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Author(s)	Masood, Ansar; McCloskey, Paul; Ó Mathúna, S. Cian; Kulkarni, Santosh
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Co-based amorphous thin films on silicon with soft magnetic properties

Ansar Masood,^{1,a} P. McCloskey,¹ Cian Ó. Mathúna,^{1,2} and S. Kulkarni¹

¹Microsystems Centre, Tyndall National Institute, University College Cork, Cork, Ireland

²School of Electrical and Electronic Engineering, University College Cork, Cork, Ireland

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The present work investigates the emergence of multiple modes in the high-frequency permeability spectrum of Co-Zr-Ta-B amorphous thin films. Amorphous thin films of different thicknesses ($t=100\text{-}530$ nm) were deposited by DC magnetron sputtering. Their static and dynamic soft magnetic properties were investigated to explore the presence of multi-magnetic phases in the films. A two-phase magnetic behavior of the thicker films (≥ 333 nm) was revealed by the in-plane hysteresis loops. Multiple resonance peaks were observed in the high-frequency permeability spectrum of the thicker films. The thickness dependent multiple resonance peaks below the main ferromagnetic resonance (FMR) can be attributed to the two-phase magnetic behaviors of the films. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5007733>

I. INTRODUCTION

Soft magnetic amorphous thin films retaining the in-plane uniaxial magnetic anisotropy, high permeability, and high ferromagnetic resonance frequency are of greater importance for technological applications such as magnetic passives,¹ digital recording media,² and energy transferring devices.¹ These devices need to integrate the soft magnetic thin films with a certain thickness. When the thickness of amorphous films is greater than a critical limit, it has been found challenging to retain the adequate magnetic softness³ and in-plane uniaxial magnetic anisotropy.⁴ The in-plane uniaxial magnetic anisotropic field (H_k) determines the permeability (μ) and ferromagnetic resonance frequency (f_{FMR}) of the films, provided that the saturation magnetization (M_s) is kept constant for a particular material.⁵ To better control the high-frequency permeability response of the amorphous thin films, homogeneous in-plane uniaxial magnetic anisotropy in thin amorphous thin films is required.

A thorough understanding of underlying mechanisms defining the frequency dependence of permeability of the ferromagnetic material is indispensable for an optimal tuning of ferromagnetic resonance behavior.⁶ The ferromagnetic resonance (FMR) occurs when electron spins absorb energy from an oscillating magnetic field at a certain frequency.⁷ A typical single peak corresponding to FMR is commonly observed in the high-frequency permeability spectrum of ferromagnetic materials and is well anticipated by Landau-Lifshitz-Gilbert equation.⁸ Nevertheless, multiple resonance peaks have been observed in the permeability spectrum of Co-Nb-Zr⁹ and Co-Fe-Si-B¹⁰ amorphous thin films, rather than single FMR. The emergence of additional peaks below the FMR is considered a detrimental factor for the soft ferromagnetic thin film materials as the hysteresis power loss, which is closely related to the imaginary permeability, becomes significantly large even well below the FMR frequencies.⁸ Different reasons have been put forth to explain the emergence of these additional peaks in the permeability spectrum.⁸⁻¹⁰ Evidence suggested that these multiple peaks are mainly attributed to the stripe domains,⁹ which in turn emerged due to perpendicular anisotropy,¹¹ and

^aElectronic mail: ansar.masood@tyndall.ie

spike-like magnetic domain structure owing to sample-edge effect.^{10,12} On the other hand, Lee *et al.*⁸ suggested that the multiple resonance peaks could be due to the magnetostatic modes or spin-wave modes. However, further analysis was suggested to explain the possible dependence of peak frequencies on film thickness. Therefore, this remains as a potential area to unveil the mechanisms responsible for the frequency dependence of permeability of ferromagnetic material to prevent their detrimental effect on material losses. In this work, we propose the emergence of multiple resonance peaks is due to the existence of two-phase magnetic behavior in Co-based thin film amorphous materials.

II. EXPERIMENTAL METHODS

Amorphous thin films of $\text{Co}_{84}\text{Zr}_4\text{Ta}_4\text{B}_8$ (atomic %) alloy were deposited by using magnetron sputtering (Nordiko) technique from an 8-inch single alloy target. The sputtering chamber was pumped down to a base pressure of 10^{-6} Pa before each deposition and a high purity argon gas was introduced to obtain a sputtering pressure of 0.13 Pa to deposit the films. The DC power of 500 W was applied to the target material. The deposition process of films was carried out in the presence of a magnetic field. The deposition rate of the films was confirmed as 11.1 nm/min. Thin films of 100-530 nm thickness were deposited on 4-inch silicon wafers at room temperature. An adhesive layer of 20 nm thickness of Ti was deposited prior to the deposition of the magnetic film. The Dektak surface profilometer was used to determine the thickness of the films. The atomic structure of the films was investigated by X-ray diffraction (XRD, Phillips Xpert diffractometer, $\text{Cu K}\alpha$ 1.54 Å) and transmission electron microscopy (TEM) techniques. The magnetic properties of films were investigated by using B-H loop tracer (SHB, MESA 200 HF). High-frequency permeameter (Ryowa) was used to measure the permeability of 3 mm x 3 mm samples.

