Properties of thin film europium oxide by x-ray magnetic circular dichroism

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Magnetic circular dichroism was measured as function of temperature for the Eu M_5 and M_4 absorption edges in single crystal EuO thin films. The M_5 edge is resolved into two major features, one with the same helicity dependence as the M_4 edge, and one with opposite helicity dependence. While the ratio of the absorption peaks with similar helicity dependence remains constant as a function of temperature, the ratio of those with opposite dependence is found to change linearly with temperature below 75 K. The physical mechanism for this linear behavior in the absorption peak intensity is not well understood. *© 2004 American Institute of Physics.*
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I. INTRODUCTION

Europium oxide is optically transparent, ferromagnetic insulator, making it an attractive material for magneto-optical applications. Additionally, doping with rare earths or an excess of Eu leads to a field dependent insulator–metal transition that gives a colossal magnetoresistance (CMR) with resistivity changes of several orders of magnitude.[1] Stable EuO may be grown epitaxially on Si in order to inject spin-polarized charge carriers into the semiconductor, giving it strong potential in spintronics applications,[2] and has a large Faraday rotation, and may be useful in a variety of integrated magneto-optic and electro-optical systems, especially in applications already requiring cooling to liquid nitrogen temperatures. Characterization of the properties of thin films of EuO is necessary before devices can be designed to take advantage of its magnetic, optical, and electrical properties.

X-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD)[3–8] provide element-specific information about the chemical, electronic, and magnetic properties of the material under investigation, making them excellent techniques for characterizing these films. X-ray emission spectroscopy of the Eu $2p_{3/2}4d$ for bulk EuO shows strong helicity dependence.[9]

II. EXPERIMENT

Epitaxial EuO films were deposited in ultrahigh vacuum ($<10^{-9}$ Torr) on sapphire and cubic zirconia substrates. The ferromagnetic, insulating films were grown by thermal evaporation onto substrates held at 300 °C, resulting in nominally flat, 5 nm single crystal films and capped with a protective V film.[4] Growth was monitored with reflection high-energy electron diffraction, which indicated two-dimensional films with single crystal structure. X-ray diffraction showed no Eu oxides other that EuO to be present in the film. Su-

superconducting quantum interference device (SQUID) magnetometry measurements show that the EuO becomes magnetic at 70 K, the same Curie temperature (T_C) as bulk EuO.

The XAS and XMCD spectra were measured at the MSU Materials X-ray Characterization Facility located at beamline U4B at the National Synchrotron Light Source. The monochromator spans an energy range of 10–1350 eV and, by accepting radiation from above (or below) the plane of the orbit of the synchrotron ring, is capable of supplying circularly polarized light of variable polarization (in this case, 75%). The monochromator was set to an energy resolution of 0.15 eV at the Mn $L_{2,3}$ absorption edge. The details of the U4B beamline have been published elsewhere.[10]

For each XAS and XMCD spectrum two fluorescence yield spectra were recorded, one with the incident light helicity (circular polarization) oriented parallel (I^+) to the remnant magnetization and one antiparallel (I^-). The XAS is the average of the two spectra while the XMCD is the difference between the two. The remnant magnetization was switched by reversing a magnetic field that was applied in the plane of the sample while the incident light helicity was held constant. The XMCD spectra have been altered to compensate for the incomplete photon polarization (75%) and the 45° sample-plane orientation with respect to the incoming beam. The spectra were collected at pressures less than 5×10^{-10} Torr and were normalized to the incident flux via the current measured through a gold mesh that was placed in the beam in front of the sample. Helicity dependent spectra were collected for temperatures from 10 to 275 K as measured by a thermocouple mounted on the sample holder. To correct for monochrometer drift, the energies of the $M4$ and $M5$ peaks were set to previously published values.[11]

III. RESULTS

Representative XMCD and helicity-dependent XAS spectra for a film held at 40 K are shown in the top panel of Fig. 1. The M_5 peak can be resolved into two peaks: a shoul-

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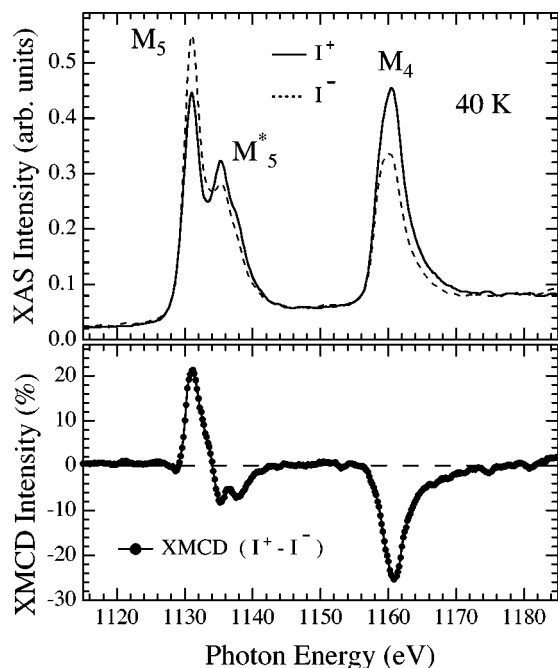[a]Author to whom correspondence should be addressed; electronic mail: holroyd@physics.montana.edu

FIG. 1. (Top) XAS for positive (I^+) and negative (I^-) helicity x rays measured at 40 K. (Bottom) XMCD determined from these spectra.

