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Influence of Ar Pressure on the Magnetic Properties of Amorphous FeGaSiB Thin Films

Qayes A. Abbas[1,2], Student Member, IEEE, Nicola A. Morley[1], Member, IEEE, August Johansson[3] and Tom Thomson[3], Senior Member, IEEE.

[1] Dept of Materials Science & Engineering, University of Sheffield, Mappin Street, UK, S1 3JD
[2] Dept of Phys, College of Education for Pure Science, University of Anbar, Anbar, Iraq
[3] School of Computer Science, University of Manchester, Manchester, UK

Magnetostrictive amorphous FeSiB and FeGaSiB thin films, thickness 50nm have been grown by the co-sputtering-evaporation technique with a range of Ar pressure (4 - 8 µbar) to control the Ga percentage within the films and study their effect on the magnetic, structural and magnetostriction properties. By x-ray diffraction, it was found that all the films had an amorphous structure and the only peaks present were for Si substrate. Using a magneto-optical Kerr effect (MOKE) magnetometer, it found that, for the FeSiB films, the anisotropy field (H_k) increased slowly as the pressure increased, while for the FeGaSiB films, the saturation field (H_s) ≈ 4000 A/m for all pressures. For both the film sets, the coercive field (H_c) was less than 800 A/m. The magnetostriction constants (λ_s) of the FeSiB thin films increased with increasing pressure. While for the FeGaSiB films, the magnetostriction constant decreased with increasing the sputtering gas pressure, with the maximum λ_s = 11.4 ppm, at the lowest pressure 4µbar. Thus it was determined that the addition of Ga atoms reduced the intrinsic stress within the films, while maintaining the amorphous morphology.

Index Terms—Magnetostrictive, thin films, amorphous, magnetic properties, MEMS.

I. INTRODUCTION

AMORPHOUS MAGNETOSTRICTIVE materials in the form of thin films have become of interest for microelectromechanical systems (MEMS) such as magnetostrictive sensors [1, 2]. The inverse magnetostrictive effect in thin films is defined as a change in magnetisation under applied stress [3, 4]. Current work has demonstrated the promise of amorphous magnetic FeGaSiB thin films for use in applications that involve low saturation field (<5kA/m) and high magnetostriction constant (>20ppm). These applications include low field sensors and artificial multiferroic heterostructures, which use electric fields to control the magnetisation via the strain induced by the piezoelectric substrate [5, 6].

Sputtering parameters such as Ar pressure affect the magnetic, structural and magnetostriction properties of the deposited films as a result of their sensitivity to the growth parameters [7]. The sputtering pressure induces intrinsic stresses, which can be either tensile or compressive dependent on varying Ar pressure [8]. Other stresses arise from the lattice misfit between the film and the substrate and the difference between the thermal expansion coefficient of both film and substrate. Javed et al [9], studied the effect of pressure on the magnetic properties and structure of magnetostrictive $Fe_{100-x}Ga_x$ (19 ≤ x ≤ 23) thin films produced by co-sputtering-evaporation. They found that increasing the pressure greater than 3µbar, changed the anisotropy of the films from uniaxial to isotropic. They also found that the composition of Ga within the films was influenced by the pressure due to inelastic collisions with the atoms of sputtering gas, hence changed the ratio of Fe:Ga atoms. While the effective magnetostriction constant for all the samples was a constant around 60 ppm with varying the pressure. Further work by Javed et al [10], investigated magnetostrictive 50nm thick $Fe_{100-x}Ga_x$ (14 ≤ x ≤ 32) thin films grown on Si substrates using the co-sputtering-evaporation technique and studied the effect of growth parameters (Ar pressure, Power of target and Ga rate) on the structure, magnetic properties and magnetostriction constant. They found that by varying the Ar pressure and holding the other parameters constant, the Fe atoms were affected thermally, such that the saturation field (H_s) decreased strongly with increasing the pressure. This was due to the intrinsic stress within the films and the change in Ga percentage. Lou et al [11] studied the addition of B to FeGa films to achieve amorphous FeGaB films for microwave applications. They found that 9% B and above gave amorphous films. For 12% B in FeGa films the coercive field was 80 A/m, the anisotropy field was 2 kA/m and the magnetostrictive constant was 70ppm. They didn't investigate further different Fe:Ga ratios nor the effect of sputtering pressure on the magnetic properties. While Ali [7] studied the magnetic properties of FeSiBC films as a function of Ar sputtering pressure. He found that the coercive field strongly depended on the sputter pressure.