III. RESULTS AND DISCUSSION

Figure 1 represents the XRD pattern and high-resolution TEM image of the Co-Zr-Ta-B thin films. The XRD pattern of the films (100-530 nm) revealed broad maxima in the range of $2\theta=40\text{-}50^\circ$ [Fig. 1(a)]. No Bragg's peak was observed in the whole XRD spectrum of the films. In addition, the local disorder atomic structure of the 230 nm thin film was investigated by the high-resolution TEM image and selected area electron diffraction (SAED) patterns [Fig. 1(b)]. The diffused hollow ring in the SAED patterns confirmed the amorphous nature of the films. From the diffraction patterns of XRD and TEM, it can be concluded that there was no sign of crystallite precipitation during the deposition process of films and the atomic structure of the film was amorphous.

The in-plane uniaxial magnetic anisotropy was induced by applying in-plane magnetic field during the deposition process of the films. Figure 2 represents the hysteretic loops along the easy and hard magnetic anisotropy axis of 100-530 nm thin films. Different magnetic properties of the

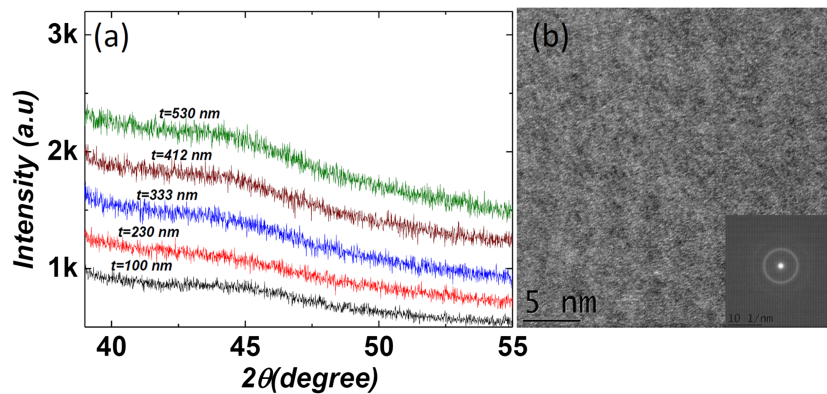


FIG. 1. (a) X-ray diffraction pattern of 100 to 530 nm thin films. (b) High-resolution TEM image of the 230 nm film. The inset represents the selected area electron diffraction pattern (SAED) of the same film.

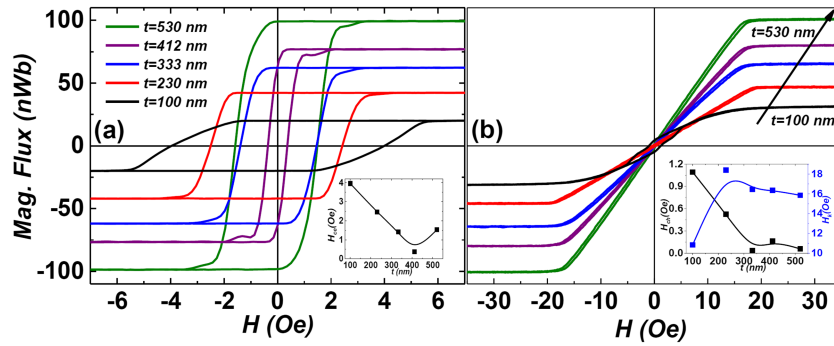


FIG. 2. (a) In-plane hysteresis loops of as-deposited films of thickness (t) ranging from 100 to 530 nm along the easy anisotropy axis. The inset represents the easy-axis coercivity (H_{ce}) as a function of film thickness. (b) The in-plane hysteresis loops of as-deposited films ($t=100$ -530 nm) along with hard anisotropy axis. The inset represents the thickness dependent hard-axis coercivity (H_{ch}) and anisotropic field (H_k).

