Hard magnetic properties of spacer-layer-tuned NdFeB/Ta/Fe nanocomposite films

W. B. Cui, H. Sepehri-Amin, Y. K. Takahashi, K. Hono

Elements Strategy Initiative Center for Magnetic Materials (ESSICM),

National Institute for Materials Science (NIMS), Tsukuba 305-0047, Japan

Abstract:

Anisotropic Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ta(x nm)/Fe(y nm)/Ti(20nm) multilayer films were prepared and the magnetic coupling mechanism between soft/hard-magnetic (SM/HM) layers were systematically studied in order to understand the potential of anisotropic SM/HM nanocomposite magnets. Recoil behaviors were also investigated in films with various thickness of Fe layer. From experimental results, the roles of exchange coupling and magnetostatic coupling in the demagnetization process were clarified, which was further supported by micromagnetic simulations. For different thickness of Ta spacer layer, the demagnetization process was analyzed and the coupling energy was estimated. Our work also indicated that by using proper thickness of Ta spacer layer the $(BH)_{\max}$ can be enhanced in the SM/HM nanocomposite magnets by a combination of weakened exchange coupling and magnetostatic coupling.

Keywords: permanent magnet; nanocomposite magnet; exchange coupling; coercivity; maximum energy product

1. Introductions

Nd-Fe-B based permanent magnets have regained a lot of attentions recently due to their broadening applications in the traction motors of hybrid and pure electric vehicles and wind generators. A large maximum energy product $(BH)_{\max}$ can be obtained in anisotropic Nd-Fe-B sintered magnet because of the high remanence and the coercivity higher than $\mu_0 M_r/2$ along with good squareness. The typical value of the $(BH)_{\max}$ of the commercial Nd-Fe-B magnet is about 400 kJ/m³ with a coercivity of 1.2 T. A higher $(BH)_{\max}$ of 475 kJ/m³ was reported for a laboratory scale Nd-Fe-B sintered magnet with highly oriented grains and minimized nonferromagnetic Nd-rich phases, but its coercivity is only 0.8 T, barely larger than $\mu_0 M_r/2$, which is too low for most applications [1].

The increasing cost and scarcity of rare-earth (RE) elements stimulates the development of permanent magnets using less amount of RE elements. In 1988, Coehoorn developed isotropic Nd₂Fe₁₄B/Fe₃B nanocomposite magnets with moderate coercivity and enhanced remanence by melt-spinning [2]. The RE content of the alloy was only 4.5%, much less compared with 12.5 ~ 14% of commercial sintered magnets. However, the isotropic magnets cannot exhibit a higher $(BH)_{\max}$ than that of anisotropic sintered magnets because of low remanence and poor squareness. For exchange coupling of soft and hard magnetic phases, there is an upper limit for the size of soft-magnetic phase. In one-dimensional, the grain size of a soft-magnetic (SM) phase must be smaller than the twice of the domain wall width δ_h of a hard-magnetic (HM) phase [3]. Here, δ_h is estimated by $\delta_h = \pi(A_h/K_h)^{1/2}$. Taking Nd₂Fe₁₄B as the hard magnetic (HM) phase, $A_h \approx 10^{-11}$ J/m, $K_h = 4.5 \times 10^6$ J/m³ [4], $2\delta_h$ is about 8.4 nm. Based on a three-dimensional calculation, in exchange coupled Sm₂Fe₁₇N₃/Fe₆₇Co₃₃ anisotropic nanocomposite magnets, a large $(BH)_{\max}$ of 1 MJ/m³ has been optimistically predicted [5].

Many experiments on anisotropic nanocomposite magnets [6,7] and their exchange coupling mechanism [8,9] had been carried out. Compared with the bulk nanocomposite magnet, controlling nanostructure is easier in thin films. So many investigations on anisotropic nanocomposite magnet films were done as proof-of-principle experiments, e.g., Nd-Fe-B/Fe [10,11] and SmCo/Fe [12,13] multilayer films. However, the reported values of $(BH)_{\max}$ were disappointing, due to small coercivity and degraded squareness caused by poor anisotropic texture. The reason for the small coercivity was thought to be the initiation of magnetic reversal from exchange-coupled SM layers. So, attentions were paid to how to

