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Enhancement of magnetostrictive properties of Galfenol thin films

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ABSTRACT

The present study investigates the role of substrate temperatures on the structural, morphological, magnetic and magnetostrictive properties of DC sputtered FeGa thin films grown on Si substrates. These films were deposited at various substrate temperatures between 50 and 350 °C. The structural characterization of the films revealed columnar growth and the transformation of surface morphology from prismatic to spherical at high substrate temperatures. Both $L1_2$ and B2 phases of FeGa existed in the films, with the $L1_2$ phase dominating. The in-plane and out-of-plane vibration sample magnetometry measurements showed the evolution of magnetic anisotropy in these films. It was revealed from the magnetostriction measurements that the films deposited at 250 °C exhibited the maximum value of 59 ppm.

1. Introduction

Functional magnetic materials with magnetostrictive properties have been investigated in recent years owing to their wide range of applications as in sonar systems [1,2], micro-electromechanical systems in the form of transducers and actuators [3,4], sensors [5], vibrational energy harvesting devices and vibration control systems [6,7] etc. Major advantages of these magnetostrictive materials include noncontact operation, high reliability, simple designs and compatibility with semiconductor manufacturing processes thus enabling integration in microelectronic technologies.

Magnetostrictive material research interest resurged in the 1970's with many groups working on single and multilayer thin films [8]. The discovery of Terfenol-D, a rare-earth Fe based alloy attracted much attention in this field of research with its high magnetostriction values of ~2000 ppm in bulk and of nearly 1000 ppm in polycrystalline thin film form [9]. Though it had high magnetostrictive properties, its large magnetocrystalline anisotropy resulting in high saturation fields, limits its use in practical applications. Other constraining factors of this material are the presence of rare-earths and high cost. Alternate materials were considered as a replacement for Terfenol-D which exhibits sufficient magnetostriction. In this pursuance, Fe-based alloys including Fe-Al, Fe-Co, and Fe-Ga showing extraordinary magnetostrictive behaviours were considered [10–12]. Among these, FeGa alloys are promising due to several attractive properties including low cost, corrosion resistance, machinable and with moderate magnetostriction (~400 ppm) [9] at lower saturation fields.

Several groups have been working on FeGa thin films and more focus is being given to improve the magnetostriction. Javed et al., have studied the influence of Ga evaporation rate, power density for Fe sputtering and Ar partial pressure on the film growth and obtained effective magnetostriction constant [13] of 80 ppm at maximum. In-plane anisotropy was induced during sputtering process by an applied magnetic field and the influence of sputter power was studied by Wang et.al. [14]. They found that anisotropy field decreases with the increasing sputter power and obtained a magnetostriction of 66 ppm in the films deposited at 60 W. The effects of Boron addition to the FeGaB films were investigated by Lou et al. [15] and a structural transformation from polycrystalline to amorphous with enhanced soft magnetic and microwave properties along with a high magnetostriction constant of

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70 ppm at a B content of 12 at.% was observed. Basumatary et al. [16] investigated the effect of substrate temperature on FeGa films and observed a maximum magnetostriction of 200 μ-strains for the film deposited at 450 °C. Eliot et al. [17] showed that Galfenol can be deposited by electrodeposition and measured a magnetostriction value of 140 ppm. However, the influence of substrate temperature on the magnetostriction and its concomitant effects on the magnetostriction has to be understood further to ensure the advances in real time applications. Therefore, in this study, we investigate the effect of substrate temperature on the morphological, structural and magnetic properties of FeGa sputtered thin films and how the magnetostriction changes with respect to substrate temperature.

2. Materials and methods

2.1. Material synthesis

A two-inch diameter Galfenol ($Fe_{73}Ga_{27}$) target with a purity of 99.99% (from Able target Inc.) was used to deposit FeGa thin films on Si substrates by DC magnetron sputtering technique. The substrates were cleaned and native oxide layer was removed using HF before the sputter deposition. The sputtering chamber was initially pumped down to a base pressure of 2×10^{-5} mbar. Argon was used as a sputtering gas and the pressure was maintained at 4×10^{-2} mbar. The substrate to target distance was maintained at ~5 cm and the substrate temperature was either 50 °C, 150 °C, 250 °C or 350 °C. The deposition process was carried out at a target power of 80 W and the total time of deposition was 30 min. After the deposition, chamber was purged with Ar for ~30 min. These films were analyzed using different characterization methods as described in the next section.

