

# Temperature dependence of the XMCD signal at the RE $L_3$ edge in REFe<sub>2</sub>

J.C. Lang, G. Srajer, C.S. Nelson, and C.T. Venkataraman  
Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439 USA

A.I. Goldman, C. Detlefs, Z. Islam, B.N. Harmon, K.W. Dennis, and R.W. McCallum  
Ames Laboratory, Iowa State University, Ames, IA 50010 USA

## Introduction

Compounds containing rare earths (RE) and 3d transition metals (TM) are of great technological importance due to their wide range of industrial applications, such as magneto-optical disks, transducers, and (most importantly) high-performance permanent magnets. In these materials the RE atom is used to increase the magnetic anisotropy while the transition metal enhances the curie temperature. The magnetic moment on the RE atom arises primarily from the highly localized 4f orbital, which has negligible overlap with the surrounding atoms, while the moment on the TM arises on the much more delocalized 3d orbital. Thus, magnetic ordering in these compounds is mediated through an indirect exchange, whereby the RE 4f orbital spin polarizes the more diffuse RE 5d orbital, which in turn has a direct exchange coupling with the TM 3d orbital. Clearly for a well-defined understanding of the magnetic properties of these materials, knowledge of the spin-polarization and exchange-coupling mechanisms involved in the 5d orbital is essential.

X-ray magnetic circular dichroism (XMCD) provides an ideal tool for providing just such information. XMCD is defined as the difference in the absorption coefficient between right- and left-handed circularly polarized x-rays. Since wave functions of the initial core levels are well understood, the structure in the measured spectra can be directly correlated to the spin polarization of the final state orbital allowed by selection rules. Furthermore, since the core level binding energies are unique to each of the constituent atoms in a material, the information obtained is specific to a particular element and orbital.

In this report, we present XMCD measurements at the RE  $L_3$  edge on a series of REFe<sub>2</sub> compounds (RE = Ho, Er, Tm, Lu). The mechanism behind the magnetic ordering in these materials is the well known RKKY interaction illustrated in Figure 1. The well-localized 4f orbital spin polarizes the 5d orbital via an exchange  $\xi_{4f-5d}$ , which in turn interacts with the TM 3d orbital through  $\xi_{5d-3d}$  exchange.

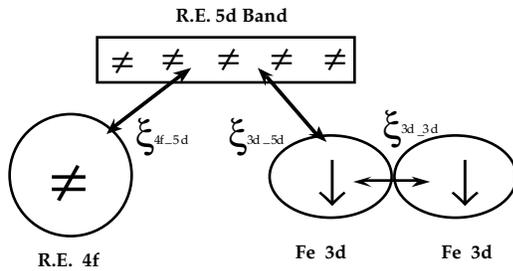


Figure 1: Schematic of magnetization process.

Additionally, the TM atoms also have a strong coupling between themselves through a direct overlap of their respective 3d orbitals ( $\xi_{3d-3d}$ ). Since the direct exchange between the TM atoms ( $\xi_{TM-TM} \sim \xi_{3d-3d}$ ) is typically much stronger than the indirect exchange between the RE and TM atoms ( $\xi_{RE-TM} \sim \xi_{4f-5d} * \xi_{5d-3d}$ ), the magnetization of the RE sublattice will fall off faster than that of the TM. Previous XMCD measurements at these edges have shown that they are composed of two distinct features. One is associated with the 5d orbital, while another has been shown to arise from transitions to the 4f orbital [1]. The effect of the different exchange mechanisms on the spin polarization of the individual orbitals can be investigated by varying the RE and changing the temperature of the material. By changing the RE, the strength of the  $\xi_{4f-5d}$  exchange is varied while keeping the  $\xi_{5d-3d}$  exchange constant. In the case of Lu, this exchange goes to zero since it has no 4f moment, thereby isolating effects of the  $\xi_{5d-3d}$  interaction alone. In the temperature behavior, features that are more tightly coupled to the TM lattice should show less temperature variation.

