Strain induced exchange-spring magnetic behavior in amorphous (TbDy)Fe₂ thin films

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In this paper, we report a strain induced exchange-spring magnetic behavior in sputter deposited (TbDy)Fe₂ amorphous thin films with phase-separated layers of (TbDy)-rich and Fe-rich at room temperature. The magnetic hysteresis loops at different strain levels were obtained with a magneto-optic Kerr effect set-up incorporating a mechanical four-point bending fixture. The unstrained film exhibits a typical ferrimagnetic hysteresis loop while the strained structure exhibits step-like hysteresis loops representative of an exchange-spring magnetic system. The mechanically strained film changes the coercivity/remanence values from positive to negative. The observed magnetic changes under strain are attributed to magnetic anisotropy modifications in the highly magnetoelastic TbDy-rich layer.

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I. INTRODUCTION

Nanocomposites of antiferromagnetically coupled hard and soft magnetic materials exhibiting an exchange-spring phenomenon have received considerable attention due to their potential applications in spintronics, magnetic data storage, and tunnel junction memory devices.¹–³ In an exchange-spring magnetic system, the soft magnetic phase exhibits magnetization reversal prior to the magnetization reversal in the hard magnetic phase due to a strong magnetic exchange coupling.⁴ A large number of exchange-spring systems studied are multilayers of RE-Fe₂/(R*Fe₂ or TM), where RE = rare earth elements, R* = Y, and TM = Fe and Co.⁵–¹⁴ In these layered systems, the film’s thickness ratio (tS/tH) and/or the materials anisotropy is used to modify the coercivity/remanence values and cannot be modified once manufactured. However, many RE-Fe₂ materials exhibit strong magnetoelastic coupling¹⁵ suggesting that the magnetic anisotropy can be changed in-situ by applying an external mechanical load (or applying an electric field induced strain), thereby tuning the exchange-spring phenomenon and/or the coercivity/remanence values. Among RE-Fe₂ materials, TbFe₂ exhibits the largest magnetostriction at room temperature but also has a large magneto-crystalline anisotropy. To reduce the magneto-crystalline anisotropy, Dy was substituted for Tb without compromising the magnetostriction significantly.¹⁶ Hence, the highly magnetoelastic (TbDy)Fe₂ is commonly studied in the literature.

In this paper, we report in-situ modification of coercivity/remanence values from positive to negative in amorphous (TbDy)Fe₂ thin films using an external mechanical load. The sputter deposited (TbDy)Fe₂ film has phase-separated layers of TbDy-rich and Fe-rich. Here, the adjective “rich” is relative to the compensation composition of (TbDy)Fe₂ at room temperature.¹⁷¹⁸ When these layered films are mechanically strained, they change from a ferrimagnetic response into an exchange-spring magnetic response with step-like hysteresis loops. This magnetic behavior change is attributed to the strain induced magnetic anisotropy modifications in the magnetoelastic TbDy-rich layer.

II. EXPERIMENTAL

Amorphous (TbDy)Fe₂ thin films with phase-separated layers of TbDy-rich and Fe-rich were produced from a single Tb₀.₃Dy₀.₇Fe₁.₉₂ alloy target (Etrema Inc., USA) using DC magnetron sputtering. The films were deposited onto a 100 mm diameter silicon (100) wafer (0.5 mm thick) with a 500 nm SiO₂ barrier layer. Process parameters used during the sputtering were argon pressure of 8 × 10⁻⁶ Torr, base vacuum of 7 × 10⁻¹⁰ Torr, DC power of 300 W, sputtering rate 1.6 nm/s, and substrate to target distance of 4 cm. The deposition process was continuous over a 5 min period. During deposition, the target temperature increased from room temperature to 550 °C while the substrate temperature intrinsically increased to approximately 200 °C. This thermal ramp leads to a phase separated layered structure. Following deposition, the wafer was diced into several pieces of different sizes for film characterization.

Several characterization tools were used to evaluate the sputter deposited films. A JEOL JXA-8230 SuperProbe, an electron beam microprobe analyzer, with a wavelength dispersive spectrometer (WDS) was employed to determine the chemical composition in samples of size 5 mm × 5 mm taken near the center portion of the wafer. The average compositions of the entire film measured with WDS were 65 at. % of Fe, 22 at. % of Dy, 11 at. % of Tb, and 2 at. % of oxygen. The film’s average composition is close to the target composition with a small amount of oxygen traces present (Target...
composition - Fe: 66 at. %, Dy: 23 at. %, and Tb: 11 at. %). The film’s magnetic properties were measured with a Superconducting Quantum Interference Device (SQUID) from Quantum Design. Saturation magnetization ($M_s$) and coercivity ($H_C$) of the as deposited films were 74 emu/cc and 80 Oe, respectively. The large reduction in $M_s$ from its crystalline counterpart (∼800 emu/cc) is due to its sperimagnetic nature where antiferromagnetic alignment between the Fe and Tb/Dy dominates. The measured $M_s$ is very close to previously reported values (Su et al.19) for the amorphous (TbDy)Fe$_2$ film. An FEI Titan Scanning Transmission Electron Microscope (S-TEM) operated at 300 kV was used to characterize the film’s microstructure. A cross-sectional TEM sample was prepared by the lift-off technique using a dual-beam, scanning electron microscope (SEM) focused ion beam (FIB), system. The specimen was further nano-milled using FISCHIONE liquid nitrogen cooled with a low energy NanoMill to remove surface amorphization caused by the FIB ion beam.

