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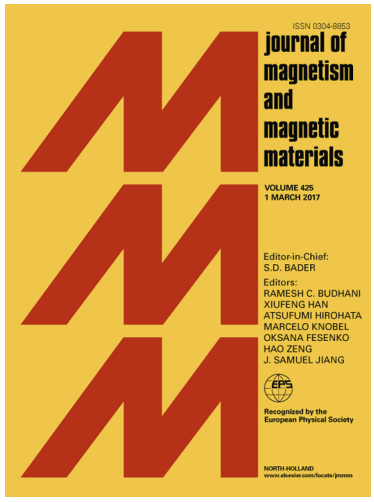

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Fabrication and Characterization of Magnetostrictive Amorphous FeGaSiB thin films.

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Abstract

In this work, amorphous FeSiB and FeGaSiB thin films have been fabricated on silicon substrates using a co-sputtering- evaporation deposition technique. The effect of adding gallium into FeSiB (Metglas) thin films on the structure, magnetic properties and magnetostriction have been studied. From x-ray diffraction (XRD), all the films were amorphous and the observed peaks were for the Si substrate. X-ray Photoelectron Spectroscopy (XPS) measurements were carried out to determine the film's composition, which was found to be $Fe_{83}Ga_{11}Si_{5.2}$ $B_{0.8}$. Atomic force microscopy (AFM) images were taken to measure the film thickness along with studying the surface topography. It was found that the film surface had an average roughness of 0.461 nm. For both FeSiB and FeGaSiB thin films, the effect of the thickness of the films on the magnetic properties and magnetostriction were investigated. The results showed that adding Ga into the FeSiB films changed the magnetic properties by reducing the saturation induction along with changing the magnetic anisotropy from uniaxial to isotropic. For the FeGaSiB films, the coercive field decreased and the saturation field (H_s) increased with film thickness. The magnetostriction constants of the FeGaSiB films were all larger than the FeSiB films for thicknesses greater than 40nm.

Keywords: Magnetostrictive, amorphous, anisotropy, MEMS, Metglas.

1. Introduction

Recently, the development of magnetic amorphous materials has become of interest for Micro-Electro-Mechanical (MEMS) application such as low field sensing applications [1,2]. One of the main materials of interest is magnetostrictive Fe-based amorphous thin films, such as FeSiB (Metglas). Thin films with amorphous structures have an absence of atomic long range order, thus they only have a random short range order, which leads to magnetocrystalline anisotropy coefficients of zero [3] compared to crystalline thin films. In this case, the magnetic properties of these films can be affected by magnetoelastic anisotropy and shape anisotropy [3], such that large magnetic anisotropy can be generated by inhomogeneous strains within the amorphous alloys [4]. Due to their amorphous structure, FeSiB thin films [5] can be magnetized and demagnetized quickly using a low applied magnetic field (<40 kA/m). Thus reducing the saturation field, while maintaining/increasing the magnetostriction constant of amorphous thin films is of interest to produce new low field sensing thin films.

From previous research, it has been found that the dimensions between the Fe-Fe atoms in Fe-based alloys can be influenced by adding non-magnetic elements, which affect the magnetostriction constant [6]. Adding of nonmagnetic materials such as Al, Be into these magnetic films changed their magnetic properties [6]. Gallium is another non-magnetic material, which has been an additive element used to improve the magnetic performance of Fe films [7], by substitution onto Fe atoms sites within the crystalline structures [7]. Influence of Ga composition has been studied in Fe-Ga thin films [8-10]. It was found that the magnetostriction constants of Fe-Ga alloys are responsive to Ga composition [11,12]. For example, Hatrrick-Simpers et al [13] determined that the magnetostriction magnitude in Fe-Ga thin films reached a maximum value of about 190 ppm when the percentage of Ga was 32 at. %., while Wang et al [14] found that the magnetostriction of $Fe_{81}Ga_{19}$ thin films was about 42 ppm at a field of 5kA/m and thickness 660 nm. Morley et al [15, 16] developed a co-sputtering-evaporation technique which is suitable for growing thin FeGa films and overcomes the difficulties of sputtering Ga. Using the co-sputtering-evaporation technique allowed Javed et al [17] to control the Ga composition within the deposited films by varying the growth parameters such as sputtering gas pressure, sputtering power, and Ga evaporation rate. Three sets of films have been grown on