## Methods and Materials

The measurements were made on the Advanced Photon Source 1-ID beamline. The beam was monochromatized using a Si(111) monochromator yielding an energy resolution of  $\sim 1.5$  eV for the edges investigated. A diamond (111) phase retarder was used to convert the on-axis linearly polarized synchrotron beam from the insertion device into a circularly polarized one,  $P_c=0.98$ , for the experiment. Powder samples were made of each material and placed on tapes for transmission measurements. The samples were placed inside a closed-loop He displx in order to vary the temperature between 10 K and 300 K. In order to magnetize the sample, a 4.0 kG permanent magnet was also placed inside the displx. The magnetic field was oriented at  $45^\circ$  with respect to the incident beam in order to allow the transmitted beam onto the detector. The entire displx with the magnet and sample inside could be rotated by  $180^\circ$  in order to take data with the field both parallel and antiparallel to the incident beam helicity. This eliminated any artifacts in the signal that are not due solely to the sample magnetization. Spectra were taken by reversing the helicity at each energy, with several spectra taken at each magnet orientation and combined to improve the statistical accuracy.

## Results and Discussion

Figure 2 shows the XMCD spectra for each RE compound at 10 K and 300 K. The spectra of Ho, Er, and Tm all show prominent features at approximately  $-8$  and  $+4$  eV, which are known to come from the 4f and 5d bands, respectively. Lu, on the other hand, shows a single peak at  $\sim 0$  eV. A peak

at this energy is also clearly seen in the Tm spectra at 300 K (arrow). Note that the Tm spectra inverts with increasing temperature since at room temperature it is above its compensation point. Taking the derivative of the XMCD spectra reveals that this feature is also present in both the Er and Ho spectra but is simply obscured by the much larger pre- and post-edge features. Clearly since this feature is present in Lu, it must be caused by the  $\xi_{5d-3d}$  interaction. To deconvolute this small feature from the much larger nearby peaks, all spectra were fit using three Gaussian curves. This is shown in Figure 3, which shows data taken at the Er edge along with the three curves used to fit the spectra. The three peaks in the spectra are labeled Q, P, and D. Peak Q is known to be associated with the RE 4f states, while peak D is known to be due to the 5d orbital. The exact nature of peak P is unclear, but as mentioned earlier (and as shown by its temperature dependence), its most likely source is the exchange between the TM 3d and RE 5d states, since it has nearly the same magnitude and occurs at nearly the same energy as the peak in the Lu spectra.

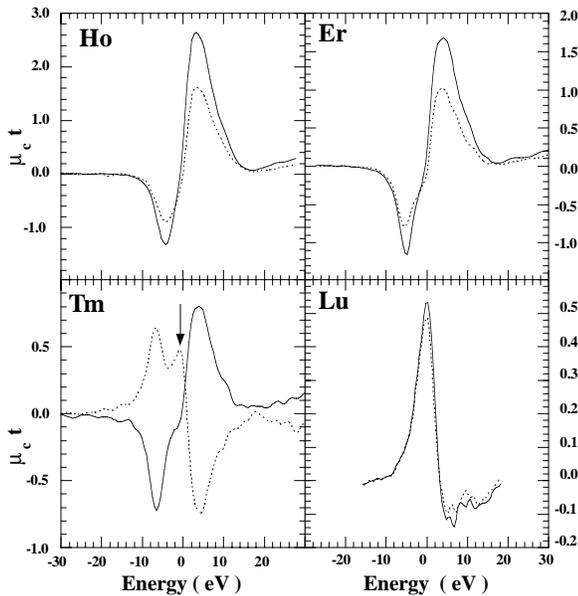


Figure 2: Dichroism signal in REFe<sub>2</sub> at 10 K (solid) and 300 K (dotted).

