
Investigation on microstructures and phases of Fe-Ga alloy films deposited by magnetron sputtering

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Abstract: Galfenol (Fe-Ga alloy), as a new magnetostrictive materials, has potentially wide applications in magnetostrictive devices. In this work, Fe-Ga alloy thin films were prepared by slice-style target magnetron sputtering and investigations on microstructures and phases of Fe-Ga alloy films were made to explore the relationship of properties and microstructures. The results show that the component of alloy thin film is related to physical properties of the material itself as well as the area ratio of the patch and target. The phase formed in the films is disorderly A_2 phase with face-centred cubic structure. The films prepared by magnetron sputtering exist in the form of polycrystalline with $\langle 110 \rangle$ crystallographic texture perpendicular to the film plane. The structure shape of as-deposited specimens present a maze domain with different contrast and resolution and the magnetic domain decreased with the increase of the Ga content. With the increase of the Ga content, the magnetic domains become more and more irregular. Fe-Ga thin film morphology is related to the growth mode of the film. The microstructures of Fe-Ga alloy films can be controlled by magnetron sputtering technology.

Keywords: Fe-Ga alloy films; magnetron sputtering; magnetic domain; A_2 phase.

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1 Introduction

There is a developing trend that multicomponent thin films are deposited adopting compound targets. This kind of compound targets can be made by means of defining an alloy target as matrix composition target and inlaying else metal on the substrate (Nakano et al., 2004; Golosov et al., 2012). It is a kind of effective method that limited components are utilised to prepare the alloy thin films with multi-ingredient. However, sputtering effect can be affected by the presence of gaps in the process of drilling and inlaying using traditional mosaic technique.

Fe-Ga alloy giant magnetostrictive film (GMF) has gained more and more attention by people because of its superior integrated properties. So Fe-Ga GMF is expected to overcome shortcomings of other magnetostrictive materials, e.g., Terfenol-D and Fe-Ga alloy (Yan et al., 2013). We have observed growing interest among Fe-Ga alloy films in the possible improvements in manufacturing integrated magnetostrictive devices in the field of industry (Javed et al., 2009; Butera et al., 1998; Seguin et al., 2009). It is known that the thin film performance is determined by its microstructure. Thus, it is very important to obtain good microstructure of thin films for improving the performance of the film (Wenzel et al., 2009). Therefore, the key problem is to deposit thin films in a needed microstructures. It appears that the use of vacuum deposition with appropriate parameters can effectively suppress the DO_3 ordered phase. Systematic studies of the

effective magnetostriction constant as a function of composition support this conclusion. It was found that films of high effective saturation magnetostriction constant and low stress could be fabricated using low Ar pressure, irrespective of sputter power or evaporation rate, giving properties useful for application in microelectromechanical systems (Javed et al., 2010). It is believed that both the (100) texture and the additional modified-DO₃ phase play a positive role in magnetostrictive properties of Fe₇₇Ga₂₃ ribbons (Liu et al., 2015).

Coercivity is mostly controlled by grain size and magnetic anisotropy. Grain size of the films largely depends on film thickness and deposition temperature. Basumatary et al. (2015) report the structure, microstructure and magnetic properties of Fe-Ga thin films deposited using DC magnetron sputtering technique on Si(100) substrate kept at different temperatures. Structural studies employing X-ray diffraction and TEM revealed the presence of only disordered A₂ phase in the film. Columnar growth of nanocrystalline grains from the substrate was observed in the film deposited at room temperature. With increase in substrate temperature the grain size as well as surface roughness was found to increase (Basumatary et al., 2015). Butera et al. (2004) have observed that the symmetry of the in-plane angular variation of the resonance field and the line width are dependent on the film thickness.

Many studies indicated that preparation methods affect magnetic properties as well as magnetostriction of Fe-Ga thin films (Yang et al., 2007). The magnetic domain structure providing fundamental and applied information is the physical base of magnetic materials. The study on microstructure of magnetic domains is very important. Magnetic domains imaging can be used as an effective approach to ascertain the presence, character, and influence of domains or defects in ferromagnetics. Extensive work has been reported on bulk single crystal and polycrystalline Fe-Ga alloys by several groups (Shih et al., 2001; Sun et al., 2004).

