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Research Article

Microstructure and Magnetic Properties of NdFeB Films through Nd Surface Diffusion Process

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Ta/Nd/NdFeB/Nd/Ta films were deposited by magnetron sputtering on Si (100) substrates and subsequently annealed for 30 min at 923 K in vacuum. It was found that the microstructure and magnetic properties of Ta/Nd/NdFeB/Nd/Ta films strongly depend on the NdFeB layer thickness. With NdFeB layer thickness increasing, both the grain size and the strain firstly reduce and then increase. When NdFeB layer thickness is 750 nm, the strain reaches the minimum value. Meanwhile, both the in-plane and perpendicular coercivities firstly drastically increase and then slowly decrease with NdFeB layer thickness increasing. The highest in-plane and perpendicular coercivities can be obtained at NdFeB layer thickness of 750 nm, which are 21.2 kOe and 19.5 kOe, respectively. In addition, the high remanence ratio (remanent magnetization/saturation magnetization) of 0.87 can also be achieved in Ta/Nd/NdFeB (750 nm)/Nd/Ta film.

1. Introduction

NdFeB permanent magnetic films have drawn extensive attention due to their excellent hard magnetic properties and potential applications in microelectromechanical system (MEMS), micromagnetic devices, and magnetic recording media [1–6]. The relatively low coercivity and poor thermal stability are the practical obstacle for applications of NdFeB films. One major approach to improve the properties is to increase the coercivity at the room temperature, which can suppress demagnetization at a higher operating temperature.

To increase the coercivity of NdFeB films, many researches have been reported. Fukagawa et al. reported that after sputtering Nd metal to the surface of NdFeB magnet and subsequent annealing an fcc interfacial phase was formed between the surface grains and the Nd layer, resulting in the recovery of surface coercivity [7]. Kim et al. reported that by Nd element diffusing from Nd layer into NdFeB layer high coercivity was achieved at the thickness ratio of Nd/NdFeB ≥ 1. However, a rather low content of NdFeB hard magnetic phase existed in the films [8]. Li et al. reported that the coercivity of $[NdFeB/Nd]_4$ films was fairly increased because that Nd element effectively diffused [9].

In this work, Ta/Nd/NdFeB/Nd/Ta films were prepared on Si (100) substrates by magnetron sputtering. The effect of the NdFeB layer thickness on the microstructure and magnetic properties of Ta/Nd/NdFeB/Nd/Ta films was systematically investigated. The high in-plane and perpendicular coercivities can be obtained in Ta/Nd/NdFeB (750 nm)/Nd/Ta film, which are 21.2 kOe and 19.5 kOe, respectively. The high remanence ratio of 0.87 can also be achieved in the film.

2. Experimental Procedure

Ta/Nd/NdFeB/Nd/Ta films were prepared by FJL560II ultrahigh vacuum magnetron sputtering system on Si (100) substrates. Here, Nd layer thickness was fixed at 250 nm. A Ta underlayer of 60 nm and Ta coverlayer of 60 nm were used to suppress the oxidation of NdFeB films. Pure Nd (99.9%) and Ta (99.95%) targets were used. The target for NdFeB layer was a commercial N33H NdFeB sintered target attached to B-chips. The base pressure of the deposition chamber was 2.0×10^{-4} Pa and high purity Ar gas was introduced during sputtering. The composition of NdFeB layer was determined to be $Nd_{10.73}Fe_{84.01}B_{5.26}$ by a Thermo System 7 energy dispersive spectrometer (EDS). The as-deposited

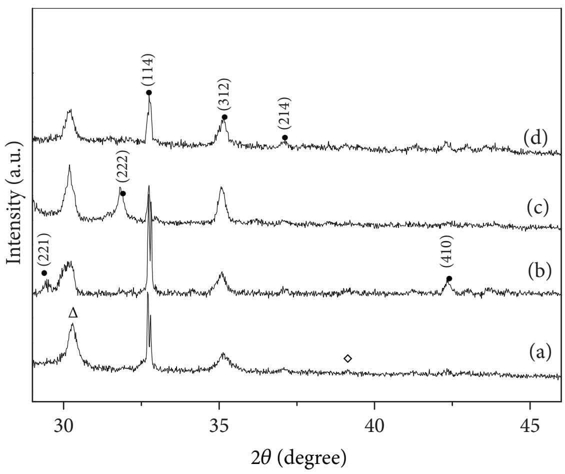

FIGURE 1: XRD patterns of Ta/Nd/NdFeB (x nm)/Nd/Ta ((a) x = 450; (b) x = 600; (c) x = 750; (d) x = 900) films.

films were subsequently annealed for 30 min at 923 K in vacuum.