The motivation of this work is the study of the effects of Ar pressure (p_{Ar}) on the growth of magnetostrictive amorphous FeSiB and FeGaSiB thin films. This paper presents an investigation of amorphous FeSiB and FeGaSiB films, comparing the result of magnetostriction constants and

magnetic properties as a function of the Ar pressure and Ga addition.

II. Experimental Work

Amorphous FeSiB and FeGaSiB thin films were deposited by a co-sputtering- evaporation system on Si (100) substrates which were cleaned before deposition with acetone and IPA. The amorphous films were made independent of rapid quenching processes. Si substrates were attached to a glass slide using PMMA. After deposition, the samples were cleaned again with acetone and IPA. The composition of the sputter target was $Fe_{85}Si_{10}B_5$. Two types of films were deposited with a thickness 50 nm, sputter power (P_{FeSiB}) of 20W, and the substrate-target distance (d) 60 mm. The first was amorphous FeSiB thin films for a range of Argon gas pressures (p_{Ar}) (4 - 8 μbar). The second was amorphous FeGaSiB thin films with the same pressure conditions above, but with the addition of Ga, evaporated at a constant arbitrary rate, R_{Ga} = 0.2. The rate was measured using a rate monitor, with the evaporation power being varied to achieve a constant rate during growth. The substrate holder was rotated during the growth, to avoid the effect of the sputter gun magnetron field, which can induce anisotropy into the films. In the transverse mode, the magnetic properties of the two film sets were measured on a magneto-optical Kerr effect (MOKE) magnetometer, with the max magnetic field applied being 40 kA/m, which was sufficient to saturate the films. Characterization of the magnetic properties was carried out by measuring the normalized hysteresis loops on unstrained films. The magnetic anisotropy and saturation fields were determined by measuring the normalised hysteresis loops at different magnetic field directions with respect to an edge of the sample, with angles from $0°$ to $180°$. To measure the magnetostriction constant (λ_s) [13] at room temperature, the inverse magnetostrictive effect (Villari effect) [10, 12] measurement was used. This was done by applying a strain to the films using different bend radii and measuring the normalised hysteresis loop for each radius. To analysis the data, the straight line method was used on the data to define the saturation field (H_s) (or anisotropy field (H_k)) [14]. The H_s (H_k) were plotted as a function of bending radii (1/R) and the magnetostriction constant (λ_s) calculated from [13, 14].

$$\lambda_S = \frac{d(H_S)}{d(\frac{1}{R})} \frac{2\mu_\circ M_S (1-v^2)}{3tE} \tag{1}$$

Where, E=130 GPa is the substrate Young's Modulus, t =380 μm is the substrate thickness, and v=0.28 is the substrate poisson ration, $\mu_o M_s$ is the films' saturation induction, and the radii were R=300, 400 and 500mm.

The Bruker D2 phaser model X-ray diffractometer with the Source Cu $K_{\alpha 1}$ (wavelength =1.54184 Å) was used to investigate the film's structure. In the θ/2θ mode, the measurements were taken twice. The first was for the 2θ range from 30° to 80° to study the structure of the films (for bcc Fe 2θ <110> = 44.61° and for bcc Fe-Ga 2θ <110> = 44.29° (10%Ga) to 43.81° (25%Ga) [15] and silicon substrate (peaks at 2θ = 69°

(<004>) and 33° (<200>). The second was for the 2θ range from 35° to 55° to avoid the Si substrates peaks. The DMS model 10 Vibrating sample magnetometer (VSM) was used to measure the magnetic moment of the samples at room temperature, with an external field about 40 kA/m. X-ray Photoelectron Spectroscopy (XPS) using a thermo theta probe was used to determine the compositions of the films. The parameters of the measurement were dwelled time 100 ms (10 scans total of hi res), pass energy of 40 eV, and the source was an X-ray monochromator Al K_α X-ray. For one sample, the measurements were run three times on three different positions on the film surface and an average taken.

