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Magnetic and magnetostrictive properties of amorphous TbFe/FeAl multilayer thin film

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Abstract: Exchange coupling multilayer thin films, which combined giant magnetostriction and soft magnetic properties, were of growing interest for applications. The TbFe/FeAl multilayer thin films were prepared by dc magnetron sputtering onto glass substrates. The microstructure, magnetic, and magnetostrictive properties of TbFe/FeAl multilayer thin film was investigated at different annealing temperatures. The results indicated that the soft magnetic and magnetostrictive properties for TbFe/FeAl multilayer thin film compared with TbFe single layer film were obviously improved. In comparison with the intrinsic coercivity $_{J}H_{c}$ of 59.2 kA/m for TbFe/FeAl multilayer film, the intrinsic coercivity $_{J}H_{c}$ of TbFe/FeAl multilayer thin films rapidly dropped to 29.6 kA/m. After optimal annealing (350 °C×60 min), magnetic properties of H_{s} =96 kA/m and $_{J}H_{c}$ =16 kA/m were obtained, and magnetostrictive coefficient could reach to 574×10⁻⁶ under an external magnetic field of 400 kA•m⁻¹ for the TbFe/FeAl multilayer thin film.

Keywords: exchange coupling; multilayer; TbFe/FeAl; magnetostrictive coefficient; rare earths

Magnetostrictive thin films are particularly promising as microactuator elements like cantilevers or membranes because they combine high-energy output, high-frequency, and remote-control operations^[1–3]. Due to this potential, interest in such giant magnetostrictive thin films has rapidly grown over the past few years^[4–6].

Recently, high magnetostriction combined with soft magnetic properties has been proposed based on spring type magnets^[7–9], which consist of two materials with different magnetic properties, exchange coupling in a multilayer system. Combination of giant magnetostrictive materials and soft magnetic materials show not only good softness but also it is expected to give novel magnetostrictive behavior.

In this study, FeAl was selected as a soft magnetic material because it is known to show higher magnetic susceptibility and larger magnetostriction at low magnetic fields than pure Fe film^[10]. The aim of this work is to fabricate heterogeneous multilayer systems consisting of a hard magnetic amorphous TbFe phase (with high magnetostriction) and a soft magnetic amorphous FeAl phase. Finally, the effect of vacuum annealing on microstructure and magnetic characteristics was investigated.

1 Experimental

Tb_{0.41}Fe_{0.59} single layer, Fe_{0.83}Al_{0.17} single layer, and a

typical {(Tb_{0.41}Fe_{0.59})(10 nm)/(Fe_{0.83}Al_{0.17})(8 nm)}₅₀ multilayer thin films were magnetron sputtered on 25 mm×25 mm×1 mm polished glass substrates using Φ 60 mm composite-type targets in an FJL560D2 device and a rotary turntable technique in a stop-and-go-mode without intentional substrate heating. The base pressure was 1.4×10^{-4} Pa. The amorphous Tb_{0.41}Fe_{0.59} layer was dc magnetron sputtered using a sputtering power of 60 W, Ar pressure of 0.6 Pa, and the target-to-substrate distance of 100 mm, whereas the amorphous Fe_{0.83}Al_{0.17} layer was dc magnetron sputtered using a sputtering power of 80 W, Ar pressure of 0.6 Pa, and the target-to-substrate distance of 100 mm. These conditions resulted in deposition rates of 0.21 and 0.13 nm/s for the Tb_{0.41}Fe_{0.59} and Fe_{0.83}Al_{0.17} layers, respectively. The individual layer thickness were $t_{\text{TbFe}}=10$ nm and $t_{\text{FeAI}}=8$ nm, the bilayer periodicity was n=50. The deposited {Tb_{0.41}Fe_{0.59}/Fe_{0.83} $Al_{0.17}$ multilayer thin film was subsequently annealed at temperatures ranging from 250 to 450 °C with an interval of 100 °C for 1 h under vacuum ($<10^{-3}$ Pa). The composition of the deposited film was verified using energy-dispersive X-ray diffraction (EDS). The amorphous and polycrystalline structures of the layers were verified using X-ray diffraction. The magnetization was measured using a vibrating sample magnetometer under magnetic fields up to 1440 kA/m at room temperature. The magnetostrictive properties were measured by deflectometry using cantilever method.