The structure of the films was analyzed by Bruker-D8 X-ray diffraction (XRD) with Cu Kα radiation. The thickness was characterized by a JSM-7001F field emission scanning electron microscope (FE-SEM). The magnetic properties were measured by a Quantum Design vibrating sample magnetometer (VSM) with a maximum applied field of 30 kOe.

3. Results and Discussion

Figure 1 shows the XRD patterns of Ta/Nd/NdFeB (x nm)/Nd/Ta (x = 450, 600, 750, 900) films. The prominent characterization peaks of (222), (114), (312), and (410) can be obviously seen in the XRD patterns, indicating the formation of tetragonal $Nd_2Fe_{14}B$ phase. In addition to the above-mentioned peaks, Nd peaks are also clearly observed in the XRD patterns, due to the existence of Nd layer.

XRD can be utilized to evaluate peak broadening with crystallite size and lattice strain due to dislocation [10]. Williamson-Hall (W-H) analysis [11] considers that the contributions to line broadening of the crystallite size and lattice strain are independent of each other and both have a Cauchy-like profile; the final line breadth is the sum

$$\beta_{hkl} = \beta_S + \beta_D, \quad (1)$$

$$\beta_{hkl} = \left(\frac{k\lambda}{D\cos\theta}\right) + (4\varepsilon\tan\theta). \quad (2)$$

Rearranging (2), we get

$$\beta_{hkl}\cos\theta = \left(\frac{k\lambda}{D}\right) + (4\varepsilon\sin\theta), \quad (3)$$

where D is the average grain size, K is the shape factor (0.9), λ is the wavelength of Cuk$_\alpha$ radiation, and ε is the strain. The strain was assumed to be uniform in all crystallographic directions. As shown in Figure 2, $\beta\cos\theta$ was plotted with respect to $\sin\theta$ for the $Nd_2Fe_{14}B$ peaks of Ta/Nd/NdFeB/Nd/Ta films. The grain size and strain were calculated from the y-intercept and slope of the fitted line, respectively. Figure 3 shows the variation of the grain size and strain with NdFeB layer thickness in Ta/Nd/NdFeB/Nd/Ta films. With NdFeB layer thickness increasing from 450 nm to 900 nm, both the grain size and the strain firstly reduce and then increase. When the NdFeB layer thickness is 750 nm, the strain reaches the minimum value, which is equal to 0.0016.

Figure 4 shows the dependence of the coercivity on the NdFeB layer thickness in Ta/Nd/NdFeB/Nd/Ta films. With NdFeB layer thickness increasing from 450 nm to 750 nm, both the in-plane and perpendicular coercivities drastically increase. However, when NdFeB layer thickness further increases to 900 nm, both the in-plane and perpendicular coercivities slowly decrease. When NdFeB layer thickness is 750 nm, both the in-plane and perpendicular coercivities reach the maximum, which are 21.2 kOe and 19.5 kOe, respectively. It can be interpreted that the strain is minimized at the NdFeB layer of 750 nm, which is favorable for the crystallization of NdFeB.

Figure 5 shows the in-plane and out-of-plane hysteresis loops of Ta/Nd/NdFeB (750 nm)/Nd/Ta film. As seen in Figure 5, the in-plane and out-of-plane coercivities reach 21.2 kOe and 19.5 kOe, respectively. The high remanence ratio of 0.87 can also be noticed in the in-plane hysteresis loop, which is also important for the application of NdFeB film. It is very interesting that the out-of-plane hysteresis loop shows a kink near the original point, implying the existence of uncoupled soft magnetic grains, which becomes more significant while the external field is applied along the out-of-plane direction [12]. Moreover, the out-of-plane hysteresis loop shows two stage types of initial magnetization behavior with the first magnetization of high susceptibility followed by the second one of low susceptibility, which also suggests the independent magnetization of the uncoupled soft magnetic grains. Zhao et al. proposed that Fe is very likely to occur in $Nd_2Fe_{14}B$ so that many single-phased materials are in fact the two-phased ones [13].

