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Tailoring anisotropy and domain structure in amorphous TbCo thin films through combinatorial methods

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Abstract.

We apply an in-plane external magnetic field during growth of amorphous TbCo thin films and examine the effects on the magnetic anisotropy and domain structure. A combinatorial approach is employed throughout the deposition and analysis to study a continuous range of compositions between 7-95 at.% Tb. Magnetometry measurements show that all samples have a strong out-of-plane anisotropy, much larger than any in-plane components, regardless of the presence of a growth field. However, magnetic force microscopy demonstrates that the growth field does indeed have a large effect on the magnetic domain structure, resulting in elongated domains aligned along the imprinting field direction. The results show that the anisotropy can be tuned in intricate ways in amorphous TbCo films giving rise to unusual domain structures. Furthermore the results reveal that a combinatorial approach is highly effective for mapping out these material properties.

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1. Introduction

Thin films of TbCo are well known for their strong perpendicular magnetic anisotropy (PMA)[1, 2] which makes them of interest for a range of magnetic storage and spin-valve technologies.[3, 4] Furthermore, it has recently been shown that all-optical magnetic switching (AOS)[5] can be achieved in Tb(Co,Fe) films,[6, 7] allowing the manipulation of magnetic domains on much shorter timescales than is possible with magnetic fields. AOS relies on the ferrimagnetic nature of the TbCo, where the anti-aligned Tb and Co magnetic sublattices compensate each other at a given temperature, resulting in zero net magnetization. The compensation temperature can also allow the generation of domain walls using thermal or composition gradients with potential applications in domain wall memories.[8, 9] Both the compensation temperature and PMA can be engineered by building heterostructures combining TbCo with other materials.[10, 11] This allows great flexibility in tuning the magnetic properties and can even open up new possibilities such as interlayer coupling through proximity induced magnetism.[12]

Amorphous films are particularly interesting in the context of heterostructures as they form exceptionally flat and homogeneous layers[13, 14, 15] and different materials can be combined without having to consider differences in lattice constants.[12] In addition, a magnetic anisotropy can be imprinted in amorphous films in an arbitrary direction by applying a magnetic field during growth.[16] In SmCo (another rare-earth-transition metal compound) it has for example been shown that such an imprinted anisotropy can be very large [17] and lead to unusual magnetic domain structures.[18]

Here we explore the imprinting of anisotropy in amorphous TbCo thin films and its effect on domain structure. We use a combinatorial approach[19, 20] whereby material is deposited from two separate sources on a large wafer under an angle, so that a continuous composition gradient is achieved. This allows us to map out the properties of a continuous range of compositions much more efficiently than with conventional methods. We find that, while the perpendicular anisotropy far outweighs the imprinted anisotropy, the domain structure is still very sensitive to the small imprinted anisotropy component.

2. Experimental details

TbCo thin films were deposited by a combinatorial technique using DC magnetron sputtering. The samples were prepared in an ultra-high vacuum chamber, with a maximum base pressure of 3×10^{-9} Torr, at room temperature using 99.999 % pure Ar as a sputtering gas at a pressure of 1×10^{-3} Torr. The films were deposited on 10 mm wide strips cut from naturally oxidized 3-inch Si(100) wafers which were pre-heated, at base pressure, to 650 °C for 20 min and cooled down to room-temperature before deposition. To ensure the amorphousness of the films, a buffer layer of amorphous $\text{Al}_{80}\text{Zr}_{20}$ with a nominal thickness of 3 nm was deposited on the native oxide of the Si.[15] The chamber geometry was such that Co and Tb targets were positioned facing each other at opposite ends of the substrate. Figure 1(a) shows the target and substrate configuration.

By co-sputtering from the Co and Tb targets, with a non-rotating substrate, a composition spread across the sample was created, as shown in fig. 1(a). In total, a continuous composition range from 7 to 95 at.% Tb was achieved over three wafer strips each with a nominal film thickness of 50 nm at the center of each slice. To protect the TbCo layer from oxidation a capping layer of 3 nm $\text{Al}_{80}\text{Zr}_{20}$ was deposited. Both the buffer and capping layer were deposited with a rotating substrate to ensure a homogeneous composition and thickness. One set of films was grown in a sample holder with two permanent magnets creating a nearly homogeneous static in-plane magnetic field of about 130 mT, see fig. 1(a), whereas another set of films was grown without this magnetic field. The magnetic field was applied in the direction $\phi = 90^\circ$, perpendicular to the composition gradient direction which we define as $\phi = 0^\circ$.