III. Results and Discussion

The substrate temperature was around 19℃ for the FeSiB thin film during the growth while it was around 31℃ for FeGaSiB thin films for the variation of pressure (p_{Ar}). This variation in temperature should not change the growth mechanism. It is likely that the increase in temperature is due to the evaporator used during the FeGaSiB thin film growth, as the temperature of evaporation for the Ga atoms was ~1000ºC, while the FeSiB was sputtered physically. Hence the addition of "hot" Ga atoms to the FeSiB films will increase the growth temperature. From the XPS measurements, for all the FeGaSiB films, Ga was detected, meaning that the Ga atoms had enough energy to travel through the plasma, without being scattered.
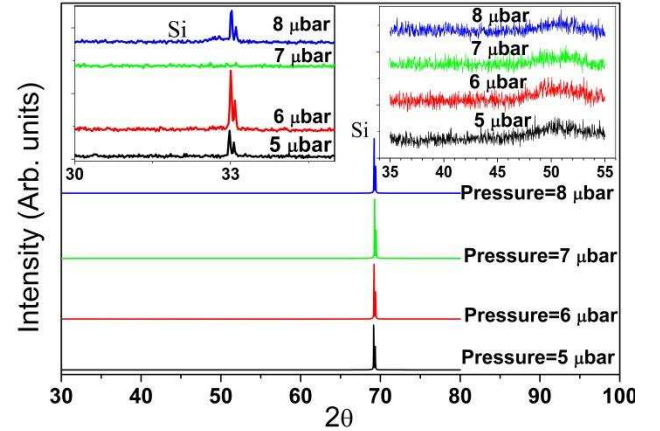
A. XRD Results

Fig. 1. XRD for the different pressures of FeGaSiB thin films. The LHS inset shows the Si peaks. The RHS shows the FeGaSiB film without the Si peaks.

Fig 1 shows the XRD of 50 nm thick FeGaSiB thin films for different (Ar) gas pressures (P_{Ar}). The results showed that all the films had an amorphous structure and all the peaks present were for the Si substrate. The left side insert fig shows the Si peaks at 2θ 33º and the right side insert fig shows the XRD pattern of the FeGaSiB thin films with a 2θ range (35º to 55º) to avoid the substrate peaks. It is clear from the right side insert fig there are no peaks present at 2θ ~ 45º to indicate the existence of crystalline Fe or FeGa. There is a broad peak at 2θ ~ 50º, which is expected for amorphous films. Hence all the

films had an amorphous structure. The results show that the adding of Ga into amorphous FeSiB thin films does not affect the morphology also changing the Ar gas pressure does not affect the film morphology. This means the film morphology is independent of pressure.

B. Magnetic Properties

Fig 2 shows the results of the VSM measurements for FeGaSiB thin films which were measured at room temperature to avoid any thermal effects of high temperatures and to avoid annealing the films. From fig 2, it can be seen that the magnetisation of the FeGaSiB films decreases with increasing pressure. This will be due to the Ar pressure changing the percentage of non-magnetic atoms (Ga, Si, B) within the films, so leading to a change in the composition of films [9, 10]. This will be due to the increase in pressure changing the mean free paths of all the atoms, as Ga is evaporated it is likely to have a higher energy than the sputtered elements, meaning that its mean free paths at higher pressures is longer than the other elements, so increasing the concentration in the films. This will change the local environment within the amorphous films, as the magnetic moment of Fe atoms depends on the nearest neighbours.
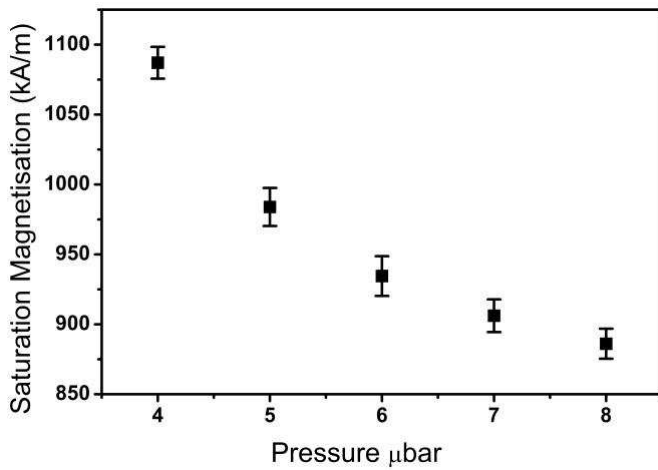

