1 T_2 = ¼ (h_{cp}/h_{sat})^2$. One would show that modulation phase-shits measured in transverse geometry could yield only T_2.

2. RESULTS

The highest priority was thus given to the commissionning of the new spectrometer that can be operated with amplitude (AM), frequency (FM) or phase modulation (PM) of microwaves.

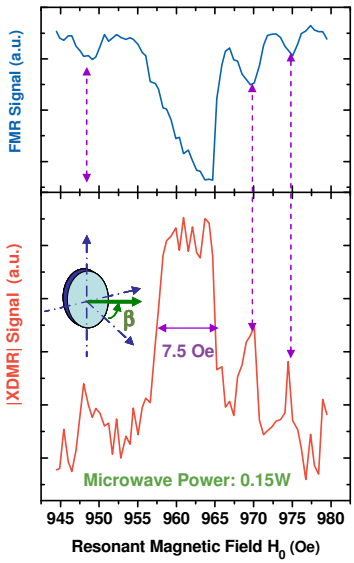
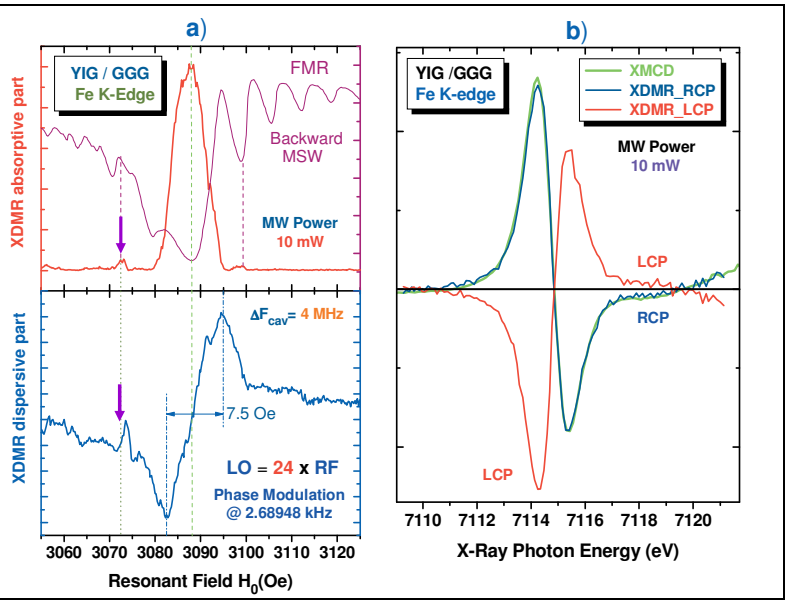


Figure 1: AM XDMR spectrum recorded in longitudinal geometry at the Fe K- edge with a YIG thin film inclined at magic angle. BMSW satellites become clearly visible at low pumping power.



Figures 2: PM XDMR spectra recorded in transverse geometry at the Fe K-edge with a YIG film inclined again at the magic angle: a) Absorptive and dispersive XDMR spectra recorded in the field-scan mode; b) Comparison of the static XMCD spectrum with XDMR spectra recorded in the energy –scan mode for left- or right CP X-rays.

We have reproduced in Figure 1 a typical Fe K-edge AM-XDMR spectrum recorded in *longitudinal* geometry with a YIG thin film rotated at magic angle in order to minimize any foldover effect[2]. With a pumping power as low as 150 mW, we could resolve -for the first time- the low order bulk (Backward) MagnetoStatic Waves (BMSW) satellites: this is the first direct evidence that *orbital* magnetization components can couple to magnetostatic modes through dipole-dipole interactions[2]. From very preliminary measurements of $\Delta\Phi_{AM}$, we could also estimate T_1 to be of the order of 80 ns.

We have reproduced in Figure 2 typical PM-XDMR spectra recorded in *transverse* geometry with a YIG film again rotated at magic angle. The absorptive and dispersive parts shown in Fig. 2a were measured in a superheterodyne detection mode with only 10 mW incident microwave power. The signal could be still measured[2] at a power as low as 100 µW. As shown in Fig. 2b, the sign of the XDMR spectra recorded in the energy-scan mode is nicely inverted when switching from left- to right-circularly polarized X-rays whereas there is no significant difference with repect to the static XMCD signal[2]. Unfortunately, for some reason which is still unclear, we failed so far to measure T_2 in the latter PM superheterodyne mode.

REFERENCES

[1]J. Goulon, A. Rogalev, F. Wilhelm, Ch. Goulon-Ginet and G. Goujon,
Element-selective X-ray detected magnetic resonance: a novel application of Synchrotron Radiation.
J. Synchrotron Radiation **14**, (2007), 257-271.

[2]J. Goulon, A. Rogalev, F. Wilhelm and G. Goujon,
X-ray detected magnetic resonance: a new spectroscopic tool.
Proc. of the V^th international school on Synchrotron Radiation and Magnetism, Mittelwihr (2008) – Springer Verlag