Rutherford backscattering (RBS) measurements were performed at several points along the Tb-Co gradient to determine the composition. The yield of each spectrum was normalized to the total count of the spectrum to enable comparison between different RBS measurements. For each spectrum the peaks of each element were integrated giving the yields Y_{Tb} and Y_{Co} . By simultaneously solving the two equations

$$\frac{Y_{\text{Tb(Co)}}}{Y_{\text{Tb}} + Y_{\text{Co}}} = \frac{x_{\text{Tb(Co)}} Z_{\text{Tb(Co)}}^2}{x_{\text{Tb}} Z_{\text{Tb}}^2 + x_{\text{Co}} Z_{\text{Co}}^2} \quad (1)$$

for each element Co and Tb together with the condition

Figure 1. (Color online) (a) A diagram of the target configuration and substrate holder. The substrate is mounted in between two permanent magnets producing a growth field $B_g \approx 130$ mT at the center. The composition gradient is represented as a colour gradient. The upper right corner shows the position of the substrate holder with respect to the targets. The angle ϕ is also defined. (b) A representative measurement of the Tb concentration gradient across a sample. The markers are the values measured with RBS and the line is the fit used to determine compositions at intermediate points.

that $x_{\text{Tb}} + x_{\text{Co}} = 1$ the elemental concentrations $x_{\text{Tb}(\text{Co})}$ in at.% were determined. Here, $Z_{\text{Tb}(\text{Co})}$ are the atomic numbers of each species. The composition gradient was found to be almost linear versus position as shown in fig. 1(b). A linear fit was therefore used to extrapolate the composition along the entire sample length giving a composition gradient of $\Delta x_{\text{Tb}} = 0.4\text{--}0.6$ at.%/mm. This implies that the variation in concentration over the probed area of each measurement point (less than 4 mm diameter), is 1.5–2.4 at.%. The error bars in fig. 1 represent this uncertainty, which is smaller than the experimental uncertainty of RBS.

X-ray reflectivity (XRR) and grazing incidence X-ray diffraction (GIXRD) were measured at several points along the Tb-Co gradient with Cu K_α radiation using a Bruker D8 Discover in a parallel beam geometry. A Göbel mirror was used on the incident side as well as beam-shaper slits to limit the measured area. The reflected/diffracted beam was measured using a Lynx EYE detector. For GIXRD an incident angle of $\omega = 1^\circ$ was used. The probed area in each measurement was about 8 mm $\times$ 10 mm, with the long dimension perpendicular to the Tb-Co gradient. In this direction the Tb-Co composition should be constant,

see fig. 1.

Both longitudinal and polar magneto-optic Kerr effect (L- and P-MOKE) measurements were used to determine the magnetic properties of the samples at room temperature. The diameter of the laser spot on the sample was about 1–2 mm. An in-plane or out-of-plane magnetic field was applied (continuously measured with a Hall probe) and magnetization loops were recorded at different points on the samples, along the Tb-Co gradient. To study the in-plane anisotropy the samples were also rotated around the azimuthal angle ϕ where $\phi = 0^\circ$ corresponds to the direction where the field is applied parallel to the composition gradient, fig. 1.

Higher field magnetic characterization was carried out as a function of temperature and composition using a Cryogenic Ltd. vibrating sample magnetometer (VSM). These measurements were performed on cleaved samples, no larger than 6.8 mm wide along the gradient direction. Magnetization loops with a field of up to 5 T applied both perpendicular to the plane and in the plane of the samples in the temperature range 10 to 320 K were measured. The in-plane measurements were performed at $\phi = 90^\circ$, i.e. the field is applied perpendicular to the composition gradient. The diamagnetic background from the substrate was subtracted by a linear fit to the high field parts of each magnetization scan. Magnetic moments were calculated using the magnetic film thicknesses extracted from XRR fitting.

Magnetic force microscopy (MFM) was performed with a Nanosurf Mobile S atomic force microscope and MFM01 series tips from NT-MDT. All measurements were done in phase contrast mode.

3. Results and discussion

3.1. Structural Properties

Some examples of GIXRD scans are shown in the inset in fig. 2. These measurements show that all samples up to about $x_{\text{Tb}} = 80$ at.% Tb are X-ray amorphous as seen by the presence of only one broad low-intensity peak typical of amorphous samples with a lack of long-range atomic order.[21] The angular position of this broad peak gives a measure of the average atomic separation in the film, which increases with Tb content consistent with the larger lattice parameter of hcp-Tb (3.60 Å) compared to that of hcp-Co (2.51 Å). By inserting the average atomic spacing and full-width at half-maximum (FWHM) into the Scherrer formula [22] the correlation length (sometimes referred to as the grain size) can be estimated, as shown in the main panel of fig. 2. For small Tb concentrations the correlation length is almost constant at about 10 Å with only a slight increase with Tb content, which

Figure 2. (Color online) X-ray correlation length versus Tb composition for the entire range studied. The inset shows examples of some GIXRD scans for different compositions (offset for clarity). The broad peak is an indication of an amorphous structure. The scan for the highest Tb concentration shows a sharp peak which is a signature of at least partially crystalline structure.