improve coercivity in NdFeB-based nanocomposite magnets. The coercivity of 0.8 T was reported for Mo-inserted NdFeB/Fe multilayer films [14] and the coercivity mechanism was studied due to the modified interfaces [15,16]. The importance of the interface between soft/hard-magnetic layers on coercivity was further demonstrated by the insertion of thin Ta layer between Nd₂Fe₁₄B and FeCo layers. By controlling the interlayer, a high coercivity of 1.38 T and (BH)_{max} of 486 kJ/m³ were achieved in a Nd-Fe-B/Ta/FeCo nanocomposite multilayer film [17]. The superior performance in the Ta-inserted Nd₂Fe₁₄B/Ta/FeCo nanocomposite raised a feasible approach for a high coercivity SM/HM nanocomposite. The direct contact between SM and HM layers will cause strong exchange coupling, which cause dramatic reduction of the coercivity of a hard phase. However, the coercivity of Nd₂Fe₁₄B/FeCo films could be kept comparable with that of single-layer Nd-Fe-B film by introducing 2 nm thick Ta spacer layer as a result of the weakened SM/HM exchange coupling [17]. These results indicate that Ta spacer layer plays a critical role between SM/HM layers, possibly leading to the different coupling mechanism. In this work, we have grown simplified Nd₂Fe₁₄B/Ta/Fe trilayer model films with various Ta and Fe thicknesses, to have a better understanding on the coupling mechanism and demagnetization process in the spacer-layer tuned SM/HM multilayer systems.

2. Experiments

An alloy target with a composition of Nd₁₃Fe₇₇B₁₀ was used to grow Nd-Fe-B hard magnetic layers. The base pressure was better than 10⁻⁶ Pa and the Ar pressure was kept at 1.3 Pa. The film structure was Ti(20nm)/NdFeB(100nm)/Nd(10nm)/Ta/Fe/Ti(20nm). Ti(20nm) underlayer and cover-layer were sputtered at room temperature to suppress oxidations. These two layers are omitted in the expression of thin film structure for simplicity. The NdFeB (100nm) layer was deposited at 600°C to get textured structure. In order to enhance the coercivity of the polycrystalline NdFeB layer, thin Nd(10nm) layer was deposited for the grain boundary diffusion process at 600°C for 30 min [18]. After cooling down to room temperature, Ta layer and Fe layer with varied thickness were deposited sequentially onto the NdFeB layers. The magnetic properties were measured by a superconducting quantum interference device (SQUID) vibrating sample magnetometer (VSM). The microstructure was characterized by transmission electron microscopy Titan G2 80-200.

To study the effect of the Ta spacer layer on the magnetization reversal of NdFeB/Ta/Fe thin films, micromagnetic simulations were performed on model thin films with NdFeB ($100 \times 100 \times 100 \text{ nm}^3$)/Ta ($100 \times 100 \times t \text{ nm}^3$)/Fe($100 \times 100 \times 10 \text{ nm}^3$) layers in which thickness of Ta layer was changed from 0 to 50 nm. The Nd-Fe-B layer was modeled with cubic shaped $\text{Nd}_2\text{Fe}_{14}\text{B}$ polycrystals with a mean grain size of 50 ± 10 nm separated from each other with the 5 nm thick grain boundary phase that was formed by the grain boundary diffusion process. The $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains were aligned parallel to the Z-axis direction with $\pm 10^\circ$ random misalignments by considering the textures in the experimentally grown films. The saturation magnetization ($\mu_0 M_s$), the magnetocrystalline anisotropy energy (K_1), and the exchange stiffness (A) were chosen to be 1.61 T, 4.5 MJ/m³, and 8 pJ/m for the $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase and 1.0 T, 0 MJ/m³ [4,19] and 8 pJ/m for the grain boundary phase, respectively. In fact, there is no report regarding to the magnetic behavior of the grain boundary phase in Nd-Fe-B thin films. Here, we assumed that the grain boundary phase has similar chemistry and magnetic behavior to that of reported recently for Nd-Fe-B sintered magnets [20,21]. The saturation magnetization ($\mu_0 M_s$), the magnetocrystalline anisotropy energy (K_1) and the exchange stiffness (A) were chosen to be 0.0 T, 0.0 MJ/m³ and 0.0 pJ/m for Ta layer and 2.15 T, 0.048 MJ/m³ and 22.0 pJ/m for Fe layer, respectively [19]. Tetrahedron meshes were applied with a size of 1.5 nm and the Landau-Lifshitz-Gilbert (LLG) equation at each node was solved by the FEMME software [22].