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2 Results and discussion

Fig.1 shows XRD patterns of $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film as-deposited and vacuum annealed at different temperatures. It can be observed that the as-deposited multilayer thin film is mainly composed of a mixture of an amorphous phase (Halo patterns were observed at near 2θ =30°–40°) and crystalline phase. Here, the high X-ray diffraction intensity at 2θ =45° is characteristic of the (110) reflection of bcc-Fe phase. According to Scherrer formula, it is estimated that the mean grain size of bcc-Fe is about 10 nm. Also, it can be observed that Halo patterns remain almost the same at all temperatures, whereas the (110) peak broadenings of bcc-Fe phase reduced with increasing annealing temperature. When the annealing temperature is below 450 °C, the grain size of bcc-Fe increases with increasing annealing temperature but remained under 20 nm.

Fig.2 illustrates the efficacy of the exchange coupling mechanism in reducing the driving magnetic field, using the TbFe/FeAl system. In Fig.2, the in-plane magnetization behavior of as-deposited $Tb_{0.41}Fe_{0.59}$ single layer and { $(Tb_{0.41}Fe_{0.59}/Fe_{0.59})$



Fig.1 XRD patterns of $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer (asdeposited and annealed at different temperatures) thin film



Fig.2 In-plane hysteresis loops of as-deposited (1) $Tb_{0.41}Fe_{0.59}$ single layer; (2) $Fe_{0.83}Al_{0.17}$ single layer; (3) typical { $Tb_{0.41}Fe_{0.59}$ / $Fe_{0.83}Al_{0.17}$ } somultilayer thin films

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Fe_{0.83}Al_{0.17}}₅₀ multilayer thin films is shown along with the hysteresis loop of a high moment as-deposited Fe_{0.83}Al_{0.17} single layer thin film. A high field is required to magnetize the as-deposited Tb_{0.41}Fe_{0.59} single layer thin film; in contrast, the as-deposited multilayer thin film can be saturated at much lower fields, much like the as-deposited Fe_{0.83}Al_{0.17} single layer thin film. In addition, the magnetization of as-deposited multilayer thin film shows higher maximum value than the two single layer thin films.

The comparison between multilayer thin film and Tb-Fe single layer thin film reveal increased saturation magnetization M_s of the multilayer thin film combined with reduced plane intrinsic coercivity $_{J}H_{c}$. In comparison with the intrinsic coercivity _JH_c of 59.2 kA/m for Tb_{0.41}Fe_{0.59} single layer film, the intrinsic coercivity $_{J}H_{c}$ for {Tb_{0.41}Fe_{0.59}/ Fe_{0.83}Al_{0.17}}₅₀ multilayer thin film drops rapidly to 29.6 kA/m. This is due to its field dependent micromagnetic structure^[11]. Following saturation in the negative direction, reversal in the multilayer thin film occurs by nucleation of "twin" domain walls. As the field strength increases, magnetization reversal occurs by lock-step motion of this twin wall across the length of the sample. The stray field emanating out of the Néel wall causes magnetization fluctuation in adjacent layers, giving rise to quasi-Néel wall. By forming such twin walls, the stray fields emanating from each domain wall are able to close its flux by magnetostatic locking-in with the stray fields from domain wall in adjacent layers, thereby leading to an overall decrease in the wall energy and coercivity. It is known that the replacement of a small fraction of Fe atoms by nonmagnetic element of Al can lower magnetocrystalline anisotropy and reduce magnetostrictive anomalies. This is caused by the enlargement of lattice parameter by substitution of Al atoms into Fe (bcc) lattice^[12].