Song et al. (2008) observed magnetic domain structure in Terfenol-D. By changing the modulation frequencies, the SEAM can be used as an effective non-destructive method to observe not only the surface topography and domain structure but also the subsurface domain structure and defects.

By now, the study on magnetic domain structure of Fe-Ga alloys films has rarely been reported. Fe-Ga alloys fabricated into thin film form will usually be polycrystalline, and may often possess crystallographic texture. Magnetron sputtering deposition is commonly used to prepare metal and alloy thin films. It is the most common method used to prepare giant magnetostrictive thin films (Qu et al., 2001; Wang et al., 2008). Thin films with different alloying elements are usually prepared using a single element or alloy target sputtering (Ueno et al., 2008; Yan and Zhou, 2007a, 2007b). The metal target containing a single element gallium can be not achieved because metal gallium has a low melting point. Therefore, these alloying films including different compositions are also not prepared employing a single element target sputtering. In addition, it is difficult to develop a series of Fe-Ga targets with certain contents because the smelting of Fe-Ga alloy is quite difficult.

There is a developing trend of preparing multicomponent films by compound targets (referred to be mosaic targets). This kind of compound targets can be made by means of defining an alloy target as matrix targets and inlaying else metal on the substrate (Nakano et al., 2004; Golosov et al., 2012). It is an effective method to utilise limited components to prepare alloying films with multi-ingredient. However, the effect of sputtering can be

affected by the presence of gaps in the process of drilling and inlaying using traditional mosaic technique.

In this work, Fe-Ga alloy thin films were prepared by slice-style target magnetron sputtering. Effects of microstructure and composition of thin films have been investigated to obtain alloying thin films with good performance.

2 Experiment

In the experiments, the direct current (DC) magnetron sputtering equipment (JZCK-600F) was used to prepare the Fe-Ga alloy film. $\text{Fe}_{81}\text{Ga}_{19}$ alloy target was conducted to prepare $\text{Fe}_{100-x}\text{Ga}_x$ ($15 \leq x \leq 20$) thin films with different thickness (1,440 nm to 1,540 nm). The experimental parameters included 2×10^{-4} Pa vacuum degree, 0.6 Pa working pressure (Ar), 90 W sputtering power and 1.5 hours sputtering time.

During the process of deposition, polished glass was acted as the substrate. Ultrasonic before sputtering was performed to clean the substrate surface. The distance between the target and the substrate was chosen as 100 mm. Some iron slices, which had a diameter of 60 mm and a thickness of 3 mm, were attached to the etching area of round alloy target under the magnetism of the target. And the area ratio of iron slices to alloy target was changed to control further the composition of Fe-Ga thin films. The purity of iron slices was up to 99.99%. Two to five discs with a diameter from 1.6 mm to 2.5 mm and 1 mm thickness were distributed uniformly along the target diameter of 42 mm.

The film morphology was observed using quanta 200 scanning electron microscopy. Furthermore, Oxford energy dispersion spectroscopy was also conducted to investigate the average composition. Bruker AXS D8 ADVANCE X-ray diffractometer was used to analyse the film phase. The magnetic hysteresis loop of Fe-Ga thin film was measured by a vibrating sample magnetometer (VSM).

3 Results and discussion

3.1 Magnetic domain structure

Magnetic properties of a specimen deposited at a certain temperature are generally determined by the domain structure. Moreover, magnetic domain spacing is one of important parameters for domain structure. Domain structure of alloy $\text{Fe}_{81}\text{Ga}_{19}$ ($\alpha = 0$) and $\text{Fe}_{84}\text{Ga}_{16}$ ($\alpha = 3.1$) are shown in Figures 1 and 2, respectively.