3. Results

Figure 1 shows the X-ray diffractions of NdFeB(100nm)Nd(10nm) single-layer (SL) film compared with those of NdFeB(100nm)Nd(10nm)/Ta($x \text{ nm}$)/Fe(10nm) films ($x = 2 \text{ nm}, 10 \text{ nm}, 100 \text{ nm}, 250 \text{ nm}$). Strong (004) and (006) peaks are observed in the XRD of the SL film. A peak around $2\theta = 31^\circ$ is from Nd-rich phase, which is of double hexagonal close packed (dhcp) structure. In the thin films with Ta(2nm) spacer layer and Ta(10nm) spacer layer, besides the (0 0 2 l) peaks, the minor peaks corresponding to (114) of $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase is observed. In the thin films with Ta(100nm) and Ta(250nm) spacer layers, the (105) peak of $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase appears along with some additional peaks from Ta spacer layer.

Figure 2 compares the hysteresis loops of the NdFeB(100nm)Nd(10nm) SL film and those of the NdFeB(100nm)Nd(10nm)/Ta($x \text{ nm}$)/Fe(10nm) films ($x = 0 \text{ nm}, 2 \text{ nm}, 10 \text{ nm}, 50 \text{ nm}, 250 \text{ nm}$ and 400 nm). In the SL film, a good squareness and a high remanence ratio are achieved along the out-of-plane direction

as seen in Fig. 2a. Along the in-plane direction, the magnetization is hard to be saturated and a wide hysteresis is observed, indicating the strong perpendicular anisotropy, consistent with the XRD results. The coercivity of the SL film is about 1.2 T. When Fe(10nm) layer is deposited on the NdFeB film directly, the saturation magnetization is increased while keeping the perpendicular anisotropy (Fig. 2b). However, due to the **exchange coupling** between the NdFeB layer and the Fe layer, the coercivity is substantially decreased to 0.48 T, following the same tendency observed in Ref. 15. When Ta(2nm) spacer layer was inserted between SM and HM layers, coercivity was recovered to 0.95 T, although not as high as 1.2 T of the SL film due to the existence of Fe(10nm) layer. Along the out-of-plane direction, the magnetization starts to decrease from a positive field smoothly to the near zero field. The squareness and remanence become lower compared with those of the SL film. In contrast, the discontinuity becomes observable along the in-plane direction, compared with the smoothness near zero fields along the out-of-plane direction in Fig. 2a and Fig.2b.

The hysteresis loops of the films with Ta thickness from 10 nm to 250 nm can be seen from Fig. 2c to Fig. 2e. Along the out-of-plane direction, a common feature is that the magnetization starts to decrease at a positive field with the smooth demagnetization process to the nucleation field of the HM layer. Along the in-plane direction, a kink is observed in all the Ta inserted films and becomes sharper with increasing thickness of the Ta spacer layer, indicating a complete decoupling between SM/HM layers along the in-plane direction. When the thickness of Ta is further increased to 400 nm, the kink appears along the out-of-plane direction. Note that a kink appears at 2 nm of spacer layer along the in-plane direction, while it appears at 400 nm along the out-of-plane direction. This indicates that the threshold thickness at which the kink appears is different along the out-of-plane direction and the in-plane direction. Fig. 2h shows the coercivity dependence on the thickness of Ta spacer layer along the out-of-plane direction. The coercivity of the films with Ta spacer layer from 2 nm to 250 nm are between 0.8 T and 0.9 T, larger than 0.47 T of Ta-free film, but lower than 1.2 T of the SL film, due to the existence of soft-magnetic layer. **However, the coercivity is recovered to 1.34 T in the thin film with 400 nm thickness of Ta spacer. Because in this film the Fe moments are totally decoupled with the NdFeB layer and rotated easily by external field. The rotation of Fe moments will not leads to the rotation of the moments in the NdFeB layer. Therefore the coercivity of this film is mainly dependent on the NdFeB layer,**

Figure 3 shows the cross-sectional bright-field (BF) TEM images and corresponding Nd, Fe and Ta energy disperse spectrum (EDS) maps of NdFeB(100 nm)Nd(10 nm)/Ta(2nm)/Fe(10nm) (a) and NdFeB(100 nm)Nd(10 nm)/Ta(50nm)/Fe(10nm) (b) films. In the thin film with a Ta(2nm) layer, a uniform NdFeB layer can be observed. Nd-rich region is clear from the corresponding Nd EDS image. The BF image and the Fe EDS image show that both plate-like Nd₂Fe₁₄B grains and columnar Nd₂Fe₁₄B grains are formed as indicted in the Fig. 3a. A thin Fe layer with 10 nm thickness is seen. Between the Fe layer and the NdFeB layer, a thin Ta spacer layer of 2 nm thickness is observed as indicated in the Ta EDS image. In the thin film with 50 nm thick Ta spacer layer, due to the Nd diffusion process, some Nd-rich regions are also observed as indicated in Nd EDS image but not homogenously distributed. Plate-like Nd₂Fe₁₄B grains are also observed. The Ta EDS image indicates that a 50 nm thick Ta spacer layer is sandwiched by NdFeB layer and Fe layer with thickness of 10 nm. From the cross-sectional TEM images, it can be observed that NdFeB layers are thoroughly separated with Fe layer by the Ta spacer.