The micromagnetic model was applied to clarify the coercivity mechanisms in permanent and composite materials. Coercivity is a linear function of H_A [14]:

$$H_{Ci}(T) = \alpha H_A(T) - N_{\text{eff}} M_s(T). \quad (4)$$

Here, H_A is the anisotropy field, $H_A = 2K_1/M_s$, which is the ideal coercivity for the coherent rotation of magnetically isolated single domain particles. M_s is the saturation magnetization. α and N_{eff} are the microstructure-dependent parameters. The parameter α describes the reduction in the anisotropy field due to the presence of crystallographic defects in the magnetically inhomogeneous region on the grain surface and misalignment of the grains. The parameter N_{eff} describes the local demagnetization field, which assists nucleation of the reversed domain under the action of the applied inverse field. The temperature dependence of K_1 is obtained from Durst and Kronmüller [15]. Figure 6 shows

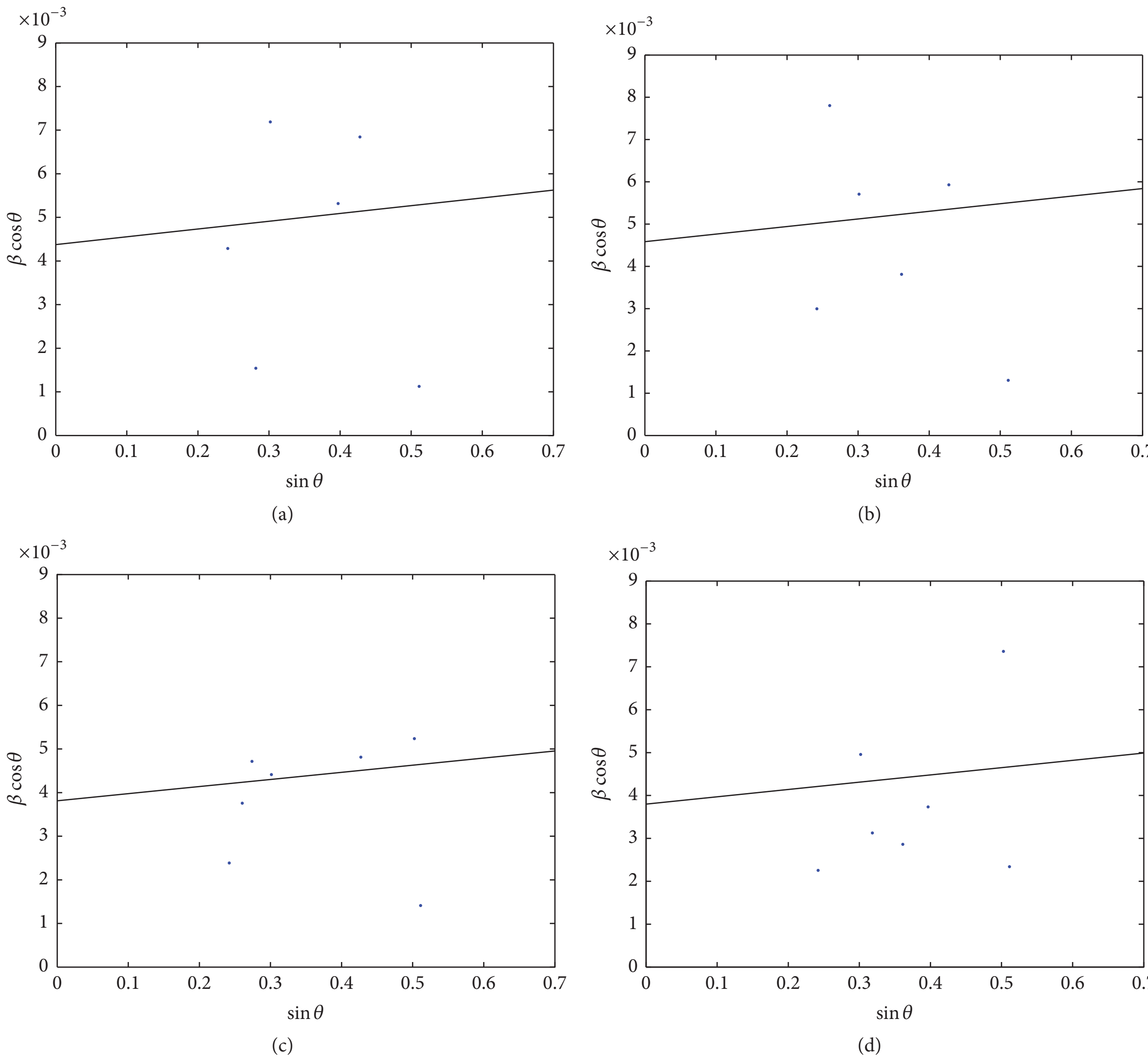

Figure 2: Plot of $\beta \cos\theta$ versus $\sin\theta$ of Ta/Nd/NdFeB (x nm)/Nd/Ta ((a) $x = 450$; (b) $x = 600$; (c) $x = 750$; (d) $x = 900$) films.

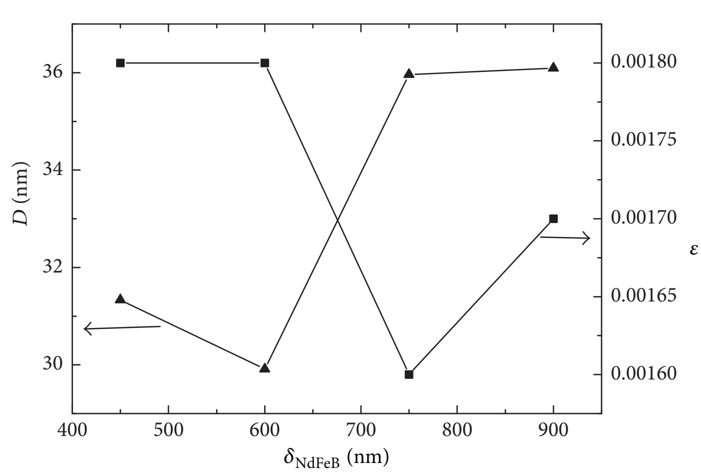

Figure 3: Variation of the grain size and strain with NdFeB layer thickness in Ta/Nd/NdFeB/Nd/Ta films.

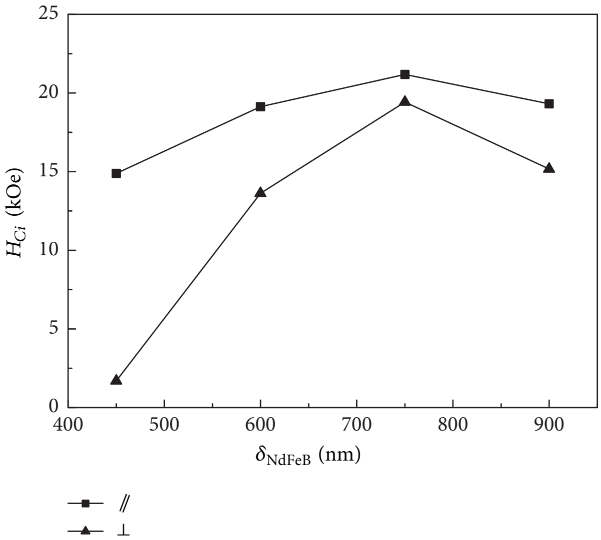

Figure 4: Dependence of the coercivity on the NdFeB layer thickness in Ta/Nd/NdFeB/Nd/Ta films.

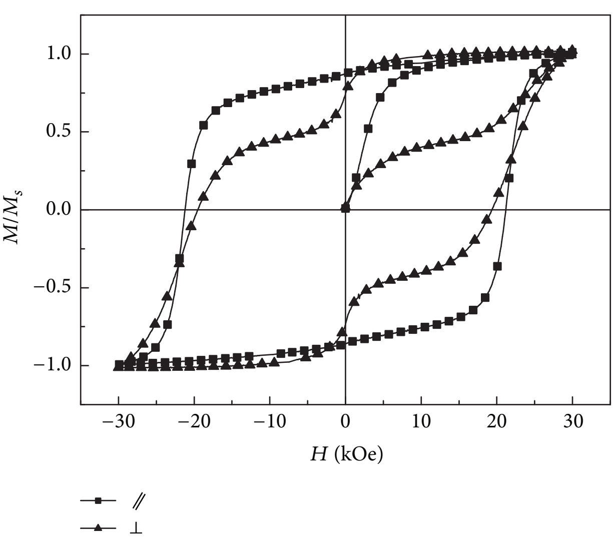

Figure 5: Hysteresis loops of Ta/Nd/NdFeB (750 nm)/Nd/Ta film.