In Figures 1 and 2, the white area and black area denote the attractions and repulsions between samples and magnetic probe, separately. In terms of the magnetic probe with vertical magnetisation, it is very sensitive to the magnetic domain structure along the direction perpendicular to the thin film. However, it is insensitive to the magnetic domain structure along the direction parallel to the Fe-Ga film. The structure shape of as-deposited specimens presents a maze domain with different contrast and resolution. It can be observed from Figures 1 and 2 that magnetic domain of thin films decreases with increasing the content of Ga element. Additionally, it has been found that with increasing Ga content, the size of magnetic domain becomes small and morphology of magnetic

domain becomes more and more irregular, indicating that the magnetic domain is becoming more and more uneven.

Figure 1 Domain structure of alloy Fe₈₁Ga₁₉ ($\alpha = 0$) (see online version for colours)

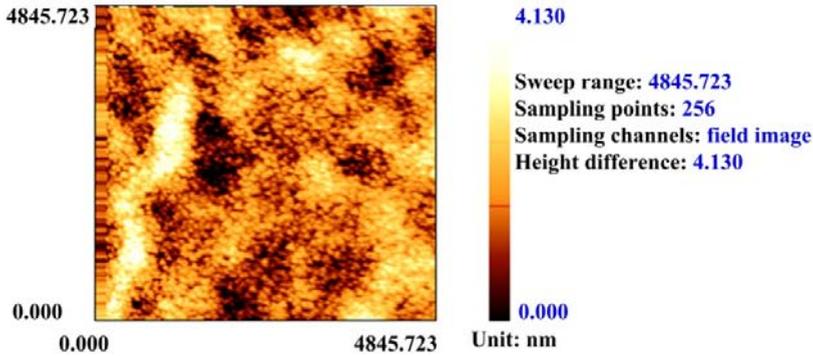
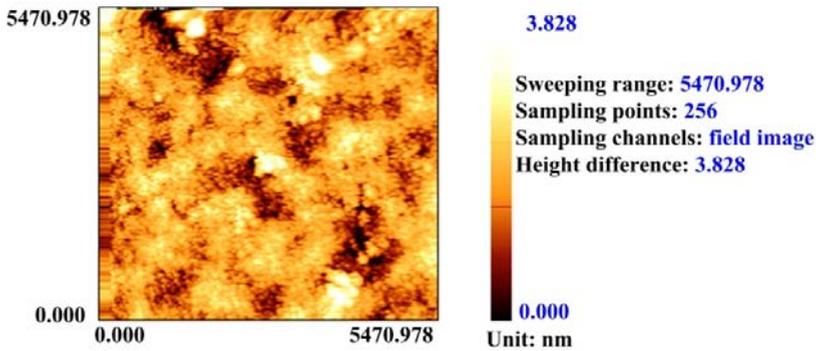


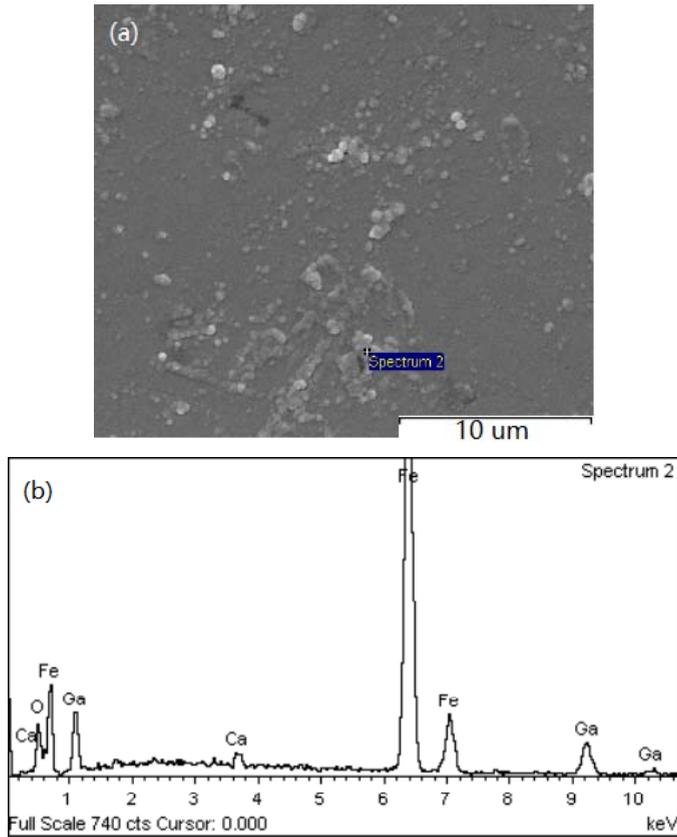
Figure 2 Domain structure of alloy Fe₈₄Ga₁₆ ($\alpha = 3.1$) (see online version for colours)