The demagnetization process is also affected by the thickness of Fe layer, as shown in Figure 4. In NdFeB(100 nm)Nd(10 nm)/Ta(2nm)/Fe(5nm) thin films, the good squareness of the out-of-plane magnetization curve indicates the strong coupling between NdFeB and Fe layers. The coercivity of 1.17 T is marginally lower than 1.2 T of the SL film. This is because both Ta spacer layer and Fe layer are thin, leading to nearly uniform reversal in both SM/HM layers. Along the in-plane direction, the hysteresis loop is continuous as observed in the SL film. When Fe thickness is increased from 20nm (Fig. 4b) to 100 nm (Fig.4c), the magnetization starts to drop at a positive field along the out-of-plane direction, in contrast to the sharp drop along the in-plane direction. With thicker Fe layer, more moments in Fe layer tend to be twisted under the external magnetic field along the out-of-plane direction. Surprisingly the demagnetization around zero field is still smooth for Fe(100nm) case. However, if the Fe thickness is increased to 120 nm, a minor kink is observed around zero fields along the out-of-plane direction, indicating that some Fe moments cannot be pinned by the HM layer and independent magnetization reversal occurs along the out-of-plane direction. Note the critical length, which is defined as the thickness above which the kink appears, is around 120 nm, significantly larger than the critical length (≈ 10 nm) predicted by one-dimensional exchange spring magnets [3,18]. In contrast, a kink appears for the thin film when Fe thickness is 10 nm along in-plane direction (as seen in Fig.2c), which is much smaller than 120 nm, the

critical length along the out-of-plane direction (as seen in Fig. 4d).

Spring-back effect is a characteristic behavior to describe the SM/HM spring magnets. However, spring-back effect was mainly studied in the isotropic magnets in previous studies [3,11]. The recoil loops of single-layer (SL) film (a) and Ti(20nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(y)/Ti(20nm) films ($y = 5\text{nm}$ (b), 20 nm (c) and 100 nm (d)) are shown in Figure 5. The nucleation fields (H_n) for these films are marked. In Fig. 5a, as long as the external field is lower than nucleation field, the magnetization/demagnetization process is reversible. When the external field is larger than H_n , the irreversible process occurs, which is due to the irreversible domain wall motions in the NdFeB layer. In the SL film (Fig. 5a), the reversible component (δM_{rev}) is hardly observed. With increasing Fe thickness from 5 nm to 100 nm (as see from Fig. 5b to Fig.5d), the slope of the recoil curve increased. The reversible magnetization/demagnetization processes are also observed as long as the external field is smaller than their respective nucleation field. δM_{rev} becomes larger with thicker Fe layer, indicating that more Fe moments become free from the hard phase because of weakened pinning by the HM layer.

4. Discussions

Kinks are considered to be the sign that a SM phase cannot be fully coupled with a HM phase and some free moments appears in the SM phase. As seen from Fig. 2 and Fig. 4, along the in-plane direction, for a fixed Ta(2nm) spacer layer, the kink appears when Fe thickness is increased from 5 nm (see Fig. 4a) to 10 nm (see Fig 2c). On the other hand, for a fixed Fe(10nm) layer, along the in-plane direction, the kink appears when Ta thickness is increased from 0 nm (see Fig. 2b) to 2nm (Fig. 2c), indicating the decoupling between SM/HM phases. It can be explained that exchange coupling is of short-range coupling and decay rapidly with increasing spacer layer, which have been proved in other FM/spacer/FM systems [23,24] and AFM/spacer/FM systems[25,26]. Supposing there is only exchange coupling in the NdFeB/Ta/Fe trilayer system, the decoupling would be observed simultaneously along both in-plane and out-of-plane direction in the NdFeB(100nm)Nd(10 nm)/Ta(2 nm)Fe(10nm) film. But it is contrary to the experimental results, because the kink is observed when Ta spacer layer is increased to 400 nm along the out-of-plane direction. Such long-ranged coupling is attributed to the magnetostatic coupling between SM and HM layers.