films, like coercivity and anisotropic magnetic field (H_k), were measured by using these hysteresis loops. The easy-axis coercivity (H_{ce}) of the films was found to be thickness dependent [see inset Fig. 2(a)] and decreased to a minimum value of 0.37 Oe for 412 nm thin films, it was 10 fold smaller as compared to 100 nm film (i.e. 3.97 Oe). It is noteworthy to mention that the unusual shape of the in-plane hysteresis loops appeared when the film thickness increased from 230 nm. A kink before the saturation magnetization of the films started to appear when the thickness of the films is 333 nm. This unusual behavior was found prominent in thicker films of 412 and 530 nm. The Fig. 2(b) represents the hard-axis magnetic hysteresis loops, while the inset of Fig. 2(b) shows the thickness dependence of hard-axis coercivity (H_{ch}) and H_k values of the films. The broad hysteresis loops ($H_{ch}=1$ Oe) and low H_k (i.e. 10.84 Oe) values were found for 100 nm thin films. Further, the H_{ch} decreased exponentially as a function of film thickness and ultra-low values of H_{ch} (<0.1 Oe) were obtained for thicker ($t \geq 333$ nm) films. The large in-plane uniaxial magnetic anisotropy was induced in thicker films (≥ 230 nm) [see inset Fig. 1(b)].

The coercivity and shape of the hysteresis loops of amorphous thin film materials are affected by the residual stress¹³ and the presence of heterogeneity in the alloy.¹⁴ For example, Fe-Cu-Nb-Si-B,³ Co-Fe-Ta-B,¹⁴ Fe-B-Si,¹¹ and Fe-Ni-B-Nb¹³ amorphous films revealed spin-reorientation transition¹⁴ where the magnetization vector transformed from in-plane to out-of-plane as a function of film thickness. The large residual stress and atomic randomness in amorphous thin films were proposed as reasons to invoke the large coercivity and perpendicular magnetization in amorphous thin films.^{3,13,14} In the present work, the thickness dependent magnetic properties can also be attributed to the large residual stress and heterogeneous composition produced during the deposition process of the thin films. Particularly, thinner ($t=100$ -230 nm) amorphous films are considered to contain large residual stress due to the interface effect between film and substrate resulting in higher coercivity. Further, the interface stress effect decreases with increasing film thicknesses resulting lower coercivity as observed in Fig. 2. The two-slope magnetic hysteresis loops of thicker films suggest the presence of two different phases of heterogeneous amorphous magnetic films. The two-phase amorphous structure may have originated due to the plasma heating effect during the deposition process of the Co-Zr-Ta-B thin films where small regions of the films may have been oxidized.

Figure 3 represents the high-frequency permeability spectrum of 100 to 530 nm Co-Zr-Ta-B thin films. All investigated films revealed high permeability (>800) and ferromagnetic resonance frequency ($f_{FMR} > 1$ GHz). The impact of low H_k of 100 nm film is resulted in the high permeability as compared to thicker films. The high-frequency permeability spectrum of 100-230 nm films revealed one main peak corresponding to the ferromagnetic resonance (FMR), typically expected for the soft magnetic thin film materials and well predicted by the Landau-Lifshitz-Gilbert (LLG) equation. However, the unusual additional peaks were observed below the FMR in the permeability spectrum from 333 to 530 nm thin films. The multiple resonance peaks are clearly visible in both real and imaginary permeability spectrum (see Fig. 3). The additional peaks were monotonically increased

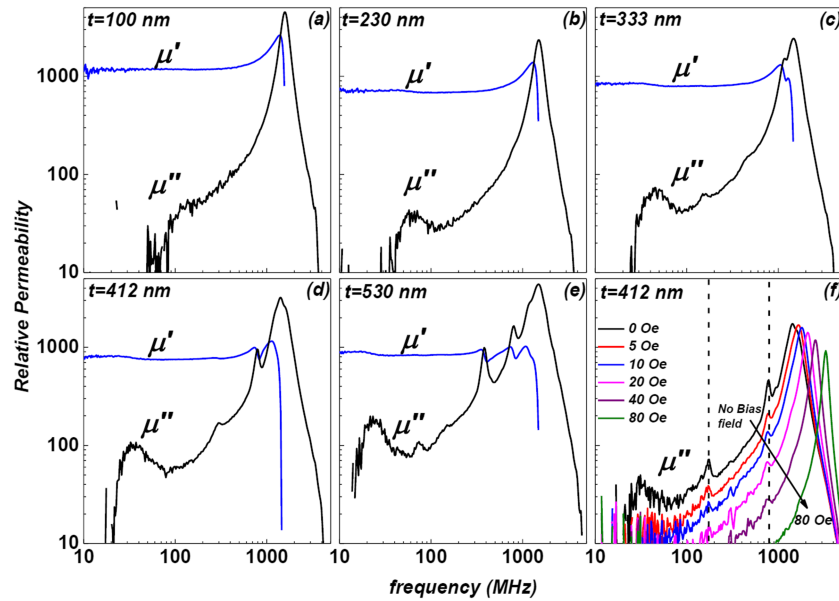


FIG. 3. (a) Permeability spectra of Co-Zr-Ta-B thin films with different thickness (t): (a) 100 nm, (b) 230 nm, (c) 333 nm, (d) 412 nm, (e) 530 nm, and (f) imaginary permeability (μ'') of the 412 nm thin film at different bias fields (0-80 Oe) applied along the easy magnetic anisotropy axis of the films.