silicon substrates, the first set, with thickness 75 nm, was deposited with a constant pressure 3 μbar and iron sputtering power 30 W while the Ga evaporation rate was varied (02, 0.3 and 0.4). The second set, with thickness 50 nm, was grown with varied sputtering power while other parameters were constant (pressure 4 μbar and Ga evaporation rate 0.3). The third set, with thickness 50 nm, was deposited with a varied pressure, constant power 20 W and Ga evaporation rate 0.25. They found that the magnetostriction constant ranged between 40 ppm to 80 ppm for the Ga concentration range of 14 at% to 32 at%. They also found that the magnetostriction constant was almost independent of the fabrication parameters. While the saturation field strongly depended on the fabrication parameters, with the softest films grown at the lower chamber pressure. The main disadvantage of these crystalline Fe-Ga films for applications was not the magnetostriction constant, but the saturation field. For all the films, it was larger than 50 kA/m, which means that future work has to focus on maintaining the highest magnetostriction constant while reducing the saturation field to below 10 kA/m.

One way to do this is to amorphise the films, thus this work has focused on the fabrication of magnetostrictive amorphous FeSiB thin films with and without the addition of Ga. Using the co-sputtering-evaporation technique to grow the amorphous magnetic thin films, by changing the growth parameters to control the composition of the FeGaSiB films, meant that a rapid quenching process was not needed to achieve the films' amorphous nature. This paper presents a study of FeSiB and FeGaSiB films, comparing how their magnetic properties and magnetostriction constants vary with the addition of Ga as a function of thicknesses.

2. Experimental

The co-sputtering–evaporation technique [15] was utilized to deposit FeSiB and FeGaSiB amorphous thin films. The formation of the amorphous films was made without rapid quenching methods. The films were grown on Si (100) substrates, which were washed using acetone and isopropanol (IPA) before deposition. Three Si substrates were used per film growth, attached to a glass slide using polymethyl methacrylate (PMMA). To determine the thickness of the films accurately, a PMMA blob was put on one Si substrate before growth using a small stick of wood (toothpick). After growth, the PMMA blob was washed away using acetone, to leave a sharp edge, so that the film thickness could be measured. Metglas 2605SA1 with composition $Fe_{85}Si_{10}B_5$ was used as the sputtered target. Two different film sets were grown with a thickness

range from 24 to 100 nm. The first set was FeSiB thin films at a chamber pressure (P_{Ar}) of 4μbar and sputter power (P_{FeSiB}) of 20W. The second set was FeGaSiB thin films with sputter power (P_{FeSiB}) of 20W, pressure (P_{Ar}) of 4μbar and Ga evaporated with a constant arbitrary rate, R_{Ga} = 0.2.

For both sets of films, the magnetic properties were measured using a magneto-optical Kerr effect (MOKE) magnetometer, in the the transverse mode, with the DC magnetic field being large enough to saturate the films (Max field applied was 39.78 kA/m). Normalized magnetic hysteresis loops were measured to characterize the films' magnetic properties. For each film, normalized hysteresis loops were measured by rotating the film within the magnetic field at different angles from 0° to 180° with 30° steps to study the magnetic anisotropy. The effective magnetostriction constant (λ_s) was determined via the Villari Effect [16] at room temperature. This involved using a set of bending tools with different radii (R), which induce a strain within the film. The film's hysteresis loops were measured for each bend radii on the MOKE magnetometer. From the loops, the straight line method was used to determine the saturation field (H_s) induced by the strain [18]. The H_s was plotted as a function of the inverse bending radii (1/R) and magnetostriction constant (??$_{??}$) was calculated using [16,18].

$$\lambda_s = \frac{d(H_s)}{d\left(\frac{1}{R}\right)}\left\{\frac{2\mu_o M_s\left(1-v^2\right)}{3tE}\right\} \quad (1)$$

Where the bend radii were R=300, 400 and 500 mm, ??$_o$ is the permeability of space, ??$_s$ is the magnetization, and for the silicon substrate Young's Modulus, E=130 GPa, thickness of substrate, t =380 μm and Poisson ration of substrate, ??=0.28. Hs is the anisotropy field created by the bending tool,

The Bruker D2 phaser technique, X-ray diffraction (XRD) with Cu $K_{\alpha 1}$ 1.54184 Å was used to analyze the films' structure. The θ/2θ mode was used, the 2θ range from 30 to 80° was used to check the structure of the film (for bcc Fe 2θ ~ 45° and for bcc Fe-Ga 2θ ~ 45°) and silicon substrate (2θ = 69°, 61.7°, and 33°), and the 2θ range from 35 to 55° was used to avoid the Si substrate peaks. A Quantum Design 6 T MPMS SQUID system was used to measure the saturation magnetization of the sample at room temperature (300K) with an applied field of 39.788 kA/m. X-ray Photoelectron Spectroscopy (XPS) utilizes a thermo theta probe, with

parameters of a pass energy of 40 eV, dwell time 100 ms (10 scans total for hi-res) and a monochromated Al K_α X-ray as a source. It was used to determine the compositions of films. Three runs were done using the XPS in three different places on the film surface area and an average was taken. Atomic force microscopy (AFM) was used, in tapping mode, to measure the thickness of the samples using the step produced in fabrication and to check the topography and roughness.