To confirm the role of magnetostatic coupling in the spacer-layer tuned SM/HM layers, the

micromagnetic simulations were applied. The geometry of modeled thin films was NdFeB (100nm)/Ta(x nm)/Fe(10nm) with various Ta thickness as shown in Fig. 6. The Nd₂Fe₁₄B grains are separated with 5 nm thick grain boundary phase to mimic the grain boundaries in the Nd-diffusion processed SL film. The Fe layer on top has a constant thickness of 10 nm. By using such geometry, the hysteresis loops of NdFeB (100nm)/Ta(x nm)/Fe(10nm) films ($x = 0$ nm and 2 nm) were simulated with and without considering the magnetostatic coupling as shown in Figure 7. Without Ta spacer layer ($x = 0$ nm), the demagnetization process occurs smoothly because the exchange coupling is strong and dominant in this case whether magnetostatic coupling is present or not. With a Ta(2nm) spacer layer, an obvious kink is observed if without considering magnetostatic coupling because the exchange coupling is weakened by 2 nm of Ta spacer layer and SM phase is reversed independently from HM phase. With considering magnetostatic coupling between SM and HM layers, the out-of-plane demagnetization curve becomes smooth, which is in agreement with the experimental results. It can be understood that when Ta spacer layer is thinner than 2 nm, the exchange coupling is dominant. Because exchange coupling decays when Ta spacer layer is thicker than 2 nm, magnetostatic coupling become dominant for Ta thickness from 2 nm to 250 nm. With further increasing thickness of a Ta spacer, the magnetostatic coupling is also so weakened that some free moments appears in the SM phase, leading to the appearance of a kink on the out-of-plane hysteresis loop (as seen in Fig. 2g). The experimental results confirm that exchange coupling and magnetostatic coupling co-exist in the multilayer films and proved by micromagnetic simulations. It also verifies the proposed beneficial role of magnetostatic coupling [27].

Figure 8 shows the magnetization configurations of modeled NdFeB (100nm)/Ta(x nm)/Fe(10nm) films for (a) $x = 0$ and (b) $x = 2$ during magnetization reversal from saturated state to the coercive field of the HM layer. The applied magnetic field is parallel to the Z direction and reduced from 7 T to -7 T. In these figures, the red color corresponds to the magnetization direction to $+Z$ and blue color corresponds to the magnetization direction to $-Z$. Applied magnetic field for each snap shot is shown in the inset. The models are fully saturated with applying $\mu_0 H_{\text{ext}} = 7$ T. A magnetization reversal starts from the Fe layer by the formation of vortex-like domain structure. This can be seen for both $x = 0$ and 2 nm cases. The formation of a vortex domain structure in the Fe layer starts from positive magnetic

field, which is the origin of reduction of the magnetic field at positive fields in the out-of-plane hysteresis loops shown in Fig. 2 and Fig. 4. By further decreasing the magnetic field to -1 T, the Fe moments precess gradually from +Z to nearly -Z direction. In the nucleation field of the HM layer ($\mu_0 H_{\text{ext}} = -1.32\text{T}$), magnetization reversal starts to occur in the HM layer from the SM/HM interface for $x=0$ due to the twisted configuration of Fe moments and NdFeB moments. By further decreasing the external field to $\mu_0 H_{\text{ext}} = -1.54\text{ T}$, magnetization reversal propagates into the NdFeB layer as seen Fig.8a. By introducing the 2 nm thick Ta layer, the antiparallel moment configuration of Fe and Nd-Fe-B layer can be accommodated as shown in $\mu_0 H_{\text{ext}} = -1.89\text{T}$ in Fig. 8b. This is because the Ta layer decays the exchange coupling and the HM/SM layers can be more easily magnetostatic coupled. Such a configuration leads to the enhancement of coercivity. (The dynamic demagnetization process can be seen in Supplementary video 1).

