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Controlling the competing magnetic anisotropy energies in FineMET amorphous thin films with ultra-soft magnetic properties

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Thickness dependent competing magnetic anisotropy energies were investigated to explore the global magnetic behaviours of FineMET amorphous thin films. A dominant perpendicular magnetization component in the as-deposited state of thinner films was observed due to high magnetoelastic anisotropy energy which arises from stresses induced at the substrate-film interface. This perpendicular magnetization component decreases with increasing film thickness. Thermal annealing at elevated temperature revealed a significant influence on the magnetization state of the FineMET thin films and controlled annealing steps leads to ultra-soft magnetic properties, making these thin films alloys ideal for a wide range of applications. [http://dx.doi.org/10.1063/1.4984235]

I. INTRODUCTION

Magnetic anisotropy is a fundamental physical quantity responsible for defining an axis of magnetization in the magnetic materials.[1] It plays a significantly different role in integrated soft magnetic thin films in comparison to the bulk magnetic material.[1] In thin film magnetic materials, a large interface or surface anisotropy can create a dominant perpendicular magnetic anisotropy (PMA) component and as a consequence aligns the magnetic spins perpendicular to the plane of the films.[2–4] The presence of PMA component significantly undermines the functionality of these thin film materials particularly for efficient energy storage applications such as magnetic cores for inductors and transformers.[5]

In soft magnetic thin film materials, the magnetic orientation depends on several additional physical factors, including thickness and shape etc.[2–4,6,7] For example, when the thickness of the film increases from few monolayers to sub-micron regime, the orientation of magnetic spins transform from out-of-plane to the in-plane configuration.[3,4,6–8] This phenomenon typically defined as thickness dependent Spin Reorientation Transition (SRT),[2,3] has been observed in several thin film crystalline materials, i.e. Fe/Cu(100),[9] Fe/Ag(100),[10] Fe/Mo(110),[11] Co/Au(111),[12] Ni/Cu(001).[13] In these systems, the emergence of SRT was attributed to different factors such as film-substrate interface effects, change in magnetic anisotropy due to structural evolution, or variation in the orbital magnetic moment due to broken symmetry of the film-substrate interface.[9–14] Recently, SRT behaviour has also been observed in amorphous and nanocrystalline thin films.[2–4,6,7,15] As the use of amorphous magnetic materials in different functional devices becomes more ubiquitous, it is important to explore the emergence of perpendicular magnetic anisotropy and approaches to minimize the PMA component to realize the ultra-soft magnetic properties in thin film materials.[16,17]

Excellent soft magnetic properties, i.e. low coercivity, along with high electrical resistivity, high permeability and controlled magnetic anisotropy are the prerequisites of amorphous magnetic thin

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films for their use in a range of applications including high-frequency power conversion devices, magnetic shielding, and current sensors etc.[17–19] However, the emergence of perpendicular magnetic anisotropy along with stripe magnetic domains in these thin film materials deteriorate the soft magnetic properties, in particular, coercivity and permeability.[5,17,20] These issues limit the possibility of utilizing such high flux density thin films in efficient energy transferring devices.[5,17] In the present work, we explain the emergence of thickness dependent perpendicular magnetic anisotropy in high flux density thin film amorphous materials. Further, this study explains the benefits of using the annealing process to overcome PMA to realize the ultra-soft magnetic properties of the amorphous thin film. This work establishes that the PMA component in the magnetization reversal process can be minimized to the same extent by increased film thickness and through post processing techniques such as thermal annealing for stress relieving. The FineMET thin film material have potential to form a key element of future highly miniaturized and integrated power conversion circuits due to their ultra-low coercivity (2.4 A/m), high resistivity (145 μΩ.cm) and high magnetic saturation (1.4 T). These magnetic properties are superior to the existing thin film soft magnetic materials such as Permalloy (crystalline) and Co-based amorphous materials which have lower resistivity and magnetic saturation along with higher coercivity.[17]