the dependence of H_{Ci}/M_s on H_A/M_s for Ta/Nd/NdFeB (x nm)/Nd/Ta ($x = 450, 600, 750, 900$) films. The micromagnetic parameters α and N_{eff} were fitted by the least squares method and are shown in Figure 6. It can be noticed that α for Ta/Nd/NdFeB (750 nm)/Nd/Ta film is 0.257, which is larger than those of other Ta/Nd/NdFeB/Nd/Ta films. It suggests a decrease in the size of the distorted region and/or a considerable release of interfacial misfit at the NdFeB layer of 750 nm [16]. Zhao et al. proposed a self-pinning coercivity mechanism, incorporating elements of both initial local nucleation process and subsequent propagation of the domain wall to the main phase [13, 17]. Such a mechanism was firstly proposed in hard-soft multilayers and then extended to composite and permanent nanomagnets. The defects in the so-called single-phased permanent magnets act as the nucleation and pinning centers, which plays a role similar to the soft phase in hard-soft composite systems. Such

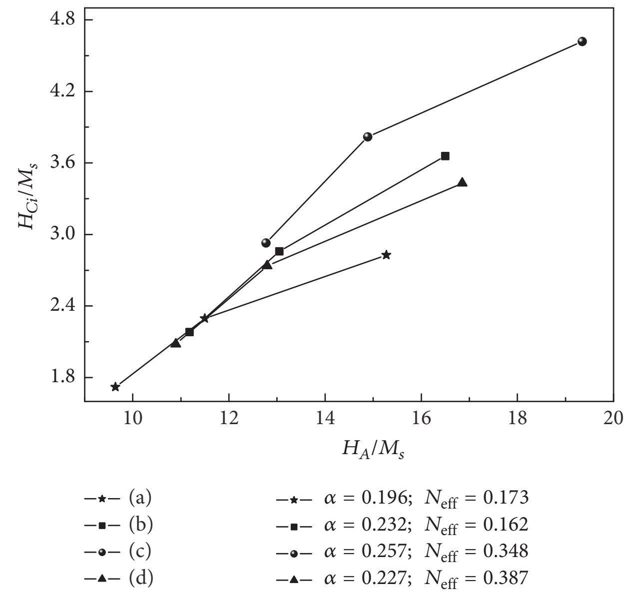

Figure 6: Dependence of H_{Ci}/M_s on H_A/M_s for Ta/Nd/NdFeB (x nm)/Nd/Ta ((a) x = 450; (b) x = 600; (c) x = 750; (d) x = 900) films.

self-pinning is attributed to the change of the intrinsic parameters associated with the phase change at the interface. In particular, for sufficiently large soft grains/defects, the pinning field can be expressed as $H_P = \alpha H_K$, where $H_K = 2k/M_s$ is the anisotropy field and α depends on the material parameters and micromagnetic structures. The coefficient α decreases as the volume occupation of the soft phase increases. For an exchange-coupled $Nd_2Fe_{14}B$-αFe system with abrupt change of parameters in the interface, $\alpha = 0.1$. For the permanent magnets in which small amount of soft grains exists, $\alpha = 0.2$–0.3. As is shown in Figure 6, the α value is between 0.196 and 0.257, which is consistent with the theoretical value of the permanent magnets, indicating the existence of soft grains. It is accordant with the results concluded from Figure 5.

4. Conclusions

In summary, the microstructure and magnetic properties of Ta/Nd/NdFeB/Nd/Ta films are strongly dependent on the NdFeB layer thickness. When NdFeB layer thickness is 750 nm, the strain reaches the minimum value. Meanwhile, the highest in-plane and perpendicular coercivities can be obtained at NdFeB layer thickness of 750 nm, which are, respectively, 21.2 kOe and 19.5 kOe, because the strain is minimized. The high remanence ratio of 0.87 can also be achieved in Ta/Nd/NdFeB (750 nm)/Nd/Ta film. Altogether, the results suggest that Ta/Nd/NdFeB/Nd/Ta film may have a significant potential as a magnetic material with excellent performance.

Competing Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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