Tuning in-plane magnetic anisotropy and temperature stability in amorphous trilayers

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Keywords: Amorphous magnetic layered structures can be realised by magnetron sputtering. Artificially layered magnetic systems display a remarkable variety of physical phenomena, with important technological applications in e.g. information processing and storage [1]. For a long time, much effort has been devoted to epitaxial growth and obtaining atomically flat interfaces [2]. However, recently the interest in magnetic materials with graded interfaces is growing [3] and it has e.g. been shown that this intermixing at the interface can improve the magnetic properties of exchange-spring magnets [4]. Exchange-spring magnets consist of combined nanometer-scale hard and soft magnetic phases. They were originally investigated mainly with the purpose of obtaining strong permanent magnets, containing less rare earth elements while still achieving a high energy product [5–8]. For permanent magnets, the goal is to obtain a rigid coupling between the two phases, so that the magnetisation of the entire soft magnetic layer follows the hard magnetic phase. The exchange-spring regime, where the spin orientation in the soft phase is non-uniform instead, is obtained if the thickness of the soft layer is larger than a critical thickness of typically 10 nm [9]. Such exchange-spring magnets are of interest for achieving suitable magnetisation reversal in spintronics applications [10,11].

Amorphous magnetic layered structures can be realised by magnetron sputtering. With this process, smooth interfaces can be achieved, with no crystalline defects or strain from lattice mismatch [12]. The magnetic properties of each layer can be tuned by the composition and the interfaces are naturally graded due to a degree of intermixing occurring as a result of the sputtering process [13,14]. For amorphous alloys with d-electrons responsible for the magnetism, the local anisotropy is to a large extent averaged out, yielding very low coercivity as explained by the random anisotropy model [15,16]. If the film deposition is homogeneous, which can be ensured e.g. by rotating the substrate during sputtering, one could naively expect isotropic in-plane magnetic response. However, uniaxial in-plane magnetic anisotropy is often observed in sputtered amorphous soft-magnetic films [17–21]. This uniaxial anisotropy may have multiple origins. Depending on the properties of the substrate, it has been shown to be due to bond-orientational anisotropy in some studies [18,22] and to magnetoelastic anisotropy from (residual) strain in the film in others [19,20,23]. Having soft magnetic films with a uniaxial anisotropy is important for e.g. spintronic applications, magnetic recording, magnetic sensors and microwave devices and it is also possible to imprint a well-defined uniaxial anisotropy by sputtering in a magnetic field [24,25].

In amorphous alloys with f electrons, e.g. Sm-Co, the random anisotropy strongly affects the interactions, and may give rise to hard magnetic behaviour if the anisotropy is too strong to be averaged out [15,16]. Amorphous Sm-Co films sputtered onto rotating amorphous substrates are typically magnetically isotropic in-plane, but a uniaxial anisotropy can be induced by applying an external magnetic field during deposition [26–28].

In this study, we have investigated trilayer structures with a configuration of either hard/soft/hard or soft/hard/soft amorphous magnetic layers.
also performed in a non-commercial Magneto-Optic Kerr Effect (MOKE) anisotropy. Measurements of angle-dependent hysteresis loops were performed in-plane, using VSM. The results show a large dependence on the easy axis direction. Frisk et al. [30] showed that for both polycrystalline and amorphous Co-Fe-Zr films the experimental M_r/M_s versus angle 

2. Sample fabrication and experimental methods

Thin film samples with amorphous layers of the magnetically soft Co_{85}(Al_{70}Zr_{30})_{15} (CAZ) and magnetically hard Sm_{12}Co_{81}Ti_{7} (SCT) were grown using DC magnetron sputtering in a non-commercial ultra high vacuum (UHV) system with a base pressure below 10^{-6} Torr (typically in the low 10^{-9} Torr range) [29,30]. Before deposition, Ar gas with a pressure of 2 mTorr was introduced to the sputtering chamber. Highly pure (99.99 at.%) sputtering targets with a 3-inch diameter were used, consisting of an Al_{70}Zr_{30} composite, as well as Co, Sm, and Ti, respectively.

The substrates, single crystalline Si(100) with a native oxide and an area of ~1 cm², were rotated during deposition at a rotational speed of 20 rpm, to ensure a homogeneous deposition. The sputtering yield of each target was calibrated in order to calculate the sputtering time and power required for the desired thicknesses and compositions. For all samples, 4–5 nm thick layers of Al_{70}Zr_{30} were used as buffer layers to ensure amorphous growth of the following magnetic layer and as capping layers to prevent oxidation of the material underneath [12]. The capping layer becomes naturally oxidised in air, and the resulting 1–2 nm thick layer of self-passivating Al_{2}O_{3} prevents any further oxidation of the sample. The magnetic trilayer samples with the two respective configurations of CAZ/SCT/CAZ and SCT/CAZ/SCT are illustrated in Fig. 1. For the configuration with the magnetically soft alloy at the centre, multiple samples with a range of soft layer thicknesses d = 10 – 50 nm were produced and investigated.