II. EXPERIMENTAL METHODS

Amorphous films of FineMET ($Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$, atomic %) alloy with a thickness of 40-517 nm, were deposited using DC magnetron sputtering from a single alloy target. The sputtering chamber was pumped down to a base pressure of 10^{-6} Pa and a high purity argon gas was introduced to obtain a sputtering pressure of 0.13 Pa. The films were deposited on 4 inch disk shape Si wafers at room temperature. Prior to the deposition of the magnetic films, an adhesive layer of Ti of 20 nm was deposited. The DC power to the target was fixed at 100 W. Dektak surface profilometer was used to determine the thickness of the films. The structure of the films was investigated using x-ray diffraction (XRD) method (step size=0.003°, scan speed = 500 sec/step) by using Phillips Xpert diffractometer with Cu K_α(1.54 Å) radiations. From the XRD patterns, the broad maxima at 2θ = 40-50° and absence of sharp crystalline peaks confirms the amorphous atomic structure of the films. Further, the films were annealed for 60 minutes at 300 °C. The annealing experiments were performed in an inert atmosphere. The Argon gas was kept on constant flow during the whole series of experiments which reduces any possibility of oxidation process. Transmission electron microscopy (TEM) was utilized to further confirm the random atomic structure of the annealed films. In-plane hysteresis loops of the films (4 inch wafer) were investigated using an SHB (MESA 200 HF) B-H loop tracer.

III. RESULTS AND DISCUSSION

In order to understand the effect of film thickness on the competing magnetic anisotropy energies, in-plane magnetic hysteresis loops of as-deposited films (40-517 nm) were performed and presented in Fig. 1. The effect of competing magnetic anisotropy energies on the state of the magnetization as a function of film thickness is clearly illustrated by the shape of the hysteresis loops (Fig. 1). The shape of the hysteresis loop of 40-316 nm films is not a square type. This particular type of hysteresis loop is known as "transcritical loop"[3,21] and widely recognized as a consequence of the perpendicular state of magnetization in thin film materials.[22] Another interesting observation from Fig. 1 is that the coercivity (H_c) and anisotropy field (H_k) vary as a function of film thickness. Increasing film thickness of FineMET material from 40 to100 nm increased the H_c (279-358 A/m) and H_k (780-1751 A/m). Further, as film thickness increased beyond 100 nm, the values of H_c and H_k were reduced continuously to 64 A/m and 127 A/m for 517 nm thick film, respectively. This improvement in magnetic properties is clearly linked to the transformation from transcritical hysteresis loops to the square type representing the in-plane soft magnetic behaviour of thicker films. This dynamic behaviour of hysteresis loop suggests that competing magnetic anisotropy energies were strongly influenced by the film thickness, and as a consequence, state of the magnetization was transformed

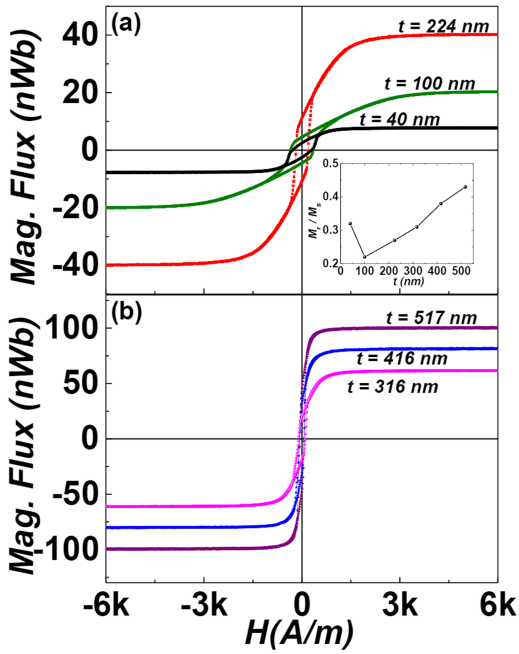

FIG. 1. (a) In-plane hysteresis loops of 40-224 nm films in the as-deposited state. Inset (a) represents the Mr/Ms ration of the films as a function of film thickness. (b) B-H loops of 316-517 nm amorphous films in the as-deposited state.

from perpendicular to the in-plane configuration by giving rise to the SRT as a function of film thickness.

Understanding this dynamic behaviour requires a careful analysis of competing magnetic anisotropy energies in thin films. The anisotropy field, H_k is directly connected to the perpendicular anisotropy energy arising from magnetoelastic effect, $K_\perp$, which can be calculated from the following expression.[6]

$$K_\perp = \frac{u_o M_s H_k}{2} \tag{1}$$

where $\mu_o M_s$ is the saturation magnetization of the material. The in-plane magnetoelastic anisotropy energy, K_d, of the films can be expressed as $K_d = \frac{u_o M_s^2}{2}$.[6] It is well known that the development of transcritical loops and perpendicular magnetization is associated with the Q-factor, $K_\perp/K_d$,[6] and found to appear spontaneously when Q > 1. For the present series of films, the $K_\perp$, K_d and Q-factor were calculated ($\mu_o M_s$ of FineMET = 1.4 T) and summarized in Table I. However, the Q-factor was found significantly lower than 1 for the present series of samples. The emergence of the PMA component in the magnetization as a function of film thickness could not be explained by the Q-factor based model. The role of different magnetic anisotropy energies, particularly magneto-elastic and shape (for amorphous thin films) at different film thicknesses were found more complex in the FineMET thin films and requires further investigations to understand the SRT phenomenon.