3. Result & discussion

XRD analysis was carried out to determine whether the films were fully amorphous or if they contained nanocrystalline cluster within an amorphous matrix. From Fig. 1a, the XRD results for both of FeSiB and FeGaSiB thin films are presented, it was found that all the films were amorphous and the peaks observed were for the Si substrate. From Fig. 1b inset, it is observed that there are no peaks for the FeGaSiB film at $2\theta \sim 45^{o}$, which is where the Fe-Ga films (110) texture peak occurs [19]. Thus, it is concluded that the addition of Ga into the FeSiB did not change the morphology of the films.

From the XPS results, it was found that the FeGaSiB films had the composition $Fe_{83}Ga_{11}Si_{5.2}B_{0.8}$. It is clear from this result that the Ga replaces more of the Si and B atoms than the Fe atoms in the $Fe_{85}Si_{10}B_5$ target. This could be due to the Ga being heavier than the Si and B, so causing more collisions within the plasma, hence allowing fewer atoms to reach the substrate.

The AFM data determined that the films thickness range were from 24±2 nm to 100±2 nm. The thicknesses were also used to calibrate the growth sensor for the FeSiB and FeGaSiB films. It was found that adding Ga atoms to FeSiB films decreased the deposition growth rate of the films.

Fig. 2 shows the topography of the 24 nm FeGaSiB film. The film's surface had an average roughness (Ra) of 0.461 nm with a root-mean-square (RMS) surface roughness of 0.944 nm, due to the 24 nm with spots on the surface. Since the films had a smooth surface, and no grains were observed, this confirms that the films were fully amorphous compared with AFM images of grainy films such as FeGa film[18]. These white spots observed are due to a contamination on the surface, which has been observed before using this fabrication technique [18].

From the saturation induction data measured using of VSM technique (Fig. 3), it is observed that the saturation induction of the 23μm ribbon FeSiB $\mu_\circ M_s = 1.56\,T$, 86nm FeSiB film $\mu_\circ M_s =$

1.15 T, and the 86 nm FeGaSiB film was $\mu_\circ M_s = 0.96T$. Thus, adding Ga atoms, which are nonmagnetic into FeSiB films reduced the saturation induction from 1.56 T to 0.96 T.

Figure 4 shows the saturation induction for all the FeGaSiB thin films as a function of thickness. It can be seen that the saturation induction changed slightly with the thickness, from 1.1T for the 24 nm to 0.96 T for the 86 nm film and then increased to 1.23 T for the 100 nm. This change in saturation induction as a function of thickness for the same composition is due to the amorphous nature of the films and the atoms have no long range order, only short range order. This means that the nearest neighbour atoms change from film to film. The magnetic moment of Fe atoms depend on the nearest neighbours and the saturation magnetisation is an average of the moments, which will change for each film, this effect of the nearest neighbour atoms can be the reson of film 100 nm to have higher saturation magnetisation compared to the other films. The induction (magnetisation) of magnetic film depends on the nearest neighbours. For example, crystalline $Fe_{40}Co_{60}$ saturation induction is higher when the sample has an ordered bcc structure compared to a disordered fcc structure [20]. As the FeGaSiB films are amorphous and have no long range order or set lattice constant, between films the nearest neighbours to the Fe will differ, thus can change the saturation induction of the films.

For the FeSiB amorphous films, it was found that all the films had uniaxial anisotropy, determined from the change in the normalized remanence magnetization as a function of field angle. For example, the saturation field of the 25 nm FeSiB film was 2.308kA/m along the easy axis and 5.232 kA/m along the hard axis. This uniaxial anisotropy is likely to be due to inhomogeneous strains within the films [4]. While adding Ga into the FeSiB thin films changed the magnetic anisotropy, as the films were magnetically isotropic. For example, for the 49 nm FeGaSiB film, the saturation field was 2.308kA/m for all field angles. This can be understood that the Ga is a diamagnetic element, thus the existence of Ga within the amorphous films weakens the anisotropy, so producing magnetic isotropic properties.

