

Temperature and field-induced magnetization flips in amorphous Er–Fe alloys evidenced by x-ray magnetic circular dichroism

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Magnetic properties of amorphous $\text{Er}_{1-x}\text{Fe}_x$ alloys with $x \approx 0.7$ have been studied. Macroscopic characterization has been performed by measuring temperature- and field-dependent magnetization. Applying a magnetic field the compensation temperature first decreases, but increases again at larger fields. This “exotic” behavior has been interpreted in terms of the sperimagnetic character of both subnetworks. The suggested scheme has been checked by measuring x-ray circular magnetic dichroism at the Er M_5 -edge. Using this atom-sensitive technique we have been able to detect temperature-induced as well as field-induced flips of the Er-subnetwork with respect to the direction of the applied field. © 1996 American Institute of Physics. [S0021-8979(96)78608-1]

I. INTRODUCTION

Amorphous alloys have received much attention in the last years due to their interesting properties and their applicability for new devices.^{1,2} Their magnetic properties are strongly affected by bond and chemical disorder causing a distribution of magnetic moments and exchange interactions. Also, the randomly varying electrostatic fields create, via spin–orbit coupling, locally varying single-site anisotropies giving rise to noncollinear arrangements of the magnetic moments. These sperimagnetic structures have been widely observed in amorphous rare-earth-transition-metals alloys $R_{1-x}T_x$ ($0 < x < 1$).¹ The large spin–orbit coupling of non- S -state rare earths gives rise to large local anisotropies and therefore to a fan structure in the R subnetwork moment. In the case of $T=\text{Fe}$ noncollinear structures produced by the R – T exchange have been observed also in the Fe subnetwork.¹

In the case of heavy R , where R and T moments are coupled antiparallel,³ the different temperature dependence of each subnetwork moment can lead to a vanishing global magnetization at a temperature T_{comp} called “compensation temperature.” The fan magnetic structure in amorphous sperimagnets is responsible for a strong dependence of the magnetization of each subnetwork on the applied magnetic field. T_{comp} are therefore strongly dependent on the applied magnetic field.

The objective of the present paper is to characterize the magnetic behavior of an amorphous $\text{Er}_{1-n}\text{Fe}_n$ ($n \approx 0.70$) compound from a macroscopic point of view and to study independently the magnetic behavior of the Er subnetwork taking advantage of the element selectivity of x-ray absorption spectroscopy. X-ray magnetic circular dichroism (XMCD) measurements were performed at the Er M -edge as a function of temperature and applied magnetic field.

II. MACROSCOPIC STUDY

Crystalline alloys of $\text{Er}_{1-x}\text{Fe}_x$ with nominal composition $x=0.60$ were prepared by melting the starting elements in a cold crucible furnace. These alloys were used as targets in a sputtering chamber, using ionized Ar gas as sputter gas. Amorphous films were simultaneously grown on monocrystalline-Si and kapton foils, cooled at 77 K. A Si-deposited sample was used in the XMCD measurements to optimize the thermal contact with the cryostat head, while the kapton-deposited samples were used for macroscopic measurements in order to reduce the diamagnetic contribution of the substrate. The magnetic properties of the samples were shown to be independent of the nature of the substrate. Amorphization was checked by x-ray diffraction. Rutherford backscattering (RBS) with α particles performed on the amorphous films gave us a composition of $x=0.73 \pm 0.01$ and allowed us to determine the sample thickness. The work presented in this paper is focused on two samples: sample “a,” 400 Å thick, and sample “b,” 80 Å thick. All the samples were protected against oxidation by Cr 50 Å thick films.

Alternative magnetic susceptibility (χ) was measured in sample “a” with a superconducting quantum interference device from 4.2 to 300 K. The ordering temperature detected by measuring the thermal dependence of susceptibility, $\chi(T)$, is $T_c=190$ K. The macroscopic magnetic behavior of this compound was characterized by isofield- $M(T)$ and isotherms- $M(H)$ measurements from 4.2 K $< T < 300$ K and $0 < H < 6T$ performed in a vibrating sample magnetometer. The Arrott-plots, $M^2(H/M)$, obtained from $M(H)$ measurements show a change of the slope around T_c , which is typical for random anisotropy systems.⁴ $M(H)$ measurements show, up to $6T$, the characteristic lack of saturation associated with noncollinear magnetic structures. We can therefore assume that our compounds present sperimagnetic structures.