3.2 Composition

Figure 3 shows the morphology and composition of the Fe-Ga thin film. It can be observed that Ga content in white specks is higher than that in grey substrates. The depositions can be also identified as the Ga-rich phases.

When preparing the thin film by using magnetron sputtering, the morphology of the film depends on two factors, including the roughness of the substrate and the growth mode of the film. As shown in Figure 3, the reason for forming the white dot on the Fe-Ga thin film surface is related to the growth mode of the film. On the one hand, low-energy atoms that can be adsorbed by the film break away from the matrix on account of the impact of subsequent atoms with high energy. During depositing the film process, the pits can be generated because of failing to supplement low-energy atoms on the surface of the film. On the other hand, the presence of the morphology may be associated with the surface of the substrate. If the glass substrate has an uneven surface and larger roughness, the deposition location of the film may form the pits.

Figure 3 Morphology and EDS diagram of Fe-Ga thin film (see online version for colours)

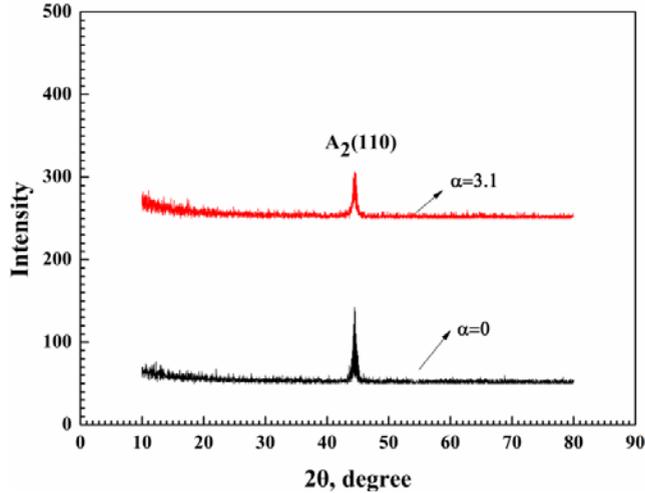
3.3 XRD analysis

To further characterise the phase of Fe-Ga alloy thin films, XRD analysis was carried out under specified conditions. Figure 4 shows the XRD patterns of the as-deposited thin films.

It can be viewed from Figure 3 that the intensity of the XRD peak is weak for as-deposited Fe-Ga alloy thin films. In order to guarantee the fine grains and microstructure in the experiment, a method was performed to prepare alloy thin films using low sputtering energy, repeated and transitory sputtering. The experimental parameters are as followed, including 90 W power and five min intervals every time. It has been found that the substrate temperature was not high as a result of a larger target distance set in this experiment. A mixed structure containing part of crystal structures and amorphous structures were produced because the diffusion ability of atoms was too weak to finish completely the crystallisation process of granular films when conducting sputtering deposition. If heated in the subsequent process, most of the stress formed within the alloy thin films could be eliminated. In addition, the atoms within the films can be migrated and segregated again under the action of a driving force produced by the temperature and residual stress. A long-range ordering for atoms inside the films occurred when the annealing temperature reached a certain critical value (crystal

transition temperature). The diffraction peak intensity was related to the thin film structure. A_2 diffraction peak intensity began to increase when the thin film structure changed from amorphous state to crystalline state. With annealing temperature rising, the intensity of A_2 would be strengthened and crystallites would grow up gradually.