Figure 3. (Color online) XRR scan (points) and a fitted curve (solid line) for $x_{\text{Tb}} = 46$ at.%. The inset shows the layer model used to fit the XRR data.

can be attributed to the change in atomic separation mentioned above. At approximately $x_{\text{Tb}} = 80$ at.% there is a sudden increase in the correlation length which can be interpreted as an onset of crystallization. GIXRD on the samples grown without a field have the same appearance and give the same atomic spacings, FWHM and correlation length as the corresponding field-grown samples.

XRR measurements were used to determine the thickness and quality of the layering in the samples. A representative XRR scan can be seen in fig. 3. Clear interference fringes arising from the total thickness of the films can be observed up to $2\theta = 6^\circ$, attesting to their smoothness. By fitting the data to the layer model shown in the inset it is possible to extract

Figure 4. (Color online) Room temperature in-plane and out-of-plane hysteresis loops for $x_{\text{Tb}} = 13$ at.%, measured with VSM. A strong out-of-plane anisotropy is observed. The slight difference in saturation magnetization can be attributed to sample alignment. The in-plane angle was $\phi = 90^\circ$, i.e. perpendicular to the composition gradient.

the layer thicknesses, densities and roughnesses. Low surface roughnesses (root-mean-squared) in the range 0.6–1 nm are obtained for films with a total thickness of 52–57 nm which is similar to other amorphous rare-earth-transition metal compound films.[17] As for GIXRD, XRR showed no significant differences between samples grown with and without a magnetic field.

3.2. Magnetic Properties

A combination of MOKE and VSM measurements in different geometries was used to map out the magnetic properties of the TbCo. Films with a Tb concentration below $x_{\text{Tb}} = 45 \pm 3$ at.% were ferrimagnetic at room temperature, which is a slightly larger composition than the 38 at.% reported by Betz et al.[23] In this composition range, the films have a strong out-of-plane anisotropy, as seen in the characteristic VSM measurements presented in fig. 4. In the out-of-plane direction, the hysteresis loop is square with a large (temperature dependent) remanent magnetization whereas in the in-plane direction the loop is smoothly varying with a small remanence and large saturation field. This is observed for films grown both with and without an applied magnetic field. However, subtle differences are seen in the in-plane magnetization for these two cases. Figure 5 shows in-plane hysteresis loops along two perpendicular directions in the plane, for samples grown with and without field. Samples grown without field [fig. 5(a)] are isotropic in the plane as seen by the identical loops along the two directions. In contrast, for samples grown in a field [fig. 5(b)] an opening is observed in the hysteresis loop in the direction parallel to the

Figure 5. (Color online) In-plane minor loops measured in the L-MOKE geometry both parallel ($\phi = 0^\circ$) and perpendicular ($\phi = 90^\circ$) to the composition gradient for (a) the sample grown without an external field and (b) the sample grown in an in-plane magnetic field along $\phi = 90^\circ$. The composition at the point measured was approximately $x_{\text{Tb}} = 10$ at.% for both samples.

growth field whereas perpendicular to the growth field the hysteresis is identical to that of the sample grown without a field. This shows that, despite the out-of-plane direction being the overall easy axis, there is a component (of the easy axis) which lies along the growth field direction. This shows that the external growth field has indeed imprinted a small in-plane anisotropy component.

Out-of-plane magnetization measurements by VSM over the temperature range 10–320 K confirm the ferrimagnetic ordering in the films. A compensation temperature T_{comp} is observed where the coercivity diverges and the magnetization is zero, as shown in fig. 6. This is due to the different temperature dependence of the magnetization of the anti-aligned Tb and Co magnetic sublattices which cancel each other out at T_{comp} . With increasing Tb content T_{comp} is seen to increase and for compositions above 21 at.% Tb, T_{comp} is above room temperature (see inset of fig. 6). This value corresponds well with previously reported values.[23, 24]

The out-of-plane coercivity is strongly dependent on temperature and composition as seen in fig. 6. For the composition shown in the main graph ($x_{\text{Tb}} = 20.2$ at.%) the coercivity is larger at room temperature since it is close to T_{comp} . For the smaller composition of $x_{\text{Tb}} = 13$ at.% [fig. 4],

Figure 6. (Color online) The temperature dependence of the out-of-plane remanence and coercivity, for a sample piece with $x_{\text{Tb}} = 20.2$ at.% at the center, as measured with VSM. The coercivity has a singularity and diverges at 271 K while the remanence goes to zero at this compensation temperature, T_{comp} . The inset shows the measured T_{comp} versus composition.

the coercivity is instead quite small. In this case the measurement is performed at room temperature which for this composition is much higher than T_{comp} . Generally, for all compositions the coercivity decreases with increasing temperature above T_{comp} while for decreasing temperatures below T_{comp} H_c initially decreases, but eventually reaches a minimum and for even lower temperatures increases slightly once again.