Figure 5 shows the comparison of the anisotropy and saturation fields as a function of thickness for the FeSiB and FeGaSiB thin films respectively, prepared under the same growth conditions. The FeSiB films have uniaxial anisotropy, this means they have easy and hard loops as a function of applied field angle, thus an anisotropy field was determined from the normalized hard axis, as the field where the normalised magnetisation reached 1 (or -1). It is observed that the anisotropy fields for the FeSiB films decreased with increasing thickness (fig. 5). While the

FeGaSiB films are isotropic, this means they have the same shape hysteresis loops for all applied field angles, thus a saturation field is determined from the isotropic hysteresis loops, again at the field where the normalised magnetisation reached either 1 or -1. It was found that the FeGaSiB films saturation fields increased with the increasing thickness (fig. 5). Thus the addition of Ga has not only changed the anisotropy from uniaxial to isotropic [21] but has also increased the saturation field.

Figure 6 shows the coercive fields for both film sets. For the FeSiB films the coercive field increases with thickness up to 40-50nm, then decreases for films thicker than 50nm, while for the FeSiGaB film, the coercive field decreased as the thickness increased. Such that for the thicker films (>50nm), the coercive field of the FeGaSiB films was lower, i.e. for the 86 nm FeGaSiB film was H_c = 0.159 kA/m which was a factor 5 smaller than the 86 nm FeSiB film (H_c = 4.775kA/m).

Figure 7 shows the hysteresis loops, for three bend radii, R, (300, 400, and 500) mm, for the 74 nm FeGaSiB film, it can be seen that there is a change in loops. The difference is due to the effect of the strain on the magnetization of the film resulting from the bending of the film. From these loos, the different anisotropy fields at each radus are determined and plotted against (1/R). The slope of this plot gives the result of the first term in equation (1).

Figure 8 shows the change in magnetostriction constant (??$_{??}$) with thickness. It is observed that the FeGaSiB films have higher magnetostriction constants than the FeSiB films for thicknesses over 40nm. The 50 nm FeGaSiB film had the largest ??$_{??}$ of 17 ppm. For the FeSiB films, ??$_{??}$ decreased with the increasing thickness, while for the FeGaSiB films with t > 50nm, ??$_{??}$ ~13ppm but were still higher than the FeSiB films. This means adding of Ga into the FeSiB thin films increased the magnetostriction constant by giving the amorphous structure dilation by increasing the space between the atoms because the radius of Fe atoms is slightly lower than the radius of Ga atoms. And this would also reduce the inhomogeneous strain, which can be the reason the films anisotropy changed from uniaxial to isotropic. Compared to the crystalline Fe-Ga films, the saturation fields for the amorphous FeGaSiB films were a factor 50 smaller, while the magnetostriction constants were a factor 2.5 smaller. Thus the saturation field has been reduced, but so has the magnetostriction constant. Further work will be carried out to maintain the low saturation field, while increasing the magnetostriction constant to values close to the crystalline Fe-Ga films.

Conclusion

Co-sputtering-evaporation deposition technique was used to fabricate amorphous Fe-based magnetic films. The addition of Ga into FeSiB did not change the morphology. The films had an average roughness of 0.461 nm and showed no grain structure. While the addition of Ga reduced the saturation induction from 1.56 T to 1.1-0.96 T. The coercive field was < 1591.54 A/m for all films studied independently of composition and thickness. The saturation field increased with the addition of Ga. The magnetostriction constants were higher in FeGaSiB thin films for thicknesses larger than 40 nm compared to the FeSiB films. The 50 nm FeGaSiB film had the largest magnetostriction constant of 17 ppm. The advantage of amorphous films over ribbons for applications includes reduced surface roughness and achieving thickness less than 1 micron. Flims are also easier to integrate with MEMS devices compared to ribbons.

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Figure Captions

Figure 1 X-ray diffraction of (a) FeSiB, FeGaSiB thin films and Si substrate (b) FeGaSiB thin films with change the thickness. Inset curve XRD for the 2θ range from 35 to 55^{o}.

Figure 2 AFM image scan of the 24 nm FeGaSiB thin film. The scan size is 4μm^{2}.

Figure 3 Saturation induction as a function of magnetic field at room temperature for the 86nm FeSiB, 86nm FeGaSiB film and 23μm ribbon (Target) FeSiB.

Figure 4 Saturation induction as a function of thickness of the FeGaSiB thin films.