TABLE I. Anisotropic field (H_k), perpendicular anisotropic energy ($K_\perp$) and Q-factor of different films.

t (nm)	H_k (A/m)	$K_\perp$ (kJ/m^3)	$Q=K_\perp/K_d$ (x 10^{-3})
40	780	0.546	0.70
100	1751	1.226	1.38
224	780	0.546	0.70
316	366	0.256	0.33
416	175	0.123	0.16
517	127	0.889	0.11

As mentioned earlier, the thickness dependent spin reorientation transition (SRT) has been widely reported in many systems.[2–4,6,8–15] For example, thin films of Fe, Co, and Ni systems were reported to exhibit SRT where the interface between the film and substrate played a crucial role in the emergence of PMA.[9–14] In addition to these crystalline thin film materials, perpendicular magnetization has been observed in amorphous and nanocrystalline thin films.[3,6,23] The orientation of magnetization in thin film materials can be determined by analyzing competing magnetic anisotropy energies such as magnetoelastic, magnetocrystalline, shape anisotropy etc.[3] Due to the unique disorder atomic structure, the magnetocrystalline anisotropy is absent in amorphous materials. In absence of magnetocrystalline anisotropy, the magnetoelastic and shape anisotropy energies play a significant role in the orientation of magnetization in these materials.[3,6] The interplay between mechanical stress from deposition process and magnetization define the magnetostriction constant, which is responsible for the magnetoelastic energies in thin films.[3,6] The rapid cooling process of amorphous thin films during the deposition process and atomic randomness play a vital role in inducing the localized stress at the substrate/film interface.[2] Due to the high magnetostriction constant (20 x 10^{-6}) of the amorphous phase of FineMET alloy, the magnetoelastic energy plays an important role in defining the state of magnetization, particularly at low film thickness.[6] This magnetoelastic energy is reduced with increasing film thickness, as the increased thickness minimizes the initial substrate/film interface stress. Further, at certain thickness the magnetoelastic energies become similar to the shape energy contribution and as a consequence magnetic spins transform from out-of-plane to the in-plane configuration.

In order to investigate the effect of stress on the perpendicular magnetization in FineMET films, films (100-517 nm) were annealed at 300 °C for 60 minutes in an inert atmosphere and its effect on structural and magnetic properties was investigated. The XRD spectrum of the as-deposited and the annealed film of thickness 100 nm is presented in the Fig. 2 (a). The absence of any Bragg's peak and presence of broad maxima in the XRD spectrum revealed the existence of amorphous atomic

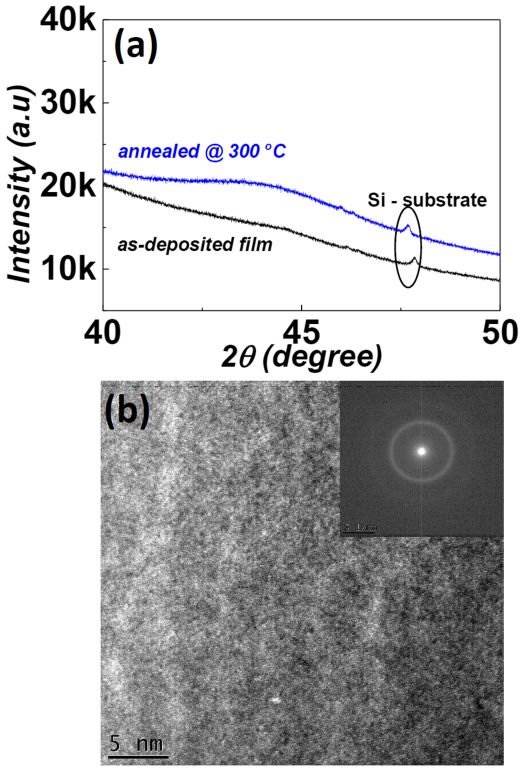

FIG. 2. (a) The X-ray diffraction (XRD) pattern of the 100 nm thin film in as-deposited state and after annealing at 300 °C for 60 min. (b) High resolution TEM image of the film annealed at 300 °C for 60 min. inset (b) represents the selected area electron diffraction (SAED) patterns of the same film.

structure of the films. In addition, high resolution TEM image and selected area electron diffraction (SAED) pattern of annealed film, presented in Fig. 2 (b), confirmed that there was no precipitation of the crystallites at the local scale and films contained amorphous atomic structure after annealing. This confirms that the annealing process only relaxed the atomic structure of the films without precipitating any crystalline phase of the material.