To confirm that the chosen sputtering parameters produced X-ray amorphous samples, grazing incidence X-ray diffraction (GI-XRD) was performed on single films of Co_{85}(Al_{70}Zr_{30})_{15} and Sm_{12}Co_{81}Ti_{7}, at 1° incidence for a 20° range of 10 – 80° in a Bruker D5000 system with Cu Kα radiation (λ = 1.5418 Å). X-ray reflectivity (XRR) was measured in a Bruker D8 Discover system, also with Cu Kα radiation, for a 20° range of 0.2 – 5°. To extract the information on film thickness, density, and interface roughness, the XRR data was fitted in the software GenX [31]. The phases Co_{85}(Al_{70}Zr_{30})_{15} (CAZ) and Sm_{12}Co_{81}Ti_{7} (SCT) are written in abbreviated form in the table, with the nominal thickness of the respective layer given in parenthesis. Both the buffer and capping layers consist of Al_{70}Zr_{30}.

3. Results and discussion

3.1. Samples and structural characterisation

It was inferred from the absence of any sharp peak in the measured GI-XRD patterns that the sputtered samples are X-ray amorphous. The thickness of each individual layer in all samples is presented in Table 1.

3.2. Magnetostatic properties

The room temperature saturation magnetisation, M_r, of all samples as a function of the relative content, in vol.% of Sm_{12}Co_{81}Ti_{7} in the magnetic part of the film is reported in Fig. 2. These measurements are performed in-plane, using VSM. The results show a linear trend for M_r versus composition, with a decreasing saturation as the amount of Sm_{12}Co_{81}Ti_{7} increases. The hysteresis loops from Sm_{12}Co_{81}Ti_{7} films are isotropic, while the Co_{85}(Al_{70}Zr_{30})_{15} film exhibits a clear uniaxial anisotropy. The normalised remanent magnetisation, M_r/M_s, is determined from angle dependent MOKE hysteresis loop measurements. Fig. 3 shows the resulting M_r/M_s versus angle ψ Succ, defined with respect to a sample edge, for the single layer Co_{85}(Al_{70}Zr_{30})_{15} film of 40 nm. According to the Stoner-Wohlfarth (SW) model, valid for a single magnetic domain with uniaxial anisotropy, M_r/M_s = |cos(ψ)| where ψ = 0° is the easy axis direction. Frisk et al. [30] showed that for both polycrystalline and amorphous Co-Fe-Zr films the experimental M_r/M_s versus angle

![Figure 1](link)

![Figure 2](link)

Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thickness (nm)</th>
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<tbody>
<tr>
<td>SCT (20)</td>
<td>4.6(1) 20.1(1) 4.1(1)</td>
</tr>
<tr>
<td>SCT (40)</td>
<td>4.6(2) 39.8(2) 4.1(1)</td>
</tr>
<tr>
<td>CAZ (40)</td>
<td>4.5(2) 40.2(4) 3.4(2)</td>
</tr>
<tr>
<td>SCT/CAZ/SCT (20/10/20)</td>
<td>4.9(1) 20.2(5)/9.5(8)/21.0(9) 3.8(1)</td>
</tr>
<tr>
<td>SCT/CAZ/SCT (20/20/20)</td>
<td>4.9(2) 19.4(3)/20.5(5)/20.3(5) 4.0(1)</td>
</tr>
<tr>
<td>SCT/CAZ/SCT (20/30/20)</td>
<td>4.7(6) 18.6(5)/30.1(6)/21.3(2) 4.7(2)</td>
</tr>
<tr>
<td>SCT/CAZ/SCT (20/40/20)</td>
<td>4.6(1) 19.0(4)/40.7(5)/21.0(6) 3.6(4)</td>
</tr>
<tr>
<td>SCT/CAZ/SCT (20/50/20)</td>
<td>4.6(1) 19.0(8)/51.0(1)/21.0(6) 4.5(3)</td>
</tr>
<tr>
<td>CAZ/SCT/CAZ (10/20/10)</td>
<td>4.5(1) 10.3(3)/21.0(5)/7.10.3(2) 4.1(1)</td>
</tr>
</tbody>
</table>
The coupling between the hard and soft phases appears to be stronger in the CAZ/SCT/CAZ (10 nm/20 nm/10 nm) trilayer, shown in Fig. 4(g), since the easy axis loop is square and saturates at lower fields compared to the SCT/CAZ/SCT (20/40/20) trilayers, while the hard axis loop is well approximated by a straight line. Selected hysteresis loops, measured with VSM, of the trilayers with equal amounts of Co$_{85}$(Al$_{70}$Zr$_{30}$)$_{15}$ and Sm$_{15}$Co$_{81}$Ti$_{7}$ are shown in Fig. 5. $M_s/M_i$ versus $H_i$ are plotted in the insets and indicate that the anisotropy is uniaxial, as for the 40 nm thick Co$_{85}$(Al$_{70}$Zr$_{30}$)$_{15}$ sample. The lines are fits to Eq. (1), yielding $\sigma_{\psi} = 0.22\degree$ for SCT/CAZ/SCT (20/40/20) and $\sigma_{\psi} = 0.13\degree$ for CAZ/SC/SCT (10/20/10). This indicates a more well-defined anisotropy easy axis for the thinner trilayer, which we relate to a stronger coupling between the hard and the soft phase in this sample, where in addition the hard phase is surrounded by the soft phase.