Magnetic force microscopy was used to examine the magnetic domain structure of the films for several different compositions as shown in fig. 7, specifically to compare the samples grown with and without an external field. At around 7 at.% Tb (not shown in fig. 7), the sample grown with an external field exhibits a similar labyrinthine domain structure to that of the sample grown in-field at 8 at.% [fig. 7(c)]. For a slightly higher Tb concentration around 9 at.%, [fig. 7(b) and (d)], there is a clear divergence in the domain structures between the two samples. For the sample grown without an external field, the domains begin to align parallel to the composition gradient, while for the sample grown in-field the domains start to align parallel to the growth field. This reinforces the idea that the growth field does indeed influence the zero-field magnetization in the sample, as was suggested by the L-MOKE results, see fig. 5. To be sure that this effect was in fact related to the growth field, several MFM images were measured for each sample and composition. Before each measurement, the sample was exposed to an external in-plane field of 700 mT (simulating an L-MOKE scan) applied either parallel or perpendicular to the composition gradient. However, the trends described above remained constant regardless of the

direction of the most recently applied field. This gives strength to the assertion that the magnetic structure of the samples is imprinted during the growth process. For higher Tb concentrations the domains grow in size. This evolution is faster for the sample grown with an external field than the sample grown without. At 11 at.% the domains of the field-grown sample become so large that several MFM scans in different surface locations failed to contain any domain boundaries. Hence, these images exhibited little contrast and are therefore omitted in fig. 7.

The origin of the PMA in Tb-Fe amorphous films has been shown to be related to different pair correlations between the Tb-Fe, Tb-Tb and Fe-Fe atomic pairs in the plane and perpendicular to the plane [25]. These differences are induced by the broken symmetry at the interfaces of the film during growth and it is likely that the same applies to the origin of the PMA in TbCo. The origins of field induced magnetic anisotropy in amorphous materials are at present less well understood. It is thought that the magnetic field induces changes in the local atomic configuration [26] in the form of alignment of atomic moment pairs via dipolar effects [27], alignment of atomic clusters via local spin-orbit coupling (single-ion anisotropy) [28], or direction dependent bonding between atoms of different elements as described above [25]. Imprinted anisotropy has also been linked with the strain induced during growth through magnetoelastic coupling [28]. Although we cannot distinguish between these mechanisms here it is clear that the directional dependence of pair correlations induced by the film surfaces far outweighs the changes in the local configuration or the magnetoelastic strain induced by the growth field.

4. Conclusions

We have shown how combinatorial methods are valuable in mapping out various material properties such as amorphicity and magnetic anisotropy versus composition. We have furthermore shown that the magnetic properties of TbCo are very sensitive to composition and are continuously tunable, meaning that the desired properties can be obtained by carefully selecting the composition. This tunability, in combination with the amorphous structure and smooth interfaces, makes amorphous TbCo films ideal for perpendicular exchange coupled multilayer structures. Furthermore, these TbCo films are a new example of the possibilities associated with imprinting magnetic anisotropy in amorphous alloys. Even though TbCo exhibits a strong intrinsic out-of-plane anisotropy for all compositions, imprinting an in-plane anisotropy is still possible resulting in a tilt of the easy axis

away from the film normal. The direction of this tilt can be controlled by the growth field and the composition gradient. The magnetic domain structure is strongly affected by the anisotropy and can thus be controlled by manipulating the composition and in-plane growth field. The possibility to control the orientation of the resulting elongated domains can be useful in applications such as magnetic storage, magnetic logic, and magnonic devices.

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Figure 7. (Color online) Domain structures for the sample grown in an external field (a) and (b), and grown without an external field (c)-(f). The compositions are (a) 8.5 at.%, (b) 9.2 at.%, (c) 8.5 at.%, (d) 9.3 at.%, (e) 11.5 at.%, and (f) 13.0 at.% Tb, all with an uncertainty of ± 0.4 at.%. Dark and light regions correspond to areas where the sample magnetization points into or out of the sample plane, respectively. Field-grown samples with compositions corresponding to (e) and (f) do not show any domain boundaries in the scale measured with MFM, and are hence left out here. The field-grown sample with 7 at.% shows a similar pattern to (c), i.e., without any elongation. This composition was not accessible on the sample grown without an external field.

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