To investigate the effects of annealing on the magnetic properties, the in-plane hysteresis loops were measured using BH loop tracer and plots are presented in Fig. 3. It is clear from the figure that after annealing the transcritical shape of the loops disappeared into simple hysteresis along with very small coercivity (<8 A/m) for all films confirming the typical behaviour of soft magnetic amorphous materials. Particularly, the minimum coercivity was found 2.4 A/m, which is ideal for high-frequency power applications. Similar to the film thickness impact on magnetic behaviour, a detailed understanding of the competing anisotropy energies is required to explain this result.

Annealing of amorphous materials at elevated temperatures relaxes the atomic structure by removing the stress produced at the substrate/film interface or it changes the overall structure of the material by precipitating crystalline phases.[19] In the present case, the amorphous phase of the FineMET alloy was not crystallized, as confirmed by XRD and TEM analysis. This confirms that the thermal annealing process releases the stress/strain produced at the substrate/film interface, by reducing the magnetoelastic contribution. This along with the compactness of the random atomic structure through the relaxation process improves the soft magnetic properties of FineMET amorphous thin films.[19,20]

Further, it is interesting to note that the magnetization reversal process for annealed FineMET films with lower thicknesses (100-224 nm) begins earlier than in non-annealed films. The anisotropy field which is a measure of the reversal process is lower in the case of the annealed films. As explained in earlier sections, the substrate-film interface stress plays a dominant role in defining the magnetoelastic energy and this stress is more prominent at lower film thicknesses (Fig 1). As annealing is a route to release this stress, the impact of annealing is also more prominent for these film thicknesses. Hence, these thinner films (100-224 nm) experience an earlier reversal process compared to non-annealed films.

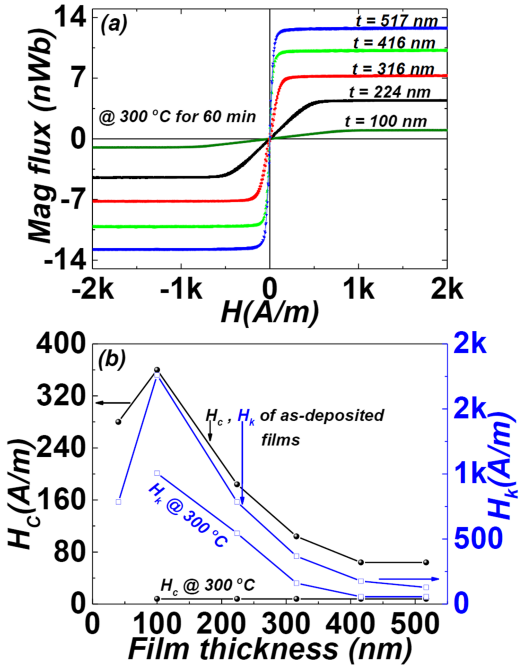

FIG. 3. (a) In-plane hysteresis loops of the films annealed at 300 °C for an hour. (b) The effect of annealing on coercivity (H_c) and anisotropy field (H_k) as a function of film thickness. The H_c and H_k values of the as-deposited films are also presented for comparison.

IV. CONCLUSIONS

To conclude, this work explains the global magnetic behaviours of amorphous FineMET thin films by stress/strain produced during the fabrication process, and finally, it focuses on the optimization of ultra-soft magnetic properties by a thermal annealing process. It reports that in FineMET thin films the magnetoelastic energy dominates and the magnetic spins configure them along out-of-plane configuration. The state of magnetization aligns into the in-plane configuration as a function of film thickness and shape anisotropy energy overcomes the magnetoelastic energy. Further, thermal annealing leads to a transition from transcritical loops to in-plane hysteresis loops by developing ultra-soft magnetic behaviours which could be attributed to the structural relaxation of the FineMET thin films. The ultra-soft FineMET amorphous thin films can be considered as a potential candidate to form a key element of future highly miniaturized integrated power conversion circuits due to their ultra-low coercivity (2.4 A/m), high resistivity (145 μΩ.cm) and high saturation magnetization (1.4 T).

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