ESRF	**Experiment title:** ***X-RAY DETECTED NUCLEAR MAGNETIC RESONANCE IN MAGNETICALLY ORDERED RARE EARTH INTERMETALLICS***	**Experiment number:** **HE-3014**
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1. THE XD_NMR PROPOSAL

In magnetically ordered rare earth (RE) intermetallics (e.g.TbZn or $TbFe_2$), the NMR of RE nuclei entails the response of an *effective* transverse magnetization $\mathbf{M_1} = \mathbf{M_1} + \mathbf{m_1}$ to the external pump field $\mathbf{h_1}(t)$. This situation results from strong hyperfine interactions[1,2]. Our proposal started with the consideration that the electron magnetization component $\mathbf{M_1} \approx \eta\mathbf{m_1}$ should largely exceed the nuclear magnetization $\mathbf{m_1}$ because the enhancement factor: $\eta = H_{hfi} / [H_0+H_a]$ could be as large as 10^3 in magnetically ordered intermetallics such as TbZn or $TbFe_2$. This implies that in those ordered systems, the hyperfine interaction field H_{hfi} of ^{159}Tb nuclei should be much greater than both the external bias field H_0 and the effective anisotropy field H_a. This stimulated us to explore whether the ^{159}Tb-NMR signal could be probed on detecting the coherent precession of the magnetization $\mathbf{M_1}$. A subsidiary question was to know whether this new detection scheme could benefit of the amplification factor $[1+\eta]^2$ which makes conventional NMR measurements at low temperature on magnetically ordered materials nearly as sensitive as FMR.

We tried to detect XD_NMR signals of ^{159}Tb (I=3/2) nuclei in a (110) TbZn single crystal and in polycrystalline $TbFe_2$ thin films grown at the IPCMS (Strasbourg). The ^{159}Tb nuclei have 100% natural abundancy and benefit of a large enhancement factor η responsible for a huge shift of the effective NMR frequency from the RF range up to the microwave S-band. For quadrupolar nuclei such as ^{159}Tb, the zero-field NMR frequencies are given by: $\nu_{m,\,m-1} = |a_t + (2m-1)P_t|$ in which $(I-1) \le m \le I$, a_t and P_t being the magnetic dipole and electric quadrupole hyperfine parameters respectively[2,3]. The ^{159}Tb NMR spectrum in TbZn thus exhibits a triplet-line with $a_t \approx 3070$ MHz and $P_t \approx 362.5$ MHz. Even though the bias field H_0 has little or no influence on the NMR frequency[1,2], a magnetic order has nevertheless to be created in the sample along a well defined direction. Whereas nearly all published NMR data refer to ***spin-echo*** measurements[2] at very low temperatures ($T < 4.5$K), the only chance for us -at this stage- to retrieve a very weak XD_NMR signal from noise was to carry out such a challenging experiment in CW mode using a high sensitivity superheterodyne X-ray detection in transverse geometry[4,5]. Unfortunately, the temperature could never be decreased below 25K under a CW microwave pumping power that could be increased up to 5 W.

2. ADVANCED INSTRUMENTATION DEVELOPMENT

Given that "Zero- field" XD_NMR spectra could only be recorded on ***scanning the microwave frequency***, we had to design and to test microwave cavities which were fully ***tunable*** over a typical bandwith of 100 MHz while preserving a high Q-factor. Some adaptation of the superheterodyne detection scheme was also needed in order to maximize the sensitivity of the experiment. Typically, we concentrated much efforts on the detection of the ^{159}Tb-NMR central line[3]: $\nu_{1/2,-1/2} = 3070 \pm 1$ MHz of the TbZn crystal magnetized along its easy direction [110].

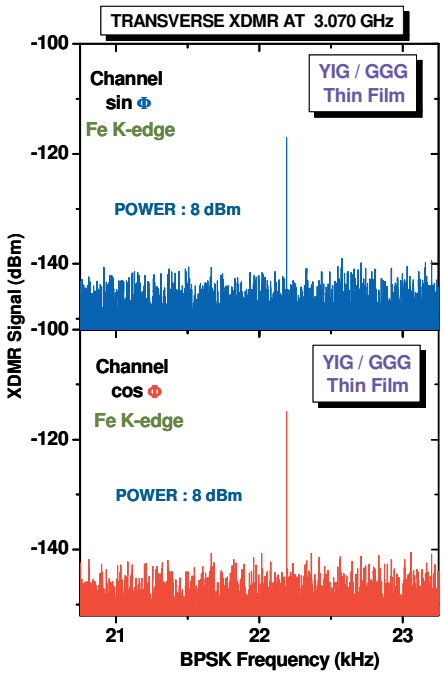

Fig. 1 Test of the superheterodyne detection using as reference the XDMR signal of a YIG thin film. The pumping power was 6.3 mW.

Our strategy was to detect a beating signal involving the amplitude modulation of the incident X-ray beam at the (local oscillator) frequency: LO = 279xRF/32 = 3070.7642 MHz. This required an unusual filling mode of the storage ring developed in close collaboration with the ESRF Accelerator & Source division. The 26/31 filling mode consists in 26 sequences of {24 filled + 8 empty bunches} followed by 5x32 empty bunches.

The detection sensitivity was further improved using a *superheterodyne* scheme in which we exploited a 180° bi-phase modulation (bpsk) of the microwave pump field at f_{bpsk} = RF/(992x16) so that: f_{bpsk} = 22.19016 kHz. The superheterodyne detection[4,5] consisted in catching the modulation satellites at frequencies IF $\pm f_{bpsk}$ with IF = 764.2 kHz. Additional electronics was developed to perform automatically a translation of the detector output in the frequency domain in order to remove IF.

As illustrated with Fig.1, we have checked the high sensitivity of the whole detection electronics by measuring the Fe K-edge XDMR signal of our reference YIG film at 3.070 GHz under a pumping power as low as 6.3 mW. Note that the exotic 26/31 filling mode of the storage ring inherently caused a signal loss of 12dB with respect to standard measurements in the S-band.

3. RESULT

Unfortunately, we failed so far to measure any signal that could be unambiguously assigned to a weak XD_NMR signature. One may envisage several explanations of this negative result: (i) since we failed to keep the sample temperature below 4K, the NMR relaxation time T_2 was probably too short; (ii) in CW XD_NMR experiments, there is no self-compensation for the *inhomogenous* line broadening whereas this is inherently the case in pulsed spin-echo measurements; (iii) since the penetration of the microwaves is restricted to a small skin-depth in intermetallics, this may have caused a dramatic loss of sensitivity owing to the much deeper penetration of X-rays. It is our diagnostic that this is the combination of these numerous handicaps, together with the 12 dB loss of signal in the exotic 26/31 filling mode, which finally concurred to a failure of this challenging XD_NMR project.

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