FIG. 3. Peak intensity ratios (left axis) for the M_5/M_4 and M_5^*/M_4 XAS intensities and the M_5^-/M_5^+ XMCD intensity (right axis) as a function of temperature.

der labeled M_5^* at higher binding energy with the same helicity dependence as the M_4 peak and the main peak labeled M_5 with opposite dependence.

The XMCD spectra, the difference between these two helicity-dependent spectra after normalization for the incomplete polarization and the incidence angle (shown in the bottom panel of Fig. 1), have a large dichroism effect of over 30% at low temperatures. In addition a third peak becomes resolved with the same helicity dependence as the M_4 peak.

The strong temperature dependence of these features in the Eu M_5 and M_4 XAS spectra are shown in Fig. 2. These spectra have been normalized to the M_5 peaks for comparative purposes. Note that this normalization does not conserve spectral intensity (the integral of the $M_{4,5}$ peaks). It can be seen from this figure that while the M_5^* and M_4 features scale proportionally, their ratio remaining constant as a func-
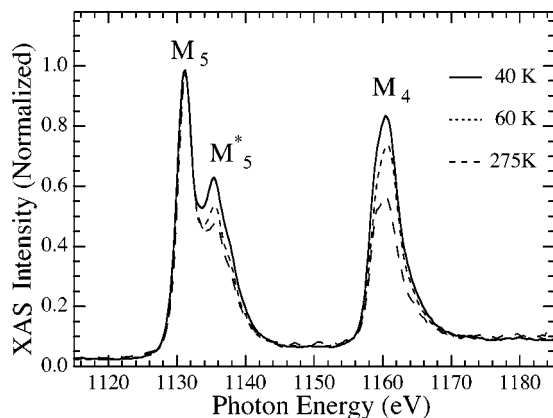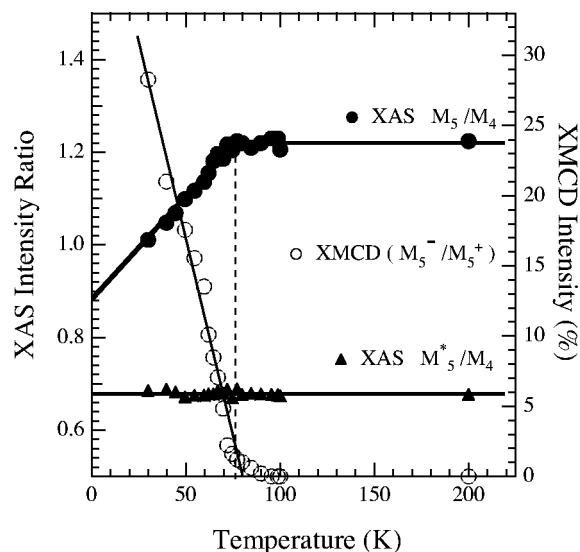
tion of temperature, the M_5 edge becomes relatively more intense with increasing temperature.

This behavior is detailed in Fig. 3, which shows the ratios M_5/M_4 and M_5^*/M_4 and the XMCD intensity as a function of substrate temperature. Above 75 K the dichroism vanishes suggesting that the material is above the material Curie temperature T_C. In addition, the M_5/M_4 and M_5^*/M_4 XAS ratios also remain constant above this temperature and do so up to room temperature (not shown in Fig. 3).

Below 75 K, the behavior of the dichroism and the XAS peak intensities are strikingly similar. The M_5 dichroism increases linearly with decreasing temperature. Similarly, as the temperature decreases, the M_5/M_4 ratio also decreases linearly with temperature. The break temperature in the XAS spectra is similar to the extracted Curie temperature for this film (the break temperature in the XMCD) as well as the measured T_C from SQUID magnetometry.

IV. CONCLUSION

The ferromagnetic insulator EuO is rich in potentially useful optical, magnetic, and electronic properties including large Faraday rotation, ferromagnetism, and a CMR transition. These properties could be utilized in thin films of EuO for integrated magneto-optic, electro-optic, and spintronics devices. The magnetic and chemical properties of thin film EuO have been examined by XMCD, and it has been found that dichroism increases linearly as temperature is decreased below 75 K. The ratio M_5^*/M_4 of the intensities of two absorption peaks with the same helicity dependence is constant, independent of temperature, and the ratio M_5/M_4 of two absorption peaks with opposite helicity dependence is found to increase linearly as temperature is reduced. The physical mechanism behind the linear behavior of these ratios is not well understood.

FIG. 2. Total XAS intensities (the average of the two helicity dependent spectra) for several temperatures.

ACKNOWLEDGMENTS

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