ESRF	**Experiment title:** ***X-RAY DETECTED NUCLEAR MAGNETIC RESONANCE IN MAGNETICALLY ORDERED RARE EARTH INTERMETALLICS***	**Experiment number:** **MI-901**
Beamline: **ID-12**	**Date of experiment:** from: 25-FEB-2009 to: 03-MAR-2009	**Date of report:** 05-FEB-2010
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1. EXPERIMENTAL REQUIREMENTS

The ultimate goal of this highly challenging project was to measure the XD_NMR signatures of ^{159}Tb (I=3/2) nuclei in a (110) TbZn single crystal or in polycrystalline $TbFe_2$ thin films grown at the IPCMS (Strasbourg). The ^{159}Tb nuclei have 100% natural abundancy and are known to benefit in magnetically ordered systems 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]. Thus, for the latter magnetically ordered intermetallics, one could envisage to detect the following triplet lines of the ^{159}Tb NMR spectra:

$TbFe_2$	$\nu_{+3/2,+1/2} = 4490 \pm 2$ MHz	TbZn	$\nu_{+3/2,+1/2} = 3795 \pm 2$ MHz
	$\nu_{+1/2,-1/2} = 3800 \pm 1$ MHz		$\nu_{+1/2,-1/2} = 3070 \pm 1$ MHz
	$\nu_{-1/2,-3/2} = 3110 \pm 2$ MHz		$\nu_{-1/2,-3/2} = 2345 \pm 2$ 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 < 4.5K), 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

Given that "Zero- field" XD_NMR spectra could only be recorded on ***scanning the microwave frequency***, we had to design and to test a series of microwave cavities which were ***tunable*** over a typical bandwith of 100 MHz while preserving a high Q-factor. Some adaptation of our heterodyne detection scheme was also needed in order to maximize the sensitivity of the experiment. The strategy was to let the weak NMR modulation signal at microwave frequency *beat* with a local oscillator (LO) signal resulting from the amplitude modulation of the incident X-ray beam due to the time-structure of the electron bunches in the storage ring. What made the present detection scheme innovative was the selection of LO frequencies that were harmonics of RF/4 or RF/32 rather than RF. This is only possible with an exotic (26/31) filling mode of the ESRF storage ring which had to be implemented and validated by the Accelerator & Source division.

We concentrated our efforts on two favourable NMR frequencies:

$\nu_{+3/2,+1/2}$ ($TbFe_2$) = 4490 ± 2 MHz LO = 51xRF/4 = 4490.5799 MHz IF=579.9 kHz
$\nu_{+1/2,-1/2}$ (TbZn) = 3070 ± 1 MHz LO = 279xRF/32 = 3070.7642 MHz IF=764.2 kHz

For the first experiment, the 26/31 filling mode consisted in 26x8 sequences of {3 filled + 1 empty bunches} followed by 5x32 empty bunches; in the second case, we had 26 sequences of {24 filled +8 empty bunches} followed again by 5x32 empty bunches. The amplitude modulation of the incident X-ray beam at either RF/4 or RF/32 is illustrated with Fig. 1a and 1b respectively.

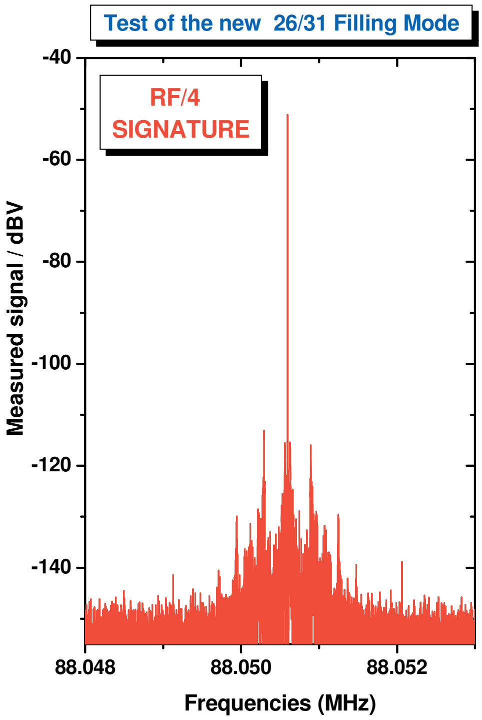

Fig. 1a : *Heterodyne* detection of the amplitude modulation of the X-ray beam at frequency RF/4. Dynamic range is *ca.* 96 dB.

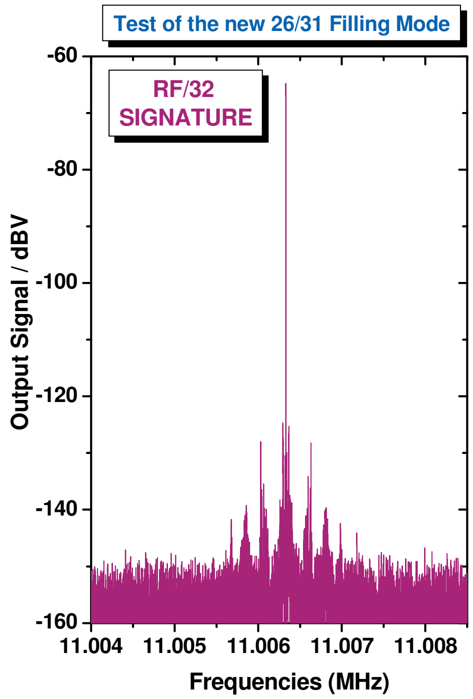

Fig. 1b : *Direct* detection of the amplitude modulation of the X-ray beam at frequency F_4 . Dynamic range is *ca.* 85 dB.

The detection sensitivity was tentatively 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) = 22.19016 kHz. The superheterodyne detection[4,5] consisted in catching the modulation satellites at frequencies IF ± f_{bpsk} whereas additional electronics was developed to perform automatically a translation of the detector output in the frequency domain in order to remove IF.

3. RESULT

Unfortunately, we failed so-far to measure any signal that could be unambiguously assigned to a weak XD_NMR signature. One may identify several causes to this failure: (i) a 12 dB loss of sensitivity due to the exotic 26/31 filling mode; (ii) a dramatic broadening of the resonance due a temperature of the sample excceding 4K; (iii) a dramatic loss of sensitivity may have been also caused by the deeper penetration of the X-rays inside the sample as compared to the penetration of the microwaves which is restricted to a very small skin-depth in intermetallics.

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