

## New Method of performing atomic-selective hysteresis cycles avoiding surface effects in nanomaterials by using XMCD in the hard x-ray region.

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### Introduction:

The great interest into new magnetic nanomaterials research needs of new experimental probes for characterizing these materials at the microscopic level. Within this framework XMCD emerges as an outstanding tool to study magnetism as each element can be probed separately by choosing the absorption edge, which opens the possibility of performing element-selective hysteresis cycles. Typically, the application of XMCD to study hysteresis cycles are obtained at the soft x-ray energy range,  $L_{2,3}$ -edges of transition metals and  $M_{4,5}$  edges of rare-earth metals. However, these materials are extremely sensitive to the surface effects at these absorption edges and the magnetic response of the bulk material to the XMCD can be hindered by the surface. As a consequence, the magnetic properties of the different components of the nanomaterial cannot be obtained at the same experimental conditions.

We have investigated the possibility of performing element selective magnetometry (ESM) by using a SINGLE absorption edge in the hard x-ray range. The method is based on the peculiar nature of the XMCD when it probes the conduction band magnetism. Our previous works in the field demonstrate that the atomic selectivity is not lost when XMCD probes the delocalised states. On the contrary, it provides a direct way of disentangling the magnetic contributions to the conduction band coming from the different elements in the material [1-3]. Then, we have tailored the present experiment to verify the possibility of performing ESM by using a single x-ray absorption edge. To this aim we have studied the modification of the XMCD spectrum recorded at the Fe K-edge as a function of the applied magnetic field in the case of both  $Y_6Fe_{23}$  and  $Er_6Fe_{23}$ .

### Experiment:

The  $R_6Fe_{23}$  samples (with  $R=Y$  and  $Er$ ) were prepared following standard procedures [4]. X-ray diffraction analysis showed that all the samples are single phase. The macroscopic magnetic measurements were performed following standard methods in magnetic fields up to 5 T, by using a commercial SQUID magnetometer (Quantum

Design MPMS-5S). XMCD experiments were performed at the beamline BL39XU of the Spring-8 Facility [5]. XMCD spectra were recorded on homogeneous layers of the powdered samples in the transmission mode at the Fe K-edge by using the helicity-modulation technique [6]. In all the cases, the XAS spectra were normalized to the averaged absorption coefficient at high energy.

### Result and Discussion:

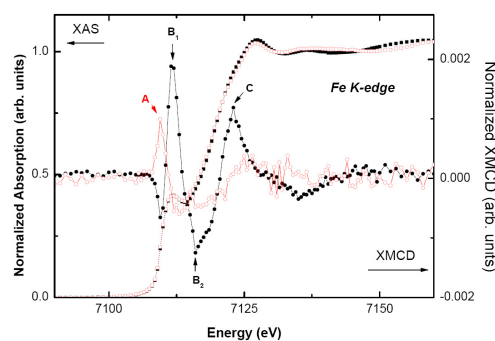


Fig.1 Fe K-edge XMCD spectra of  $Y_6Fe_{23}$  (○) and  $Er_6Fe_{23}$  (●) and at 5 K and  $H = 5T$ .

The comparison of the Fe K-edge XMCD spectra recorded at room temperature in both  $Y_6Fe_{23}$  and  $Er_6Fe_{23}$  is shown in Fig.1. The XMCD spectrum of  $Y_6Fe_{23}$  shows a narrow positive peak at the absorption threshold (A) followed by a negative dip at higher energies (C). Despite the magnetic properties of the Fe sublattice in both  $Y_6Fe_{23}$  and  $Er_6Fe_{23}$  compounds is thought to be similar, their Fe K-edge XMCD spectra present noticeable differences. Indeed, as shown in Fig.1, the Fe K-edge XMCD spectrum of  $Er_6Fe_{23}$  also exhibits the narrow peak at the edge (A). However, the former negative dip in  $Y_6Fe_{23}$  splits into two intense peaks ( $B_1$  and  $B_2$ ) of opposite sign. This strong modification of the Fe K-edge XMCD spectral shape cannot be easily assigned to differences of the Fe sublattice magnetic properties between  $Y_6Fe_{23}$  and  $Er_6Fe_{23}$  and, on the contrary, it points out the contribution of the Er magnetic sublattice to the Fe K-edge XMCD spectrum.

Upon our hypothesis, peaks  $B_1$ ,  $B_2$  and C of the  $Er_6Fe_{23}$  spectrum reflects the Er contribution to the Fe K-edge, while both Fe and Er contribute to the A

peak. In order to verify the validity of our hypothesis we have performed hysteresis cycles by recording the XMCD as a function of the applied magnetic field for fixed energy points: A (7109.5 eV), B1 (7111.5 eV), B2 (7116 eV) and C (7123 eV).