Figure 5 Anisotropy and saturation fields as a function of thickness for FeSiB and FeGaSiB thin films.

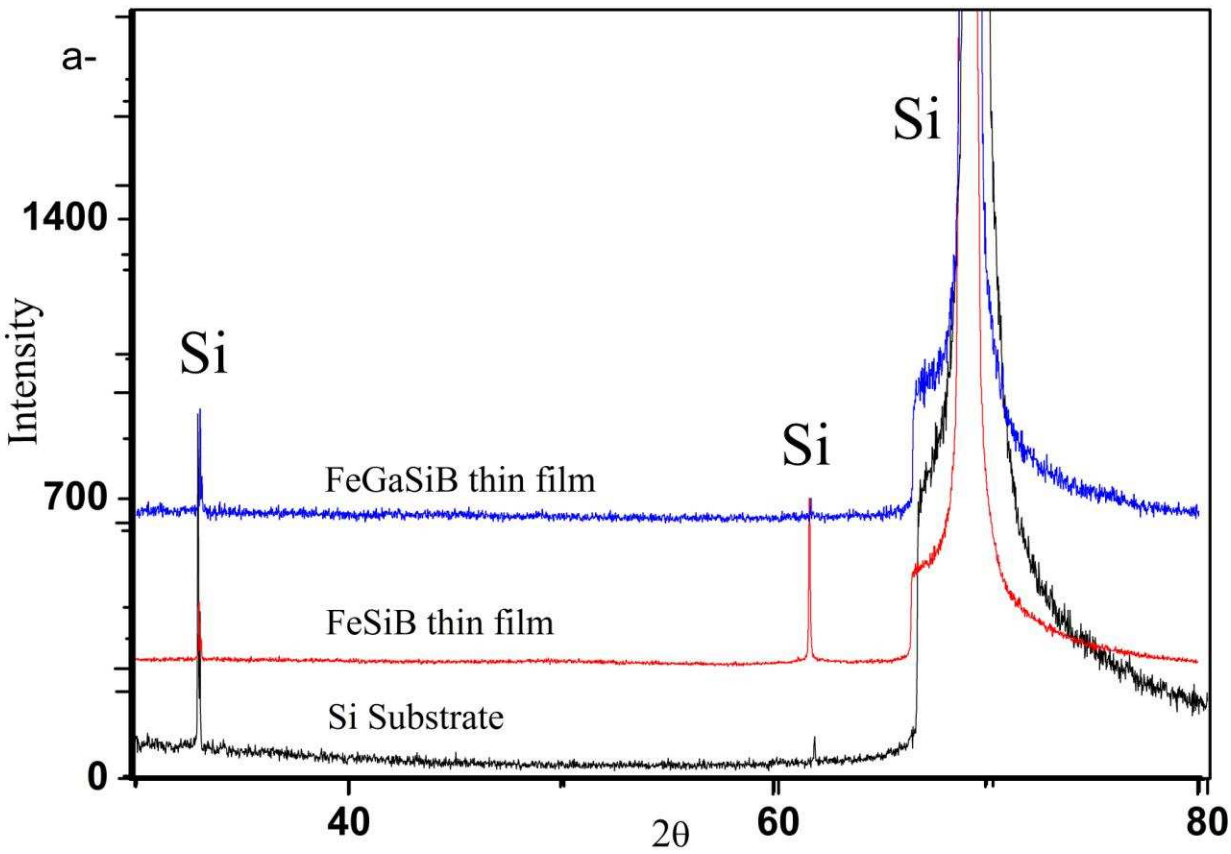
Figure 6 Coercive fields as a function of thickness for FeSiB and FeGaSiB films.

Figure 7 Magnetizing curves for FeGaSiB thin film, a bended film with three bend radii.

Figure 8 Magnetostriction constants for FeGaSiB and FeSiB films as a function of thickness.