Fig.3 shows the in-plane hysteresis loops of as-deposited and annealed $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film. It can be observed that both the as-deposited and annealed



Fig.3 Magnetic hysteresis loops of as-deposited and annealed $\{Tb_{0.41}Fe_{0.59}/\ Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film

precipitated bcc-Fe grains (Fig.1). In the case of $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film, the saturation magnetic field H_s and intrinsic coercivity $_JH_c$ first decrease to minimum values and then increase (Fig.4). After annealing at 350 °C for 60 min, the saturation magnetic field and intrinsic coercivity show a minimum value of 96 and 16 kA/m, respectively. This can be explained by the relaxation of internal stress during the first stage of annealing. Moreover, the thickening of the soft magnetic layers should also play a role in the softening of magnetic properties of the samples by the enhancement of total magnetization. Such enhancement of the magnetization is usually related to coercivity decrease in exchange coupling of multilayer thin film^[13].

increased ferromagnetic Fe-Fe interactions, arising from

The comparison between the in-plane magnetostriction coefficient of as-deposited and annealed {Tb_{0.41}Fe_{0.59}/ $Fe_{0.83}Al_{0.17}$ multilayer thin film (Fig.5) confirms the obtained magnetization results of the same series. Under an external magnetic field of 400 kA/m, the magnetostrictive coefficient of {Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}}₅₀ multilayer thin film first increases to maximum values and then decrease. After annealing at 350 °C for 60 min, the magnetostrictive coefficient shows a maximum value of 574×10^{-6} . The magnetostrictive coefficient of the {Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}}₅₀ multilayer thin films mainly depend on the stress state between film and substrate, composition and thickness of the different layers with the high ferromagnetic exchange coupling between hard and soft magnetic phases at the interfaces. When the annealing temperature is low, annealing releases the compressive stress, but there still exists some residual



Fig.4 Saturation fields (H_s) (1) and intrinsic coercivity $(_JH_c)$ (2) as a function of annealing temperature for $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film

compressive stress owing to the difference of the thermal expansion coefficient between the multilayer thin films and glass substrate. The presence of compressive stress is believed to weaken the exchange coupling between the hard and soft magnetic phases. As a result of the altered stress state, annealing leads to an in-plane magnetic easy axis with reduced values for the magnetic saturation and coercive fields (Fig.4). The optimum data of the $\{Tb_{0.41}Fe_{0.59}/$ Fe_{0.83}Al_{0.17}}₅₀ multilayer thin films being annealed at 350°C reflect the almost stress-free state of the multilayer thus vanishing all stress induced anisotropies, the absence of any significant diffusion under these conditions and the onset of nanocrystallization in the TbFe layers leading to an increase in the saturation magnetostrictive coefficient. The reasons for the reduction of magnetostrictive coefficient for annealing at 450 °C are that a recrystallization effect of iron occurs at the interfaces, thickening the soft magnetic layer, and would lead to various compositions of the different layers. The soft magnetic layer thickness is kept up the domain wall thickness to accelerate the formation of domain wall parallel to the interfaces, whose presence would otherwise lead to a substantial reduction in magnetostrictive coefficient.



Fig.5 Magnetostriction of as-deposited and annealed $\{Tb_{0.41}Fe_{0.59}/Fe_{0.83}Al_{0.17}\}_{50}$ multilayer thin film

3 Conclusion

The soft magnetic and magnetostrictive properties for { $(Tb_{0.41}Fe_{0.59})(10 \text{ nm})/(Fe_{0.83}Al_{0.17})(8 \text{ nm})$ }₅₀ multilayer thin film as compared with those of the TbFe single layer were obviously improved. In comparison with the intrinsic coercivity $_JH_c$ of 59.2 kA/m for Tb_{0.41}Fe_{0.59} single layer film, the intrinsic coercivity $_JH_c$ for { $(Tb_{0.41}Fe_{0.59})(10 \text{ nm})/(Fe_{0.83}Al_{0.17})$ (8 nm)}₅₀ multilayer thin film dropped rapidly to 29.6 kA/m. After optimal annealing (350 °C×60 min), magnetic properties of H_s =96 kA/m and $_JH_c$ =16 kA/m were obtained and magnetostrictive coefficient could reach to 574×10⁻⁶ under 400 kA/m external magnetic field for the TbFe/FeAl multilayer thin film.

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