Figure 4 XRD patterns of the as-deposited thin films (see online version for colours)

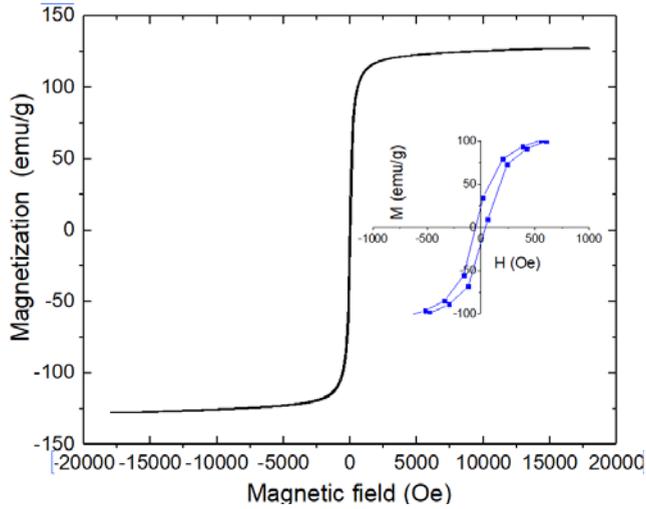


Based on the XRD results, we observed that all thin films are in polycrystalline form with $\langle 110 \rangle$ crystallographic texture perpendicular to the film plane. The phase that generated in the alloyed films was a disorderly A_2 phase with body centred cubic structure. In addition, no higher-order peaks have been noticed.

Frankly speaking, multifarious phase structures, for example, A_2 , DO_3 and so on, may be produced during depositing Fe-Ga films. However, no evidences can demonstrate the occurrence of the DO_3 phase from above detection. The reason for this phenomenon is that the peaks may be too weak to be observed. It can be also noticed from Figure 3 that the position of the body-centred cubic (bcc) $\langle 110 \rangle$ peak shifts slightly towards higher values of 2θ with an increase in the value of α . It can contribute to an increase in the content of Ga with increasing the value of α . In this case, the Ga content within the Fe-Ga alloy thin films will decrease, resulting in reducing the number of A_2 phase. According to the centroid of the $\langle 110 \rangle$ peaks, the lattice parameter of each film can be evaluated. It can be proved that the lattice parameter can increase with decreasing the value of α .

3.4 Magnetic properties

The magnetic hysteresis curve is given in Figure 5. The saturation magnetisation of the proposed thin film is 124.47 emu/g, when applying a 15.3 kOe magnetic field. The details of the hysteresis loop is shown in the little figure, the coercive force H_c is 48 Oe. So the hysteresis width can be determined as 96 Oe. The proposed thin film displays a large saturation magnetisation and narrow magnetic hysteresis loop.

Figure 5 Magnetic hysteresis curve of Fe-Ga thin film (see online version for colours)

4 Summary

In this work, we prepared a Fe-Ga thin film and investigated its microstructure and magnetic properties. Some conclusions of this study are summarised as follows:

- 1 All films are in polycrystalline form with $\langle 110 \rangle$ crystallographic texture perpendicular to the film plane. The phase formed in the alloyed films is a disorderly A_2 phase with body centred cubic structure.
- 2 The domain structure of Fe-Ga alloy thin films is a maze domain. With increasing Ga content, the size of magnetic domain becomes small and morphology of magnetic domain becomes more and more irregular. Fe-Ga thin film morphology was associated with to the growth mode of the film.
- 3 It can contribute to an increase in the content of Ga with increasing the value of α . In this case, the Ga content within the Fe-Ga alloy thin films will decrease, resulting in reducing the number of A_2 phase. According to the centroid of the $\langle 110 \rangle$ peaks, the lattice parameter of each film can be evaluated. It can be proved that the lattice parameter can increase with decreasing the value of α .
- 4 The Fe-Ga thin film performs good magnetic property in our work.

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