in number on the increment of film thickness. The emergence of additional peaks below the FMR is considered a detrimental factor for the soft ferromagnetic thin film materials as the hysteresis power loss, which is closely related to the imaginary permeability, becomes significantly large even well below the FMR frequencies. The high-frequency permeability spectrum was measured by applying different bias fields along the in-plane easy axis of the samples. A typical imaginary permeability spectra of the 412 nm film with applied bias fields ranging from 5 to 80 Oe is presented in Fig. 3 (f). The imaginary permeability spectrum of the 412 nm film without applying any bias field is also presented for the comparison. The effective increase in H_k , originated due to the applied bias fields, was reflected in the main FMR peak which shifted to the higher frequencies. The number of additional peaks below the FMR and peak frequency was remained unaffected by increasing the bias field.

The various mechanisms have been proposed to explain the possible source for multiple resonance peaks in the permeability spectrum of soft magnetic thin films.^{8-10,12} As for example, the formation of the stripe magnetic domains originated by perpendicular anisotropy⁹ and the needle-shaped magnetic domain structure near the sample boundaries^{10,12} have been reported to originate this kind of behavior. The stripe magnetic domain structure along with perpendicular anisotropy component has been observed in several ferromagnetic amorphous thin films.^{11,14} The films with perpendicular anisotropy reveal the two slopes “transcritical” type hysteresis loops and are well understood.^{11,14} In the present investigations, the hysteresis loops of the films were found “square-type” [see Fig. 2 (a)], instead of “transcritical-type”, with in-plane uniaxial magnetic anisotropy. The formation of stripe magnetic domains in the investigated thin films is not possible, hence this type of domains cannot be the source of multiple resonance peaks in high-frequency permeability spectra of thicker Co-Zr-Ta-B films. Other reports of the multiple resonance peaks are explained through the presence of the spike-type magnetic domain structure near the sample boundaries,^{10,12} possibly originated due to the shape effect of the sample. This type of magnetic domains was suggested to be a possible source of multiple resonance peaks in Co-Fe-Si-B¹⁰ and Co-Nb-Zr⁹ amorphous thin films. The magnetic domains can be wiped out by applying the bias field during the permeability measurement.⁸ In our case, the additional resonance peaks remained unaffected after applying the bias field, which suggest that the origin of the multiple resonance peaks were probably not associated with any type of magnetic domain structure which could be possibly depleted out by applying the bias field higher than the anisotropic field

of the film (i.e. $H_k=17$ Oe). This suggests that the source of additional peaks in the thicker films is not associated with the spike-domain structure.

The observed additional peaks in the permeability spectrum of the thicker films are related to the two-phase magnetic behavior of the amorphous films. The high-permeability spectrum of thinner films (100 to 230 nm) revealed single FMR which is usually observed in soft-magnetic materials. The thickness dependent multiple resonance peaks appeared from 333 to 530 nm, at the same time the two-phase hysteresis loops were also revealed for the same films. This suggests that the origin of multiple resonance peaks in the high-frequency permeability spectrum is associated with the two-phase magnetic behavior of the thicker films. As explained earlier, the two-phase magnetic behavior of the thicker films may be generated due to the inhomogeneity produced in amorphous composition as a result of the plasma heating effect during the deposition process of the thicker films. Further studies may be required to find out the origin of the heterogenous amorphous structure of the Co-Zr-Ta-B thin films.

IV. CONCLUSION

The amorphous thin films of 100-530 nm thickness were magnetron sputtered from a single alloy target of Co-Zr-Ta-B, and their static and magnetic properties were investigated. The in-plane hysteresis loops revealed the two-phase magnetic behavior of the amorphous films when the thickness of the films increased from 333 nm. The coercivity and uniaxial magnetic anisotropy induced in amorphous films during the deposition process show thickness dependence and attributed to the residual stress quenched at the film-substrate interface. High-frequency permeability study revealed that thin films can retain the high permeability (>800) and high resonance frequency (> 1 GHz) in the as-deposited state of the films. Thickness-dependent multiple resonance peaks below the ferromagnetic resonance (FMR) were revealed that were attributed to the two-phase magnetic behavior of the thicker films.

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