The magnetic response ($M$ vs $H$) of the film was measured in a transverse mode magneto-optic Kerr effect (MOKE) set-up incorporating a mechanical four-point bending fixture. The MOKE measurement consists of a linearly polarized laser light with 633 nm wavelength passing through a photoelastic optical phase modulator (PEM) and onto the film’s surface (0.5 mm spot size) with a penetration depth of ∼50 nm. The reflected light, now in an elliptically polarized state with a rotated major axis, is detected by a photodetector after it passes through an analyzer and is correlated with the sample’s magnetization change. A four point bending fixture was used to apply mechanical loads based using American Society of Testing Materials ASTM standards.20 The fixture has two inner fixed/supporting pins and two outer loading pins placed at an equal distance of 5 mm and 10 mm, respectively, from the center point as shown in the inset of Figure 2(a). This produces a uniform axial strain in the film between the two inner fixed pins without thickness variations due to the relatively thin film thickness compared to the substrate thickness. (TbDy)Fe$_2$ specimens (25 mm × 5 mm) diced from 4 different wafers with strain gauges attached to the back surface, were mounted in the four-point bending fixture placed in the MOKE. The magnetic testing was performed under the transverse mode with two orientations, i.e., first with the magnetic field applied parallel to the specimen’s long axis ($H||X$) (see the inset in Figure 2(a)) and second with the magnetic field applied perpendicular to the long axis (i.e., $H\perp X$). MOKE measurements were made on 4 different specimens at 10 different strain levels ($X$ directed strain along the long axis) varying from 0% up to 0.12%.

**III. RESULTS AND DISCUSSION**

Figure 1 shows cross sectional TEM micrographs of a 400 nm thick film deposited on thermally grown SiO$_2$ barrier layers and a protective Pt layer (Figure 1(a)) comprised of alternating 15–20 nm thick layers (Figure 1(b)). There is apparent waviness in the alternating layers and we attribute this to the self-assembly process rather than an artifact of microscope since the adjacent SiO$_2$ and Pt layers lack waviness. The alternating layers with lighter and darker contrasts indicate the presence of different magnetic properties as reported by Amish et al.21 Based on WDS measurements coupled with MOKE measurements, the alternating layers appear to be TbDy-rich and Fe-rich layers. Once again the adjective “rich” is relative to the compensation composition of (TbDy)Fe$_2$ at room temperature. The high resolution TEM micrograph (Figure 1(c)) shows an absence of long-range order indicating that the layers are amorphous, which is consistent with measured x-ray diffraction data.

Figure 2 shows representative MOKE in-plane hysteresis loops, measured along the sample’s long axis, the $X$ axis parallel to applied field $H$, i.e., $H||X$ for one representative specimen. Figure 2(a) shows that the specimen’s response without a mechanical load produces a coercivity of 80 Oe and a normalized remanence of 0.72. MOKE measurements along the shorter $Y$ axis, $H||Y$, reveal a similar magnetic response indicating that the film is magnetically isotropic in-plane. Figure 2(b) shows that the $H||X$ response for an applied strain of 0.057% produces a step-like hysteresis behavior in both forward and reverse magnetic field directions on the same sample. The first magnetization reversal (i.e., decreasing magnetic field) takes place at $-60$ Oe with a normalized remanence of 0.72 and the second magnetization reversal occurs at $-300$ Oe. Similarly, Figure 2(c), $H||X$ measured with an applied strain of 0.094%, shows a similar step-like hysteresis loop but with the first magnetization reversal at 10 Oe (negative coercivity) with a normalized remanence of 0.2 and the second reversal at $-330$ Oe (i.e., decreasing field). Figure 2(d), $H||X$ measured with an applied strain of 0.12%, produces a first magnetization reversal at 40 Oe (negative coercivity) and a normalized remanence at $-0.44$ (negative remanence). Each of the magnetic hysteresis loops shown in Figures 2(b)–2(d) is representative of magnetic exchange-spring systems. However, previous magnetic exchange-spring material systems studied required modification to the samples thickness or material constituents to change the magnetic exchange-spring system rather than a single specimen as presented in Figure 2.

The magnetic response of the unstrained layered amorphous film shown in Figure 2(a) displays hysteresis loop representative of a soft ferrimagnetic material, i.e., Terfenol-D is soft ferrimagnetic. This suggests that spin reorientation during 180 degree reorientation is dominated by the external magnetic field against a weak demagnetization field (i.e., shape anisotropy of the thin film). Specifically, since the films are sufficiently thin, the shape anisotropy produces an easy axis in-plane, which is supported by the isotropic M vs H curves measured along the longer $X$ and shorter $Y$ axes. When sufficient magnetic field is applied in a direction opposite to the layers’ magnetized state, the magnetic spins in both layers (see Figure 1(b)) coherently reorient 180 degrees producing a typical ferrimagnetic M vs H curve.