Fig. 2. Saturation magnetisation as a function of pressure for the FeGaSiB films

The anisotropy and saturation fields were determined from the unstrained hysteresis loops measured on the MOKE, as a function of the angle between the applied magnetic field and an edge of the sample. It was found that for the FeSiB films, all the films had uniaxial anisotropy, while the FeGaSiB films were isotropic. As all the films were amorphous, the anisotropy will be dominated by the stress within the films. Thus the FeSiB films have a large intrinsic stress, while the addition of Ga has reduced this stress within the FeGaSiB films. As the anisotropy field can be proportional to the stress in the film and is given by [16, 9]:

$$H_k \approx \frac{2K}{\mu_o M_s} \approx \frac{3\lambda_s \sigma}{\mu_o M_s} \qquad (2)$$

Where K is the anisotropy constant, $\mu_o M$s is the saturation induction and σ is the intrinsic stress.

Fig 3 shows the effect of Ar pressure on the anisotropy/saturation fields for FeSiB and FeGaSiB films. For the FeSiB films, H_k increased gradually as the pressure increased, suggesting that the intrinsic stresses within the films increased with pressure. While for the FeGaSiB films $H_s \approx 4000$ A/m for all pressures. This means the addition of Ga into FeSiB reduced the stress in the films, so giving stability to the saturation field over the pressure range. Fig 4 shows the coercive fields as a function of Ar pressure for FeSiB and FeGaSiB thin films. For both the film sets, H_c is less than 800 A/m, showing that neither pressure nor Ga changed the coercivity. This is expected as the coercive field strongly depends on intrinsic properties such as the grain size, and as both film sets were amorphous, the films will contain no grains.
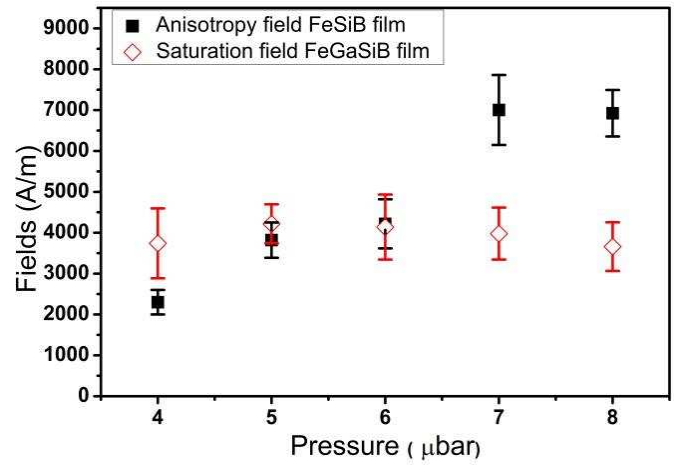