2.2. Material characterization

The thickness of the thin films was about 500 ± 30 nm and the films were not subjected to any post-deposition treatment. The as prepared films were analyzed by Glancing Incidence X-ray Diffraction (GI-XRD) technique with Cu-Kα radiation to study the crystal and phase structure. A scanning electron microscope (SEM) with a field-emission electron source (Supra 55 by Carl Zeiss) was used to study the microstructure of the films. Both the surface morphology (top view) and fracture cross-section analysis of the films were performed. Energy Dispersive X-ray Spectroscopy (EDS) was employed to analyze the composition of the films. In-plane and out-of-plane room temperature magnetic measurements were carried out using a Vibration Sample Magnetometer (VSM). The topology and roughness of the films were measured by Atomic force microscope (AFM). Magnetostriction of the samples was measured by a custom built magnetostriction measurement system [18].

3. Results and discussions

3.1. Morphological and structural analysis

Fig. 1(a–d) show the cross-sectional image of films grown at 50 °C, 150 °C, 250 °C and 350 °C, respectively. High magnification top view images are shown in the inset. At lower substrate temperatures of 50 °C, thin columns consisting of smaller grains are observed. When the substrate temperature rises to 150 °C, the columnar diameter as well as spherical grain diameter increases. With further increase in substrate temperature to 250 °C, the columnar structure of the films is gradually evolved and separate columns are observed. This columnar morphology dissolves into planar structure towards the surface. At the higher substrate temperature of 350 °C, the columnar structure has collapsed and one gets random orientation in growth leading to planar growth. This loss of structural anisotropy results in the non-uniform growth of thin film with varying particle sizes and shapes in the prepared thin films.

Fig. S1(a–d) shows the top view images of the films grown at substrate temperatures of 50 °C, 150 °C, 250 °C and 350 °C. It is observed that there is a significant change in morphology with increase in substrate temperature. At 50 °C, the morphology of the film was prismatic in nature constituting of smaller particles (~8 nm). These smaller particulates cumulated to form the prismatic structures (~50 nm). This prismatic morphology was homogeneous over the entire film surface. As the substrate temperature increases to 150 °C, the regular prism-like shape of the particles is deteriorated. The films still consist of smaller particulates (~10 nm) and these particulates combines together to form the bigger (~40 nm) partly prismatic particles. A further change in morphology is observed when the substrate temperature was raised to 250 °C. The particles, at this condition, no longer retain the prism-like shape, but have become more or less spherical in shape. The size of the particle is widely distributed (average particle size ~75 nm) though the particles still maintain their innate nature of being constituted of smaller particulates (~25 nm) [19]. At 350 °C, the particles do not retain any certain shape and is flake - like with particle size varying to give an average of ~30 nm.

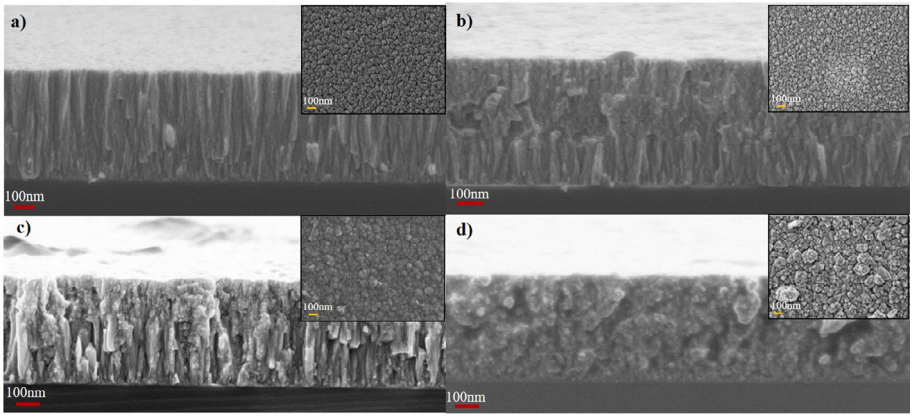

Fig. 1. The fracture cross-sectional SEM images showing the microstructure of the FeGa thin films grown at substrate temperature of (a) 50 °C, (b) 150 °C, (c) 250 °C and (d) 350 °C. The insets show the corresponding magnified plane-view images.