$M(T)$ measurements were performed on the sample “a” for different values of applied magnetic field. At low fields

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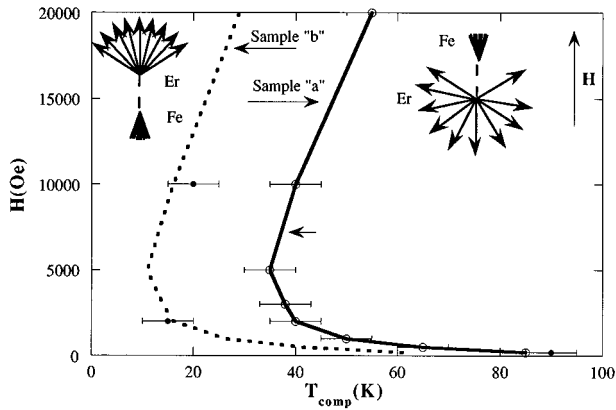


FIG. 1. Configuration phase diagram with respect to the applied field for two samples of $\text{Er}_{1-x}\text{Fe}_x$ ($x=0.73$): sample "a" (400 Å thick) and sample "b" (80 Å thick).

(around $0.05T$) $M(T)$ shows a compensation temperature at $T_{\text{comp}}=65$ K. By increasing H , the minimum of the magnetization shifts to lower temperatures and to nonzero magnetization values ($T_{\text{comp}}=35$ K at $H=0.5T$). For larger fields this tendency is reversed and the minimum is shifted to higher temperatures. In Fig. 1 we present the evolution of T_{comp} with the applied field.

For each magnetic field, we can identify the minimum in magnetization as a *pseudo*-compensation temperature, although no vanishing of magnetization occurs. As in the case of crystalline compounds, for $T < T_{\text{comp}}$ the R subnetwork is parallel to the external applied field. For $T > T_{\text{comp}}$ the T subnetwork magnetization dominates and the R moments flip to antiparallel to H . The strong evolution of T_{comp} as a function of H is quite unusual for $R-T$ alloys and has to be explained in terms of the sperimagnetic (fan) structure of the amorphous films. As the magnetic anisotropy of Fe is weaker than the Er one, a low magnetic field orients the Fe moments more easily than the Er moments. The reinforcement of the net Fe moment projected along the field gives rise to a decrease of T_{comp} . For higher fields this effect saturates, Fe moments are collinear with the applied field and the increase of the field reinforces the collinearity of the Er subnetwork, causing an increase of T_{comp} .

The minima detected in the $M(T)$ measurements of sample "b" (T_{comp}) show the same tendency of sample "a", but the minimum in the $T_{\text{comp}}(H)$ is shifted to lower temperatures. This change in compensation temperature is very likely due to a slight difference in the composition of samples "a" and "b." It has been reported that for $\text{Tb}_{1-x}\text{Fe}_x$ amorphous alloys a variation of composition of 1 at. % can cause a change of T_{comp} up to 40 K.¹

The evolution of $T_{\text{comp}}(H)$ reported for these samples and the interpretation that we give to this phenomenon suggest that a flipping of Er and Fe subnetworks might be induced by the applied field, if a fixed-temperature slightly higher than the minimum of $T_{\text{comp}}(H)$ is chosen (Fig. 1). Macroscopic isothermal measurements performed at these temperatures, $M(H)$ or $\chi(H)$ do not allow us to probe the change of the direction of the two subnetworks. To test the validity of the proposed scheme, we need a technique able to

separate the magnetic contribution of each subnetwork and their direction, and suited to amorphous systems.