Fig. 3. Saturation/Anisotropy Fields as a function of pressure for the FeSiB and FeGaSiB films
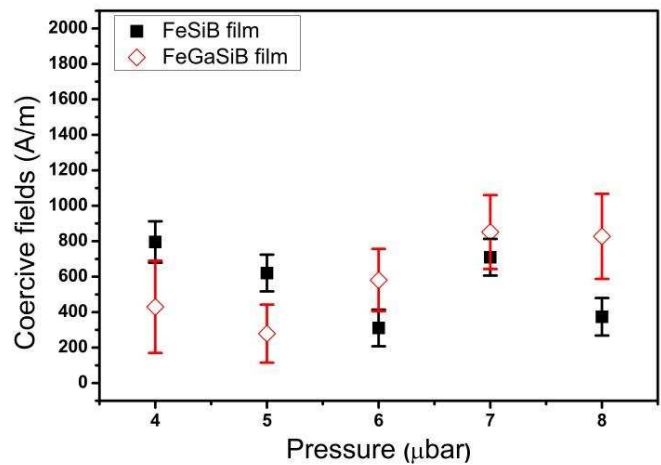

Fig. 4. Coercive Fields as a function of pressure for the FeSiB and FeGaSiB films
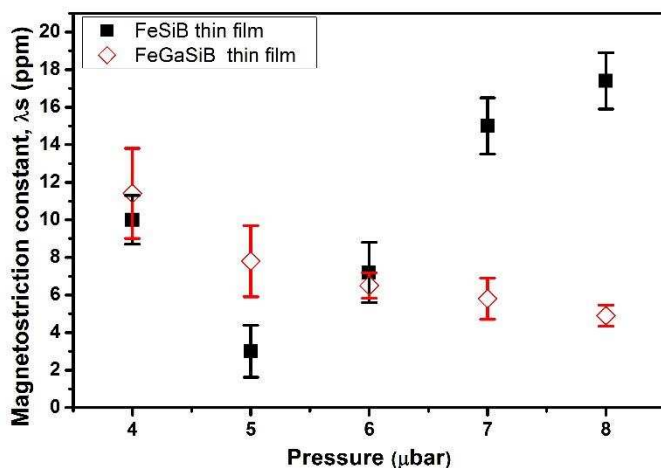
Fig. 5. Magnetostriction constant λ_s as a function of pressure for the FeSiB and FeGaSiB films

From fig 5, it is observed that the magnetostriction constant of the FeSiB thin films increased with increasing pressure. From equation (2), it is observed that the magnetostriction constant, λs, is related to the intrinsic stress in the film. From previous work, the intrinsic stress is tensile as this leads to an increase in elastic properties, hence an increase in the magnetostriction constant, λs,. Thus the increasing pressure causes an increase in tensile stress within the FeSiB films, which increased both the magnetostriction constant λs and anisotropy field. While for the FeGaSiB films, the magnetostriction constant, λs, decreases with an increase of sputtering gas pressure. The films have higher magnetostriction constants, λs, at the lower pressures compared to the FeSiB film, with the maximum λs = 11.4 ppm, at pressure 4μbar. From the anisotropy and saturation field measurements, it is concluded that the addition of Ga reduces the tensile stress within the films, thus these intrinsic stresses no longer dominate the magnetostriction constant, λs,. The reduction in stress is due to the distribution of Ga atoms in the amorphous structure changing the local environment around the Fe atom. This means that the magnetostriction constant, λs, now depends on the saturation magnetization (refer to eqn (1) and (2)), which depends on the Ga concentration (Fig. 2), as both the saturation magnetisation and magnetostriction constant, λs, both decrease with the increase in pressure.

IV. CONCLUSIONS

For FeSiB and FeGaSiB films changing the sputtering pressure and the addition of Ga did not affect the film morphology, as all were amorphous. For the FeSiB films, increasing the pressure increased the tensile stress within the films, which increased the saturation field and magnetostriction constant, and meant all the films had uniaxial anisotropy. Thus the large intrinsic stresses dominated the magnetic properties. The addition of Ga atoms to the FeSiB films, reduced this intrinsic stress within the films, as all the films were isotropic and had the same saturation field. The magnetisation of the

FeGaSiB films decreased as the pressure increased, suggesting an increase in Ga within the films. As the intrinsic stress was reduced in the FeGaSiB films, the magnetostriction constants were no longer dominated by the intrinsic stress, so depended on the films magnetisation. Hence, they decreased with increasing pressure.

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