Since the sputtered layers are X-ray amorphous we do not expect the samples to exhibit any crystalline texture which could introduce anisotropy. As mentioned in the introduction, the origin of in-plane uniaxial anisotropy in amorphous soft magnetic layers is still not fully understood. In a recent investigation of amorphous Co$_{1-x}$(Al$_{70}$Zr$_{30}$)$_{15}$ thin films [21], anisotropy is suggested to arise from substrate effects, in agreement with previous studies on other systems [17, 18]. If an in-plane magnetic field is applied during growth, the Sm$_{15}$Co$_{81}$Ti$_{7}$ layers will also be affected [26–28]. It is interesting to note that in our sample series the magnetic anisotropy is only manifest in the presence of the soft phase, and that it increases with the soft layer thickness in the trilayers (Fig. 4). This indicates that any substrate effect causing the anisotropy has to be propagated through the 20 nm thick hard phase, which we know is isotropic from the single layer $M(H)$ measurements. Strain at the substrate-buffer interface can persist throughout the layers, but bond-orientational bulk anisotropy contributions are also possible [18]. Very careful and extensive experimental investigations are necessary to distinguish between possible mechanisms [17, 18].
We also investigated the Curie temperatures, \( T_c \), of the Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) films. For the 20 nm thick Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) film, \( T_c \) was determined to be 363 K, based on the minimum of the derivative from a field-cooled (FC) magnetisation measurement, with an applied field of 1.6 kA/m, shown in Fig. 6. The \( T_c \) of the thicker (40 nm) Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) film is above 400 K and could not be determined in an FC measurement. By extrapolating the results of Thórarinsdóttir et al. \cite{Thórarinsdóttir2019} we estimate \( T_c \) of amorphous Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\) to be about 830 K.

The thermal behaviour of the magnetisation in ferromagnets at low temperatures, where magnon excitations dominate the reduction of the saturation magnetisation \( M_s \) at a temperature \( T \), can be described using Bloch’s power law \cite{Bloch1932}:

\[
M_s(T) = M_0 \left( 1 - \frac{T}{T_c} \right)^{\frac{1}{2}},
\]

where \( M_0 \) is the magnetisation at 0 K. This relation is used to create a guiding line in Fig. 7, indicating the qualitative relationship between the thermal stabilities of the samples, by looking at the temperature dependence of the magnetisation compared to idealised Bloch law case. In Fig. 7, \( M_s/M_0 \) is plotted versus temperature for the 40 nm thick Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) and Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\) films as well as for the CAZ/SCT/CAZ (10/20/10) trilayer. It appears that the 40 nm Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) film has a significantly lower thermal stability of the saturation magnetisation than the two other samples.

### 4. Conclusions

Magnetic trilayer samples are studied, consisting of layers of magnetically hard Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) and magnetically soft Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\). Saturation magnetisation is found to scale linearly with the volume fraction of Sm\(_{12}\)Co\(_{81}\)Ti\(_7\). Uniaxial in-plane anisotropy is observed, from the angular dependence of remanence, in a single-layer sample of Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\) and in trilayers with both phases, but not in single-layer Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) films. The uniaxial in-plane anisotropy inherent in the Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\) layer(s) persists in the trilayers with Sm\(_{12}\)Co\(_{81}\)Ti\(_7\), also with the hard/soft/hard layer configuration, and is more well defined for trilayers with thicker soft layers and if the hard layer is sandwiched in between two soft layers (soft/hard/soft configuration). Further investigation of the origins of this uniaxial anisotropy would be of interest. The coercivities of the trilayers are significantly reduced compared to the single layer of Sm\(_{12}\)Co\(_{81}\)Ti\(_7\), but enhanced compared to Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\). The Curie temperature, \( T_c \), of Sm\(_{12}\)Co\(_{81}\)Ti\(_7\) depends on the layer thickness and is much lower than that of Co\(_{85}\)(Al\(_{70}\)Zr\(_{30}\))\(_{15}\).
CRediT authorship contribution statement

J. Löfstrand: Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization. Parul Rani: Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization. Petra E. Jönsson: Formal analysis, Investigation, Writing – review & editing, Supervision, Funding acquisition. Gabriella Andersson: Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

Financial support by the Swedish Research Council (project grant 2017-03725) and the Swedish Energy Agency, Sweden (research project grant P48716-1) is gratefully acknowledged.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.jmmm.2023.171186.

References