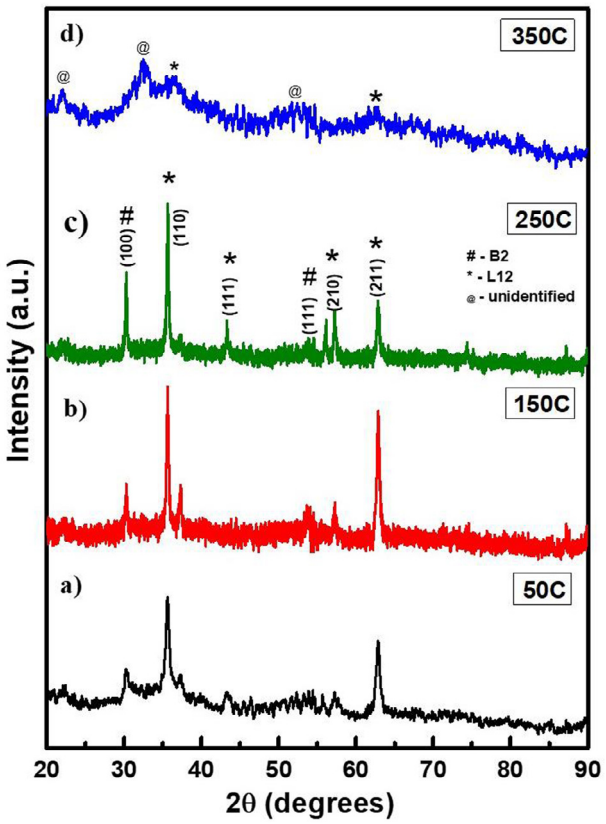

Fig. 2. (a–d). XRD pattern of FeGa thin films grown at temperatures of 50 °C, 150 °C, 250 °C and 350 °C.

It is clearly evident from the images that the substrate temperature plays a vital role in modifying the morphology of the grown thin films. The compositional analysis of the thin films was carried out by EDS and no significant deviation was found in the chemical compositions of the target and the films. The variation in the changes in the elemental atomic percentage was within the instrumental error limit of 4 atomic percent.

The XRD pattern of FeGa thin films deposited at different substrate temperatures are given in Fig. 2(a–d). It was observed that the peaks are sharp indicating highly crystalline thin films. There is a coexistence of two phases of FeGa in the sputtered films namely, $L1_2$ and B2, which was expected from the compositional range of the thin films. There are previous reports backing multiple phase existence in the literature [20,21]. Similar to our previous report [19], it was observed that $L1_2$ phase dominates in the thin films. It can be seen that at lower substrate temperatures, $L1_2$ phase was present. B2 phase seems to emerge only at higher temperatures, though its presence can be noticed at lower temperatures but with less clarity. The crystalline size was calculated and was seen to increase from 20 nm to 35 nm with increase in temperature. As the temperature increases, the crystallinity of the thin film also improves. The peaks present at 2Θ values of 32.29° and ~54° are the (100) and (111) planes corresponding to B2 phase. The other peaks at ~35.61°, 43.3°, 57.23°, 62.89° also correspond to FeGa thin films with ordered f.c.c. $L1_2$ phase [20]. As the temperature increased to 350 °C, the crystallinity of the thin film was reduced. The sizes of the particles present in this film are much smaller when compared to the films grown at lower temperature. Although with higher temperature, better crystallinity was expected, the XRD pattern for the film grown at 350 °C states otherwise. This deviance was explained by the collapsed columnar microstructure at this substrate temperature. The change in growth pattern of the thin film was altered, resulting in poorly crystalline film formation.