Highlights of this work

1- The current work is motivated on fabrication amorphous FeGaSiB thin films by co-sputtering-evaporation technique without quenching methods.
2- Study the effect of adding gallium into FeSiB (Metglas) thin films on the structure, magnetic properties and magnetostriction constant.
3- Study the effect of thickness on the magnetic properties and magnetostriction constant.
4- Compare the results of both films.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FeSiB thin film
Si Substrate
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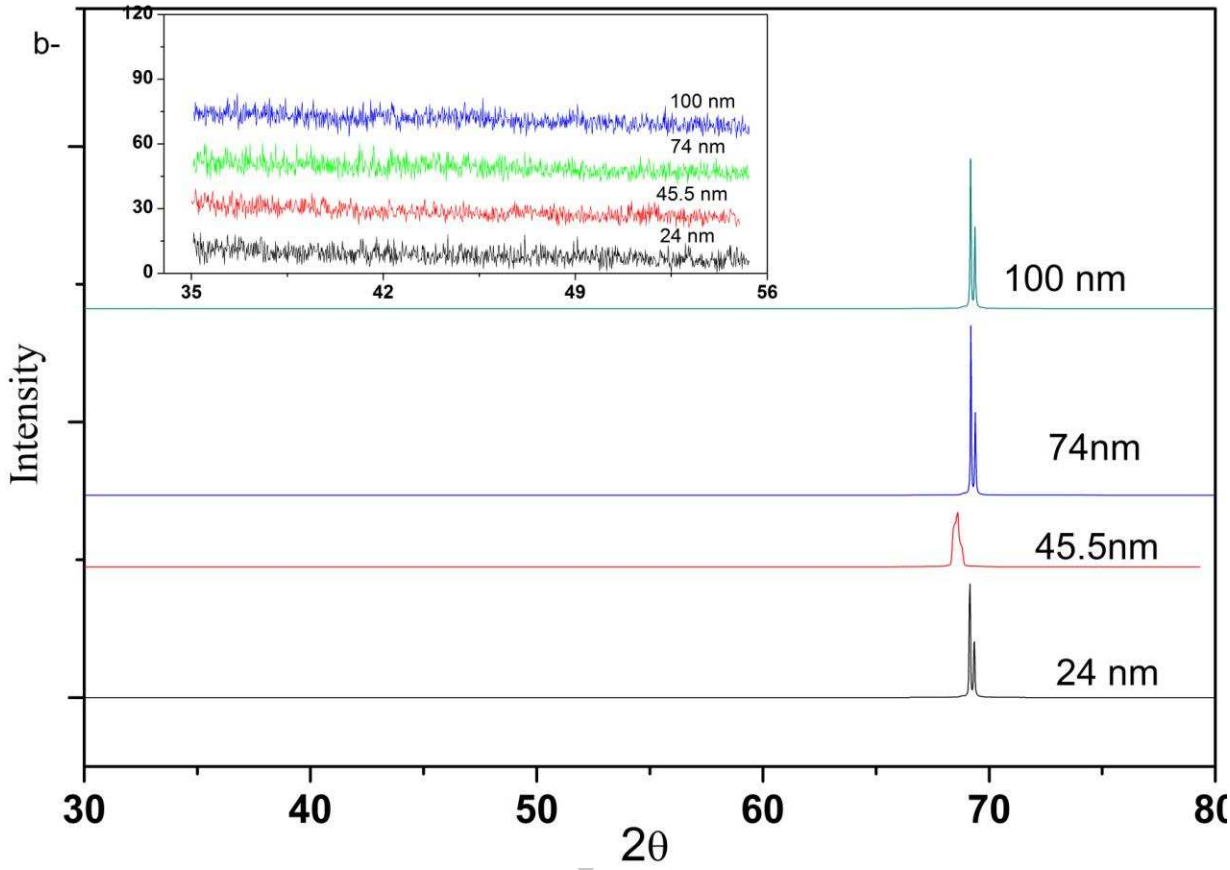
60
80
2θ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90
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30
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35
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56
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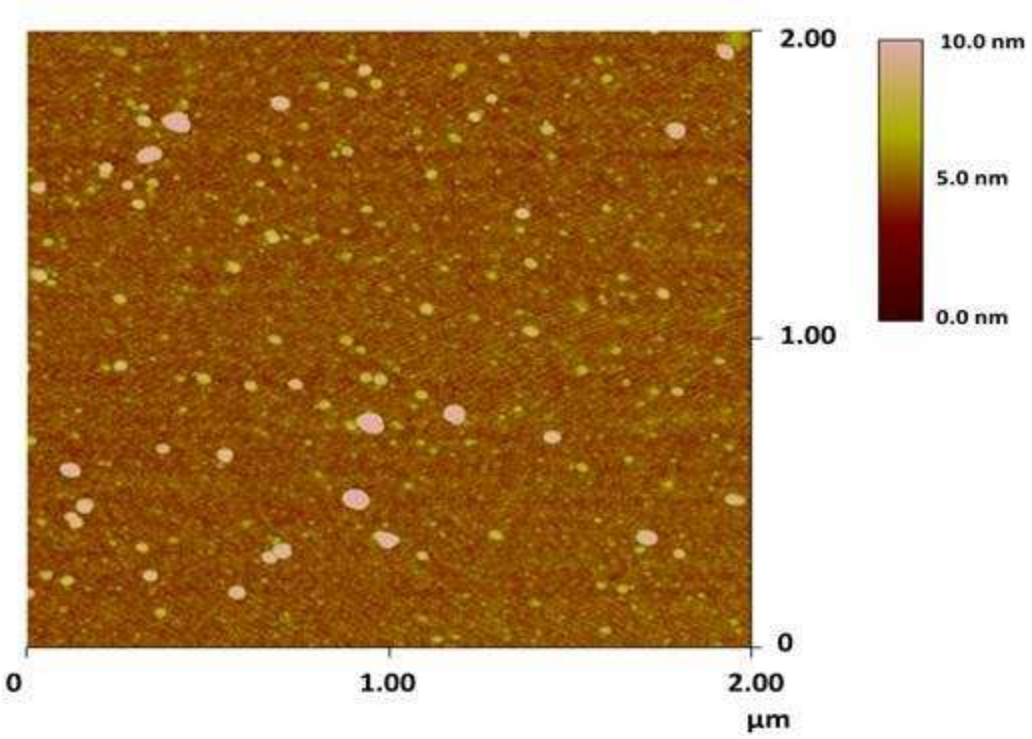