III. XMCD STUDY

X-ray magnetic dichroism is the measure of the dependence of the x-ray absorption (XAS) spectra on the helicity of x-ray photons.⁵⁻⁷ This technique is particularly valuable for our problem since: (a) it is atom selective. By tuning the photon energy to an absorption edge, it allows us to separate the magnetic contributions of the Er and Fe subnetworks; (b) it is sensitive to the direction of the magnetic moments with respect to the applied field and can therefore detect the flipping of the Er or Fe subnetwork upon application of temperature or field; (c) it does not need long-range order and can be applied to any kind of material, crystalline or amorphous, as long as a net magnetic moment exists on each subnetwork.

The XMCD measurements were carried out at SuperACO (LURE) on the SU22 beamline.⁸ The calculated circular polarization rate is 12% in the studied energy range.⁹ Due to the low XMCD signals at the Er M_4 edge¹⁰ only M_5 -edge absorption spectra ($3d \rightarrow 4f$ transitions) were carried out. Experiments were performed on sample "b" in total electron yield detection for temperatures from 4.2 to 300 K and in magnetic fields up to 5 T. The magnetic field was

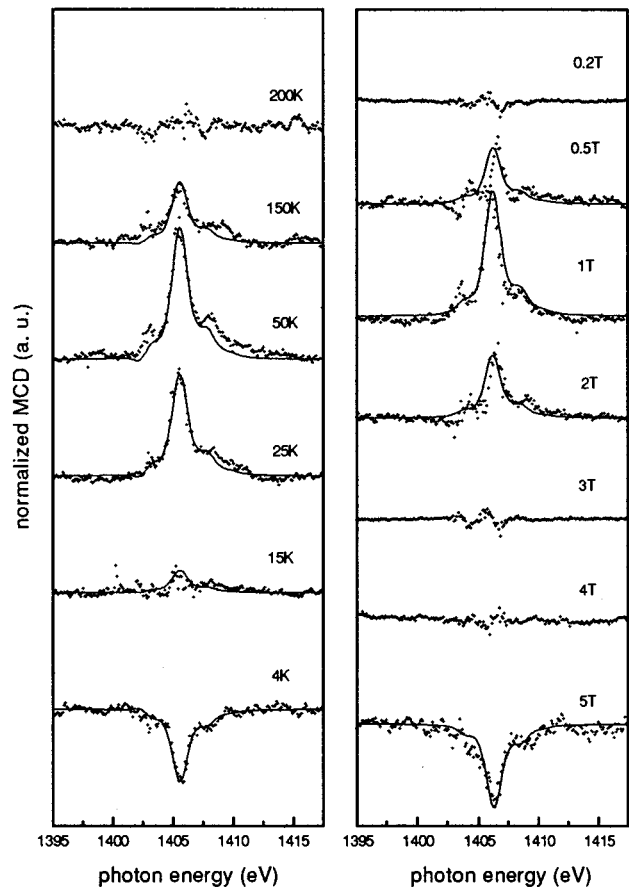


FIG. 2. (a) Temperature evolution of the Er M_5 -XMCD signal of amorphous $\text{Er}_{1-x}\text{Fe}_x$ ($x=0.73$) in a magnetic field $H=1T$. (b) Evolution of the Er M_5 -XMCD signal of the same compound as a function of the applied magnetic field and at a constant temperature of 25 K. The full lines are the theoretical fits to the data.

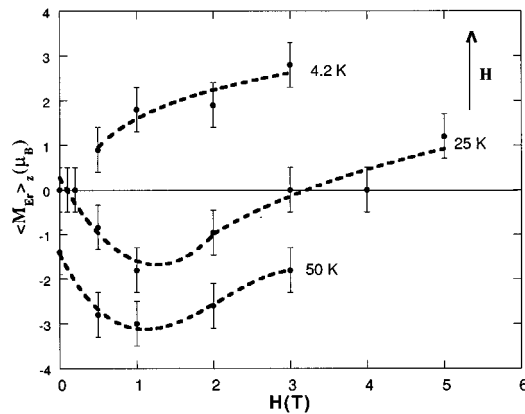


FIG. 3. Field dependence of the Er moment projected along the direction of an applied field at $T=4.2$, 25 and 50 K. Dashed lines are a guide to eye.

applied perpendicular to the sample surface and along the photon propagation direction. XMCD signals are normalized difference of two absorption spectra measured with a fixed helicity and opposite directions of the applied magnetic fields, $\sigma^+(E)$ and $\sigma^-(E)$.