The film’s magnetic exchange-spring behavior, shown in Figures 2(b)–2(d), indicates that the TbDy-rich and Fe-Rich layers become antiferromagnetically coupled with an applied mechanical strain. Specifically, Figure 2(d) with negative remanence and negative coercivity indicates that
FIG. 1. TEM micrographs of the 400 nm thick sputter deposited amorphous (TbDy)Fe₂ thin film. (a) SiO₂ barrier layer with protective Pt layer. (b) Layered structure consisting of 15-20 nm thick layers as can be seen with darker (Layer 1) or lighter (layer 2) contrast. (c) High resolution TEM (HRTEM) micrograph confirming that each of the two layers is amorphous.

FIG. 2. MOKE hysteresis loops measured along the X axis (Hₓ) at four different strain levels. (a) Hysteresis loop of the specimen with zero applied mechanical strain, right bottom inset shows the schematic of four point bending geometry. (b) Hysteresis loops measured along the X direction with an applied mechanical strain of 0.057%. (c) Hysteresis loop measured at 0.094%, right inset shows the magnified plot near zero magnetic field. (d) Hysteresis loop at an applied mechanical strain of 0.12% exhibiting negative coercivity and negative remanence, right inset shows the magnified plot near zero magnetic field.
antiferromagnetic exchange coupling must be present for this reversal to occur prior to removing the magnetic field (i.e., before \( H = 0 \)). These data also indicate that one layer is relatively harder while the other has become relatively softer (i.e., compared to Figure 2(b)) based on the change in coercivity and the step like behavior observed. We believe that this is caused by the TbDy-rich RFe\(_2\) layer, which is strongly magnetoelastic while the Fe-rich RFe\(_2\) layer is weakly magnetoelastic. We base this conclusion from previous studies on amorphous TbFe\(_2\) showing that Tb-spin dominates for compositions above \( \sim 23 \) at. % Tb\(^{\text{17}}\) while the Fe-spin dominates for compositions below this compensation composition at room temperature. Furthermore, the saturation magnetostriction, \( \lambda_s \), of Tb-rich amorphous TbFe\(_2\) is one order of magnitude larger than the Fe-rich Tb-Fe\(_2\) layer.\(^{\text{22}}\) Since both TbDy-rich and Fe-rich layers are amorphous without magnetocrystalline anisotropy, the TbDy-rich produces greater changes in the elastically induced magnetic anisotropy when a mechanical strain is applied. Specifically, one can approximate the magnetic anisotropy as \( K = \frac{1}{2} \lambda_s E \varepsilon \) where \( E \) is the Young’s modulus and \( \varepsilon \) is the uniaxial strain applied. Since the TbDy rich region has an order of magnitude larger saturation magnetostriction than the Fe-rich layers, the \( K \) increases more rapidly as the strain increases for the TbDy rich layers.\(^{\text{22}}\) This \( K \) increase produces a noticeable step-like hysteresis loop, exchange-spring magnet signature, which appears when a mechanical strain is applied.

To provide more quantitative changes produced in the (TbDy)Fe\(_2\) layered system as a function of strain, Figure 3(a) provides representative response of the soft and hard magnetic layers superimposed on the measured magnetic response for the strain \( 0.043\% \). The figure defines coercivity values \( H_C \) and magnetization changes \( \Delta M \) for the hard (TbDy-rich) and soft layers (Fe-rich) (i.e., subscripts H and S). Figure 3(b) plots these coercivity values (\( H_C^H \) and \( H_C^S \)) and magnetization changes (\( \Delta M^H \) and \( \Delta M^S \)) as a function of the applied strain. The results show that \( H_C^S \) remains relatively constant (80 Oe) from 0% to 0.043% and decreases linearly with larger strains thereafter. In contrast, the \( H_C^H \) increases substantially as the strain increases from 0% to 0.043% recognizing that the \( H_{CH} = H_{CS} = 80 \) Oe for 0 strain. For strains larger than 0.043%, \( H_{CH} \) increases more moderately as contrasted with strain values below 0.043% (i.e., see data points from 0.06%–0.12%). Focusing on \( \Delta M \) values presented in Figure 3(b), i.e., \( \Delta M^H \) and \( \Delta M^S \) initially increase (soft) or decrease (hard), respectively, for the strain values below 0.043%. At strain levels above 0.043%, both \( \Delta M^S \) and \( \Delta M^H \) are relatively constant with a ratio defined by \( \sim \Delta M_S/\Delta M_H = 2.5 \).

IV. CONCLUSION

This investigation experimentally demonstrated a strain induced exchange-spring magnetic behavior in a layered ferrimagnetic system at room temperature. Strain induced exchange–spring behavior can also be obtained by using a piezoelectric substrate with a voltage induced strain. The piezoelectric substrate with patterned electrodes can also be used to overcome the substrate clamping\(^{\text{23}}\) issue normally observed. Furthermore, the advantage of this method over a conventional exchange-spring magnet system studied is that the coercivity/remanence of the system can be mechanically tuned.

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