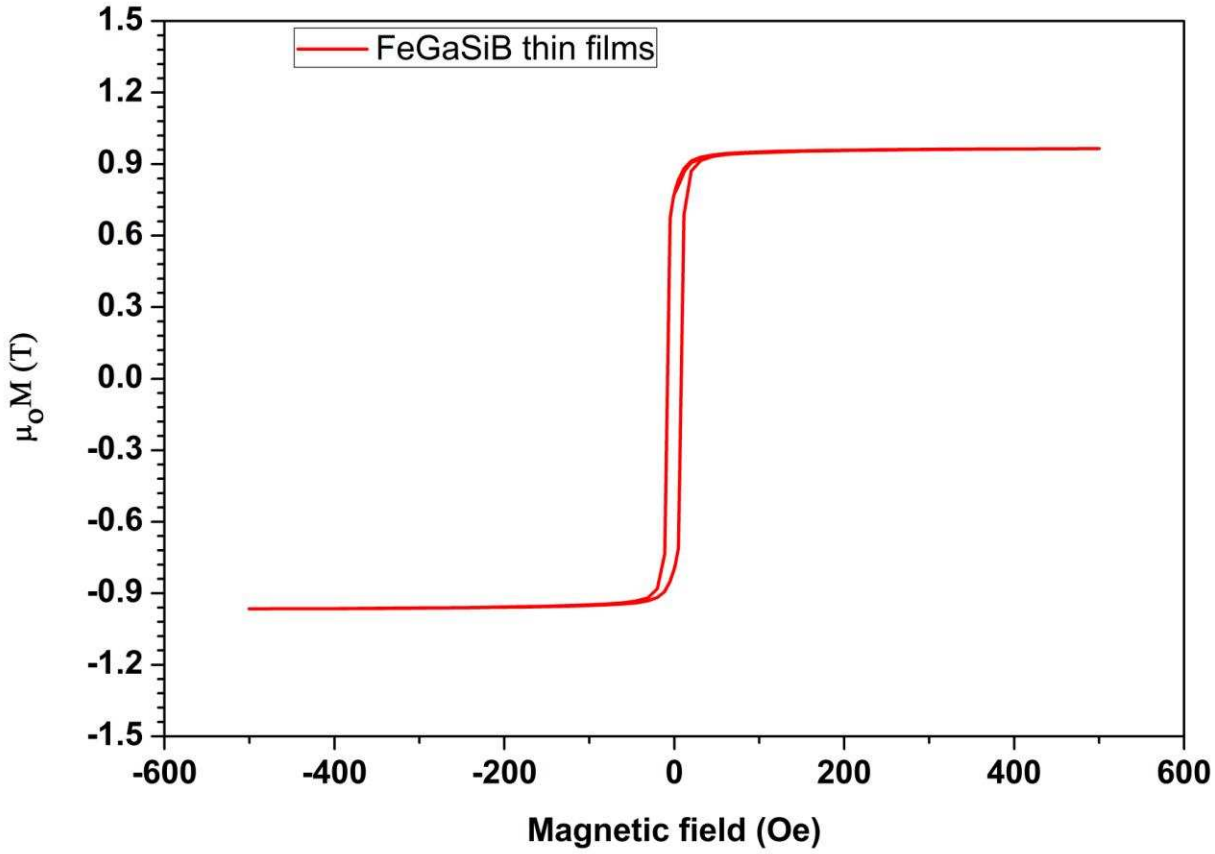
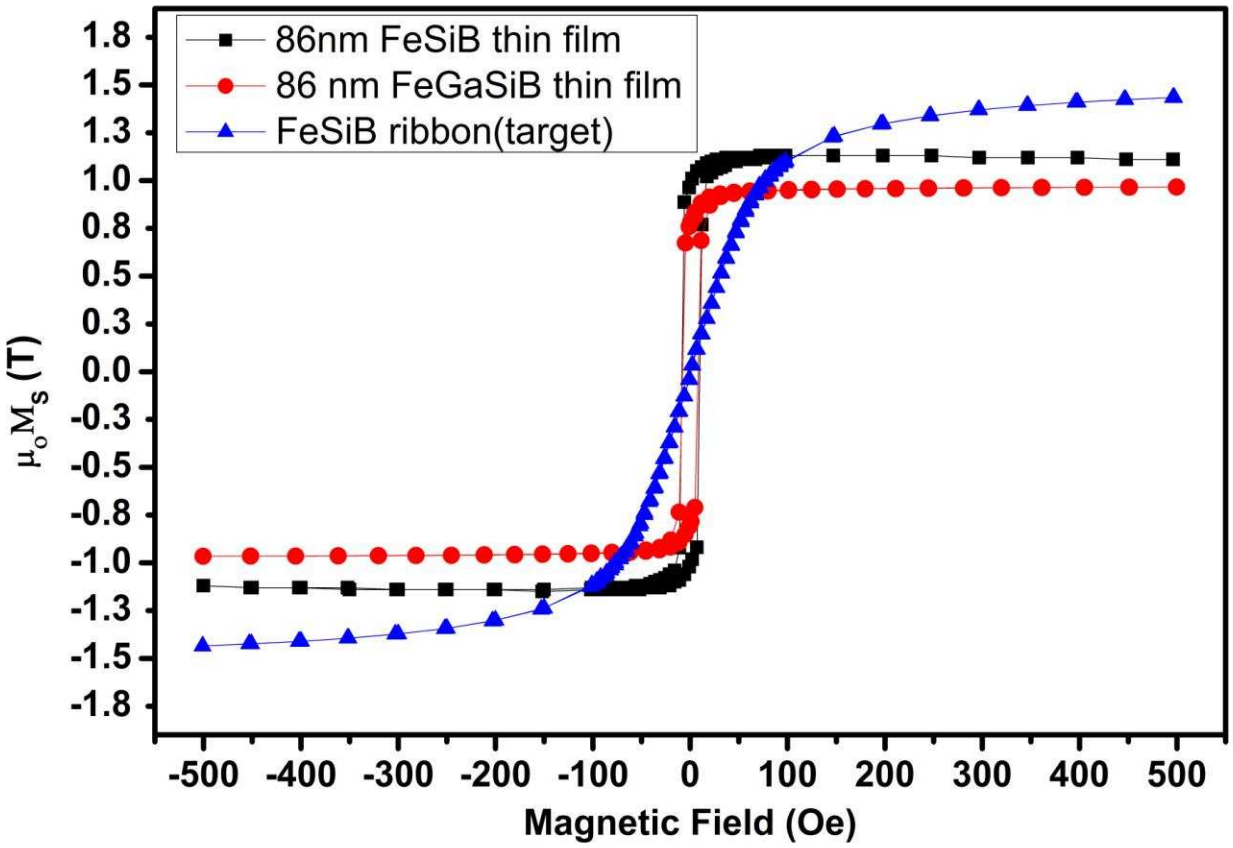



FeGaSiB thin films
1.5
1.2
0.9
0.6
0.3
0.0
-0.3
-0.6
-0.9
-1.2
-1.5
$\mu_o M$ (T)
-600
-400
-200
0
200
400
600
Magnetic field (Oe)


86nm FeSiB thin film
86 nm FeGaSiB thin film
FeSiB ribbon(target)
$\mu_o M_S$ (T)
1.8
1.5
1.3
1.0
0.8
0.5
0.3
0.0
-0.3
-0.5
-0.8
-1.0
-1.3
-1.5
-1.8
-500 -400 -300 -200 -100 0 100 200 300 400 500
Magnetic Field (Oe)