



	Experiment title: <i>INDUCED MAGNETISM IN BISMUTH SUBSTITUTED IRON GARNET THIN FILMS</i>	Experiment number: HE-2119
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1. MAGNETIC PROPERTIES OF Bi-SUBSTITUTED IRON GARNET THIN FILMS

High quality Bi substituted (RE-Y)IG films with RE = Lu, La...were grown at the *Laboratoire de Magnétisme de Bretagne* (Brest) using Liquid Phase Epitaxy (LPE) on carefully oriented GGG substrates¹. Bi-Lu-YIG films represent a class of materials of outstanding technological importance, not anymore for bubble memories but for THz optical modulation^{2,3} and monolithic microwave integrated circuits (MMIC) devices⁴. With a closed shell structure Bi³⁺ cations are expected to be diamagnetic, but they strongly affect the magnetic and magneto-optical properties of those films combining a large Faraday rotation ($\theta_F \approx 1.2^\circ/\mu\text{m}$), *in-plane* magnetic anisotropy and ultra-high frequency response. Bi-substituted (RE-Y)IG films were also found to exhibit a “*giant*” magnetoelectric (ME) rotation linear with the applied electric field⁵. Simple considerations regarding symmetry lowering⁶ in a film grown on a cubic m3m GGG substrate suggest that the true point group could then be 3m, 4mm or only m depending on whether the film is grown along the [111], [001] or [210] directions. We are not aware yet of any determination of the true magnetic groups of Bi-substituted iron garnet films on any particular GGG substrate.

XMCD did appear to us as an attractive tool to elucidate the long pending question of the existence of *induced* magnetic moments at the sites of a variety of important “*diamagnetic*” cations (Y³⁺, La³⁺, Lu³⁺, Bi³⁺). Moreover, the XMCD sum rules add to the inherent element/edge selectivity of X-ray absorption spectroscopy some valuable potentiality to discriminate between orbital and spin moments. It was therefore the main objective of proposal HE-2119 to carry out detailed XMCD studies on series of iron garnet films but we also envisaged to complement ultimately this work on collecting *X-ray Magneto-Electric Linear Dichroism* (XMELD) spectra on selected films. Unfortunately, no beamtime was left free for such delicate experiments.

2. X-RAY DICHROISM STUDIES OF SUBSTITUTED IRON GARNET FILMS

In this report, we shall focus on partial results collected on three films: **1** = unsubstituted YIG (Y₃Fe₅O₁₂) ; **2** = {Y,La,Lu}IG (Y_{1.3}La_{0.47}Lu_{1.3}Fe_{4.84}O₁₂) ; **3** = {Y,Bi}IG (Y_{1.69}Bi_{1.31}Fe_{4.9}Ga_{0.1}O₁₂). Both X-ray circular and linear magnetic dichroisms (*i.e.* XMCD & XMLD) spectra were measured at the Fe K-edge. High quality XMCD data were collected in the fluorescence excitation mode at the spin-orbit split Y, La, Lu L_{2,3}-edges. After proper

corrections for fluorescence re-absorption and for variable circular polarization rates, we could make use of the *differential* formulation of the magneto-optical sum rules to resolve the 4d- or 5d-projected **orbital** or **spin** DOS, *i.e.* the differential operators l_z and $2s_z$ whose spectra are reproduced in Figures 2 and 3 which produce clear evidence that the Y 4d states and the La, Lu 5d states are strongly spin-polarized even though the corresponding integral spin moments cancel out : $\langle s_z \rangle_d \leq 0.01\mu_B$.