In order to extract an approximate value of $\langle M_J \rangle_{4f}$ of Er from the experimental spectra, the $\sigma^+(E)$ and $\sigma^-(E)$ absorption spectra were calculated using an atomic approach. A combination of a magnetic field^{5,11} and an axial crystal field¹¹ was used to calculate a perturbed level scheme of the Er^{3+} ion which fitted the experimental spectra.

The evolution of the Er M_5 -XMCD signal as a function of temperature in an applied field of 1 T is shown in Fig. 2(a). At low temperatures (4.2 K) the signal is negative as expected for Er magnetization parallel to the applied field.¹⁰ At $T=15$ K, we are close to the compensation temperature for this field and the XMCD signal is zero. Above this temperature the signal changes sign, indicating that the Er-subnetwork magnetization is now antiparallel to the external field (flipping). Finally, the decrease of the signal for higher temperature is associated with the approach of the ordering temperature. The Er-4f moments were calculated from a theoretical fit of the experimental data. The largest moment, detected at 50 K, is of the order of $3\mu_B$ which is by far smaller than the saturated $9\mu_B$ Er-moment. Reminding that XMCD gives the average Er-moment projected along the magnetic field, this result suggests a strongly asperomagnetic structure of Er-subnetwork, as proposed in Fig. 1. This result is consistent with the low values of the net projection of the R moment which were extrapolated from macroscopic measurements on other sperimagnetic $R_{1-x}\text{Fe}_x$ alloys.¹

XMCD experiments as a function of the applied field were performed at different temperatures corresponding, in the diagram of Fig. 1, with thermal ranges in which the net moment of the R subnetwork is parallel to the field (4.2 K), antiparallel (50 K) and at a temperature (25 K) in which the isotherm crosses the dashed line of sample “b.” In Fig. 3, we show the evolution of the Er-4f moments calculated from the theoretical fit of XMCD for the three isotherms. At 4.2 K, the projection of the Er 4f-moment along the field is always

positive and increases with the field, which agrees with the fan structure proposed in Fig. 1. At $T=50$ K, this projection is always negative and a minimum in its dependence with the field is observed. The net Er 4f moment along the field is always antiparallel to the applied field as proposed in Fig. 1, increasing at low H values and decreasing at higher ones. The decrease at high fields is expected, since the field is applied in the opposite direction to the net magnetic moment direction. The increase at low fields can be explained considering the orientation of the Fe subnetwork along the field. Due to antiferromagnetic coupling between Er and Fe subnetworks, the antiparallel projection of the Er subnetwork has to follow the close-up of the angular distribution of the Fe moments.

Figure 2(b) shows selected experimental XMCD spectra measured at $T=25$ K, together with the corresponding theoretical fits. The H dependence of the fitted magnetic moments at 5, 25, and 50 K is shown in Fig. 3. At 25 K the signal is close to zero for low fields ($<0.2T$) since we are very close to the compensation temperature for these fields. For 0.5T and 1T the signal is positive, i.e., the Er moment is antiparallel to the applied field. For 3T and 4T the signal is again zero, indicating the approach of a compensation temperature. Increasing the field up to 5T induces a further flip of the Er magnetization, as expected on crossing the dashed line of Fig. 1.

In conclusion, the minima detected in the $M(T)$ curves, the Er subnetwork flips with respect to the H direction. This must be correlated with a flip in the Fe subnetwork. The different anisotropy of Fe and Er subnetworks explains the reported $T_{\text{comp}}(H)$ behavior in this sperimagnetic compound. This “exotic” behavior gives rise not only to temperature-induced but also field-induced flips of the Er.

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