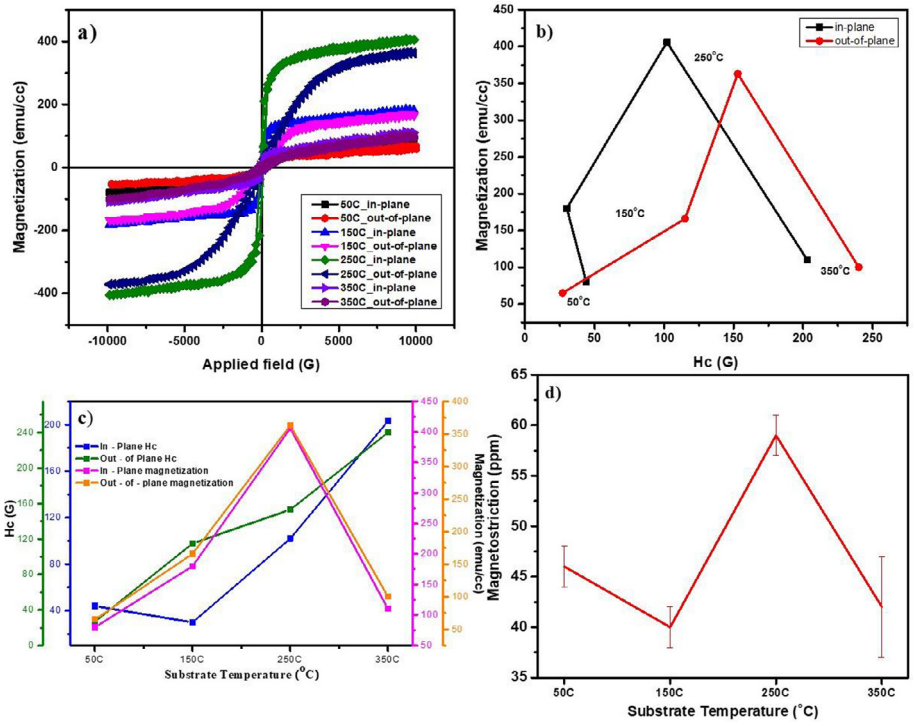

Fig. 3. (a) Room temperature in-plane and out-of-plane M-H curves for films deposited at 50 °C, 150 °C, 250 °C and 350 °C (b) relation between coercivity (H_c) and magnetization (M_s) obtained from the different substrate temperature grown thin films. (c) In-plane and out-of-plane coercivity (H_c) and magnetization (M_s) as a function of substrate temperature. (d) Effective Magnetostrictive constant (λ_{eff}) as a function of substrate temperature.

Surface topology of the thin films was analyzed using Atomic Force Microscopy. The topography presents spherical particles (Fig. S2(a–d)) with the different sizes as observed from SEM. The RMS roughness of the thin films (Fig. S2(e)) was seen to increase with respect to temperature. This was consistent with SEM images. There was a huge increase in surface roughness of the thin film deposited at 350 °C. This was expected since we observed columnar structure collapse and widely varying particle sizes from the SEM images. As there is topological non-uniformity, we obtain higher RMS roughness for the higher temperature grown thin film.

3.2. Magnetic and magnetostrictive studies

The magnetic properties of the sputtered thin films were analyzed using room temperature VSM in both in-plane and out-of-plane directions (Fig. 3a). Fig. 3a gives the in-plane and out-of-plane hysteresis loop of FeGa thin films deposited at increasing substrate temperatures. It can be observed that the magnetization of these films increases from 80 emu/cc to 400 emu/cc on increasing the substrate temperature to 250 °C in in-plane direction (Fig. 3c). Further increase in temperature, however, decreased the magnetization. A similar trend was observed in out-of-plane M-H curves of the thin films as well. Here the lowest magnetization value was 65 emu/cc for 50 °C deposited film, while the highest magnetization was 363 emu/cc for 250 °C grown thin film.

The increase in substrate temperature also brought changes in the coercivity (Hc) values of the films. An increase in Hc values was observed as the temperature rose from 50 °C to 350 °C. The Hc for in-plane measurements were 44, 30, 102 and 203 G for 50 °C, 150 °C, 250 °C and 350 °C grown thin films. For out-of-plane measurements, Hc values were 27, 115, 153 and 240 G as the temperature increased from 50 °C to 350 °C (Fig. 3b). Fig. S3 brings out the correlation between the surface roughness and the coercivity. It is observed that the coercivity increases with the surface roughness due an increase in surface pinning states. However, the contribution of surface roughness to coercivity would be only to a few tens of nm thickness from the film surface. Hence this enhancement in coercivity in these thin films are also attributed to the increase in the number of dislocations and defects that are present due to the higher deposition temperatures. This is consistent with the observation from the cross-sectional SEM images, where morphological evolution from truly columnar to dissolution of columns to planar and finally to collapsed columnar structure is evident. The dislocations also act as the pinning centres that can also increase the coercivity. The Hc, remnant magnetization (Mr) and saturation magnetization (Ms) values derived from the M-H hysteresis curves in both in-plane and out-of-plane measurements are tabulated (Table S1).