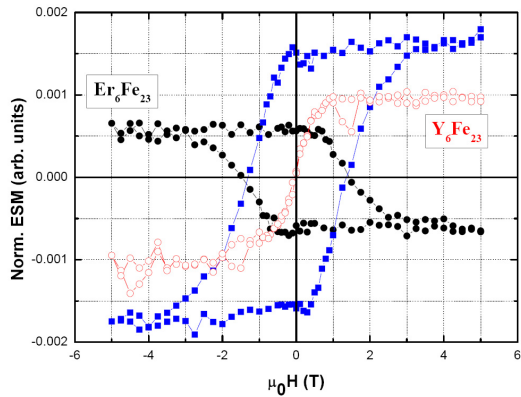


Fig.2 ESM curves recorded at  $T = 5$  K at the A peak of the Fe K-edge XMCD spectra of  $Y_6Fe_{23}$  (○) and  $Er_6Fe_{23}$  (●) and at the B1 peak (●) of  $Er_6Fe_{23}$  (see text for details).

The results, reported in Fig. 2, clearly shows the different magnetic behaviour of the contributions to the XMCD spectrum even when Fe K-edge is tuned. In the case of  $Y_6Fe_{23}$  the ESM curve yields a coercive field  $H_C = 172$  Oe, in agreement with the macroscopic data. By contrast, the  $H_C$  obtained in the ESM curves recorded in  $Er_6Fe_{23}$  is two orders of magnitude larger ( $\sim 14000$  Oe). It should be noted that, as shown in Fig. 3, the coercive field measured macroscopically for  $Er_6Fe_{23}$  is  $\sim 2000$  Oe.

These results confirm our hypothesis, i.e. that both Fe and Er contribute to the Fe K-edge XMCD signal. Moreover, they demonstrate that despite we are tuning the Fe K-edge, the hysteresis cycles at different energy points are completely different in  $Y_6Fe_{23}$  and  $Er_6Fe_{23}$  and they can be assigned to the properties of both Fe and Er sublattices separately. As a final demonstration we have compared in Fig. 3 the macroscopic hysteresis cycle of  $Er_6Fe_{23}$  and that obtained as the weighted addition of those derived from the XMCD. As shown in the figure the coercive field obtained from the XMCD contribution is  $\sim 100$  Oe, in good agreement with the macroscopic data.

### Conclusion:

These results suggest that when probing the conduction band, XMCD is a simultaneous fingerprint of the magnetism of both components of the material, the rare-earth and the transition-metal, even when only an atomic element is tuned. By using the method above we have provided the disentangled contribution of both Fe and Er sublattices to the whole coercivity of the systems by tuning a SINGLE absorption edge.

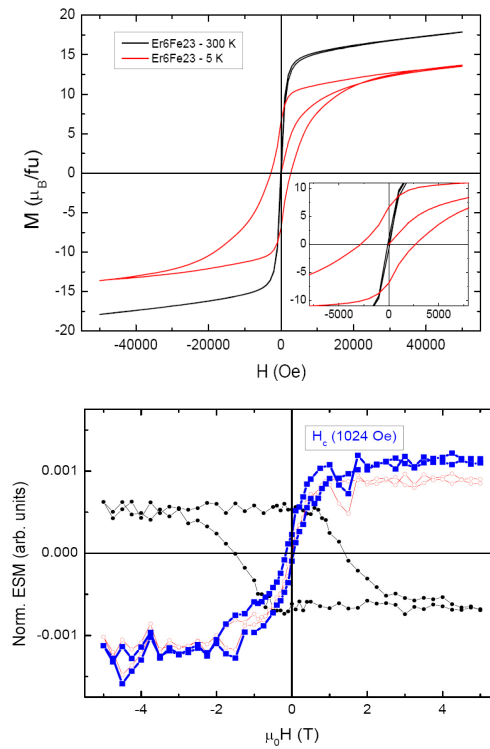


Fig.3 Top: Comparison of the macroscopic magnetic hysteresis cycle of  $Er_6Fe_{23}$  measured at different temperatures. Bottom: Comparison of the ESM curves obtained from the Fe K-edge XMCD of  $Y_6Fe_{23}$  (○) and  $Er_6Fe_{23}$  (●) at  $T = 5$  K at the A peak of the Fe K-edge XMCD spectra of  $Y_6Fe_{23}$  (○) and  $Er_6Fe_{23}$  (●) and their weighted sum (●).

These results suggest that it is possible to perform element-specific magnetometry by using a SINGLE absorption edge in the hard x-ray domain, thus avoiding all the inherent difficulties of using soft x-rays to obtain the magnetic characterization.

The fact that the energy of the x-rays is raised from the soft to the hard x-ray region has the main advantage of avoiding surface effects inherent to the size reduction. In addition, this method guaranties that the sampling depth is enhanced and then the XMCD becomes a real BULK probe. Moreover, using the hard x-ray region to obtain the magnetic characterization of the material enhances the flexibility of the sample environment. In this way, extreme conditions of temperature, applied magnetic field and even applied pressure can be studied.

### References

- [1] J. Chaboy et al., Phys. Rev. **B69**, 134421 (2004)
- [2] M.A. Laguna-Marco et al., Phys. Rev. **B72**, 094408 (2005)
- [3] J. Chaboy et al., J. Synchrotron Rad. **15**, 440 (2008).
- [4] J. F. Herbst and J. J. Croat, J. Appl. Phys. **55**, 3023 (1984).
- [5] H. Maruyama, J. Synchrotron Rad. **8**, 125 (2001)
- [6] M. Suzuki et al., Jpn. J. Appl. Phys. **37**, L1488 (1998)