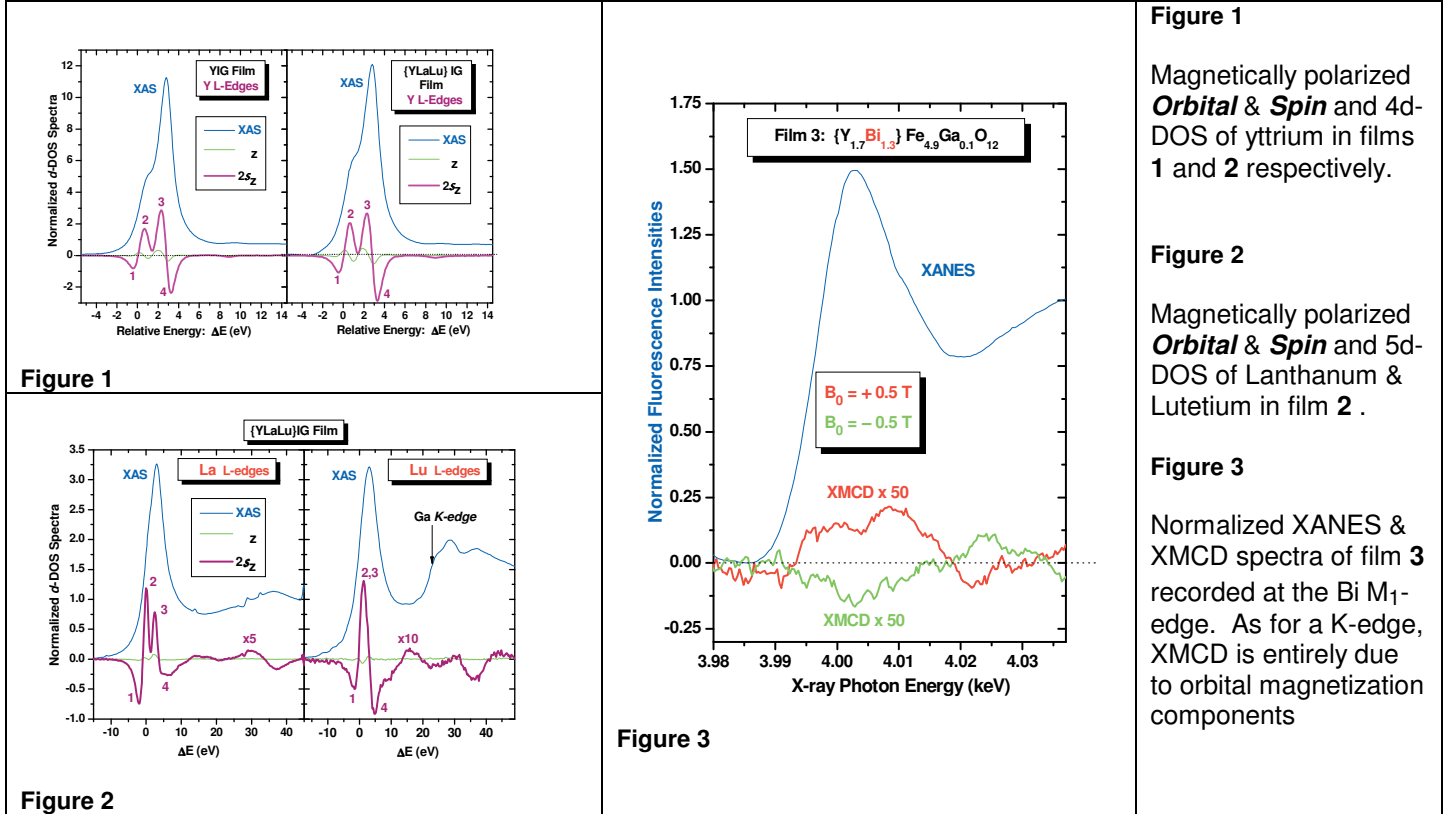


Figure 1

Magnetically polarized **Orbital & Spin** and 4d-DOS of yttrium in films 1 and 2 respectively.

Figure 2

Magnetically polarized **Orbital & Spin** and 5d-DOS of Lanthanum & Lutetium in film 2.

Figure 3

Normalized XANES & XMCD spectra of film 3 recorded at the Bi M₁-edge. As for a K-edge, XMCD is entirely due to orbital magnetization components

The experiments were much more challenging at the Bi M₁-edge but nearly hopeless at the Bi M_{4,5}-edges where the circular polarization rates of the monochromatic X-ray beam are vanishingly small. Nevertheless, the unambiguous detection of a weak XMCD signal at the Bi M₁-edge established for the first time the existence of a weak **orbital** magnetic component involving the 6p states of Bi. Recall that a careful NMR study⁷ concluded that the internal field at the ²⁰⁹Bi nuclei was too high to be explained only by the dipole field of the ferric ions and suggested that *localized* magnetic moments *might* exist at the Bi sites. Difference spectra also revealed subtle changes¹ in the Fe K-edge XMCD spectra of films 1 and 3.

Interestingly, weak signal of *natural* X-ray linear dichroism were systematically observed at the Fe K-edge with all iron garnet films as well as with a bulk YIG crystal cut parallel to the (111) planes: this proved that, at room temperature, the crystal cannot satisfy all requirements of perfect cubic symmetry, crystal distortions preserving at best a trigonal symmetry (R₃ or R_{3m}). A very weak -but perfectly reproducible- XMLD signal¹ was also measured in the Fe K-edge prepeak of the XANES spectrum of YIG: this signal revealed the presence of a tiny electric quadrupole moment in the ground state charge distribution of the iron atoms in YIG.

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