The main purpose of analyzing the magnetic properties of the FeGa thin films in in-plane and out-of-plane modes was to study the anisotropy in these thin films. As can be seen from Fig. 3a, all the films show anisotropy with respect to the measurement direction. The maximum anisotropy was observed in the thin film grown at 250 °C. The thin films have their easy axis of magnetization along the substrate. The saturation field required is much lower in the substrate plane compared to perpendicular to it.

The calculated effective magnetostriction values of these films are represented in Fig. 3d. All the sputtered films are magnetostrictive and their values range from 42 to 59 ppm. Initially, the films deposited at 50 °C gave a striction value of 46 ± 2 ppm, which increased to 59 ± 2 ppm at 250 °C substrate temperature. Increase in substrate temperature decreased the magnetostriction to 42 ± 5 ppm, which was lower to that of 50 °C deposited film. The magnetostriction of the thin film depends on certain factors like magnetization, crystallinity and phases present. The magnetostriction of the FeGa is strongly dependent on the phases present and out of the many phases present in the material, phases such as A2 and B2 contribute to positive magnetostriction and those such as L_{12} and DO_3 contribute to negative magnetostriction [22]. The higher magnetostriction constant for 250 °C substrate temperature deposited thin film can be attributed due to the presence of a higher fraction of B2 phase.

Basumatary et al. [16] studied the influence of substrate temperature on the structure, microstructure and magnetic properties of FeGa thin films. They carried out the deposition at a pressure of 5 mTorr and power of 100 W at room temperature, 300 °C, 400 °C, 450 °C, 500 °C and 550 °C. It was found that these result in polycrystalline thin films with single disordered A2 phase. They also observed increase in magnetostriction with temperature and obtained a maximum value of 200 μ-strains at 450 °C, but further increase in temperature decreased the magnetostriction. This decrease is attributed to the increased surface roughness which introduces considerable defects hampering the development of large magnetostriction. In the present case, the experimental parameters are different, like Ar partial pressure of 4E-2 mbar and sputter power of 80 W with substrate temperatures ranging from 50 to 350 °C. Similar to Basumatary et al., a rise in magnetostriction with increase in substrate temperature has been observed. But the magnitude of the striction was 60 ppm, which is lower to the value obtained by them. The films deposited by Basumatary et al., had a single disordered phase, which is known to possess maximum magnetostriction. Our films had coexistence of two phases, $L1_2$ and B2, of which one is an ordered phase, that naturally brings down magnetostriction. Though the value of magnetostriction obtained is much lower than Ref. [16], our value is fairly moderate when compared to the other reports in literature for FeGa thin films. This study also reports the interplay and influence that phases have on the magnetostriction, along with the morphology, on the magnetostriction of these thin films. The magnetostriction of the thin film depends on the magnetization, so higher the magnetization, higher the magnetostriction. A strong correlation between magnetization and magnetostriction is seen as both increases with growth temperature, reaches maximum at 250 °C and decreases on further increasing the growth temperature to 350 °C. Crystallinity also favors higher magnetostriction whereas decrease in crystallinity deteriorates magnetostriction. XRD results imply that the degree of crystallization decreases for the sample grown at 350 °C as the intensity of the diffraction peaks are relatively less and FWHM of the peaks are broad. The thin film deposited at the substrate temperature of 250 °C is highly crystalline and further increase in substrate temperature turns the film to be of poor crystallinity. Hence, the highest magnetostriction constant of 59 ± 2 ppm was obtained for thin films deposited at 250 °C. Abbas et al. [23] has reported the decrease in magnetostriction constants for amorphous FeGaSiB thin films by a factor of 2.5, when compared to crystalline films.

4. Conclusions

FeGa thin films were deposited at different substrate temperatures while keeping all the other growth parameters unaltered. $L1_2$ and B2 structural phases coexisted in the films with their percentage varying with substrate temperature. The B2 phase becomes more pronounced at higher substrate temperature. Morphological evolution from uniform prismatic to non-uniform spheres and planar flakes of varying sizes at higher substrate temperature was observed. The magnetization of the films increased with substrate temperature till 250 °C but decreased on further increase in temperature. Magnetic anisotropy, which is vital in obtaining high magnetostriction, was highest in 250 °C grown thin film. Further increase in temperature decreases anisotropy. It was

observed that magnetostriction value measured also corroborated very well with in-plane and out-of-plane anisotropy. The thin film prepared at the substrate temperature of 250 °C possesses high magnetostriction and is soft magnetic with reasonable coercivity to render it a potential candidate for sensor and actuator applications.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.jmmm.2017.11.030.

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