The changes in each of the features in the XMCD spectra as a function of temperature are plotted in Figure 4. Also plotted in Figure 4 are single-crystal neutron diffraction measurements [2] of the RE sublattice magnetization for Ho, Er, and Tm. The dotted line on the Lu data is meant merely as a guide to the eye, since no neutron measurements for this material were available. There are several things to notice about the temperature variations in the signal. First the 4f and 5d features have approximately the same temperature dependence. This would indicate that the 5d band polarization probed by feature D scales nearly identically with the 4f magnetization, and therefore is dominated by the  $\xi_{4f-5d}$  exchange, which is consistent with recent theoretical results [3]. Feature P, on the other hand, falls off much less with increasing temperature in Ho,

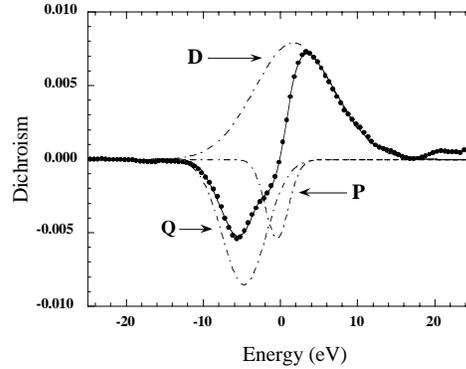


Figure 3: Deconvolution of the dichroism spectra into three Gaussian curves.

Er, and Tm, than the D and Q features. This would be consistent with this feature being strongly associated with the  $\xi_{5d-3d}$  exchange, since it scales with the expected magnetization of the TM sublattice. Lastly, while the D and Q features follow the general trend of the neutron measurements, there are substantial deviations for the Er and Tm spectra. At low temperatures the sample becomes much harder to magnetize due to increasing anisotropy, thus the XMCD data tends to deviate away from the neutron data, which was taken on single-crystal samples. Additionally the Tm data might be affected by the proximity of some of the measurements to the compensation temperature ( $T = 250$  K where the net magnetization is zero).

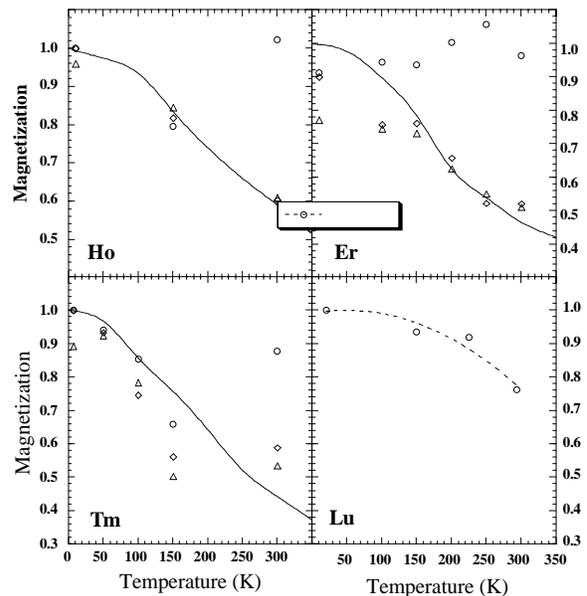


Figure 4: Temperature dependence of each feature in the dichroism spectra [Q (triangle), D (diamond), and P (circles)].

## Acknowledgments

Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

## References

- [1] J.C. Lang, G. Srajer, C. Detlefs, A.I. Goldman, H. Konig, X. Wang, B.N. Harmon, R.W. McCallum, *Phys. Rev. Lett.*, **74** 4935 (1995).
- [2] A.E. Clark, in *Handbook of the Physics and Chemistry of Rare Earths*, K.A. Gschneidner and L. Eyring eds., (North-Holland, 1979).
- [3] M. van Veenendaal, J.B. Goedkoop, and B.T. Thole, *Phys. Rev. Lett.*, **78** 1162 (1997).