Summarized from the hysteresis loops in Fig. 2, the change of the demagnetization process between near zero field and coercivity field for different thicknesses of Ta spacer layer is schematically shown in Figure 9a. Strong coupling leads to a good squareness and higher remanence. In the saturated state, all the moments of Fe layer and NdFeB layer are parallel. At the nucleation field, some moments in the SM layer are already tilted from the out-of-plane direction. Therefore, there will be a reduction from the saturation magnetization, termed as δM as seen in Fig.9a. Figure 9b shows δM as a function of the thickness of Ta spacer layer. When the Ta spacer layer is thin, exchange coupling is dominant. Then, most of Fe moments are pinned by the HM layer, leading to low δM . With thicker Ta, magnetostatic coupling becomes dominant and some Fe moments rotate freely from the hard layer, leading to larger δM . It can be seen that when Ta spacer is thicker than 5 nm, δM is almost constant at $3 \times 10^{-3} \text{ emu/cm}^2$, just equal to the twice of magnetization of Fe(10nm) layers (see inset figure of Fig. 9b). It means that once all Fe moments are reversed by the external field, magnetization reversal starts in the NdFeB layer, consistent with the micromagnetic simulations in Fig. 8. Therefore, between the zero field and the nucleation field, for each dM_i , the external field H_i is needed to rotate the moment from remanence state to antiparallel alignment with NdFeB moments. So, the energy needed in this process is $d\sigma = -H_i dM_i$, which is numerically equal to the coupling energy of dM_i pinned by HM layer. Therefore, the coupling energy along the out-of-plane

direction can be expressed as :

$$\sigma = \sum d\sigma_i = - \int_{M_{NdFeB}-M_{Fe}}^{M_r} H_i dM_i$$

where M_r is the remanence. $M_{NdFeB}-M_{Fe}$ is the magnetization at nucleation field, at which state all Fe moments are almost antiparallel to the NdFeB moments. Here, σ includes the contributions from both exchange coupling and magnetostatic coupling, schematically represented by the area as indicated in Fig. 9c. Figure 9d shows the dependence of σ on the thickness of Ta spacer layer along the out-of-plane direction. When Ta spacer is very thin, σ is about 5.5 mJ/m², which is comparable with 3 ~ 11 mJ/m² reported in Ref. 28 and 29 estimated by ferromagnetic resonance (FMR), indicating the reliability of such approach to estimate the coupling energy.

Figure 10 shows the $(BH)_{max}$ as a function of the thickness of Ta in Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm)/Ti(20 nm) films ($x = 0 \sim 50$ nm) and the thickness of Fe in Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(y nm)/Ti(20 nm) films ($y = 0 \sim 20$ nm). In Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm)/Ti(20 nm) films, the initial increment of $(BH)_{max}$ from 113 kJ/m³ in the film without Ta spacer to 205 kJ/m³ in the film with 2 nm of Ta spacer is due to the recovery of coercivity as seen in Fig. 2. In Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(y nm)/Ti(20 nm) films, with increasing thickness of Fe layer, the $(BH)_{max}$ increased from 195 kJ/m³ at $y = 0$ nm to the maximum of 269 kJ/m³ at $y = 5$ nm. Too thick Ta layer and Fe layers leads to either diluted magnetization or poor squareness and coercivity, causing the reduction of $(BH)_{max}$. It is noticed that the volume fraction of the SM phase in this model system is low. The squareness is also reduced by 2 nm of Ta spacer layer and free surface of Fe layer. The highest $(BH)_{max}$ cannot be as high as the optimized one in multilayer film [17]. From Fig. 10, it is indicated that **strong exchange coupling** is not indispensable in gaining higher $(BH)_{ma}$, in agreement with the numerical simulation in ref. 23. By properly controlling the thickness of Ta spacer layer and SM layer, weakened exchange coupling can also lead to the improved $(BH)_{max}$ assisted by the magnetostatic coupling. By combination of weakened exchange coupling and magnetostatic coupling, it is promising to realized superior $(BH)_{max}$ and moderate coercivity in anisotropy nanocomposite, as we reported in multilayer systems [17].

5. Conclusions

The anisotropic Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ta(x nm)/Fe(y nm)/Ti(20nm) thin films are prepared with variant thickness of Ta spacer layer and Fe layer. Demagnetization process and recoil behaviors were experimentally studied. The effects of exchange coupling and magnetostatic coupling in NdFeB/Ta/Fe trilayer films on their hard magnetic properties were discussed based on the experimental observations as well as micromagnetic simulations. It was found that exchange coupling was the dominant when Ta was thinner than 2 nm. With increasing Ta spacer thickness, the exchange coupling decreased quickly while magnetostatic coupling became dominant. If there is no Ta spacer, coercivity is substantially decreased. But it can be recovered by the insertion of a Ta spacer layer. This work also shows that properly tuning the strength of exchange coupling and magnetostatic coupling by spacer layer insertion is essential to gain an enhanced $(BH)_{\max}$ in anisotropic NdFeB/Fe nanocomposite magnet and maintain coercivity.

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Figure captions

- Fig. 1 X-ray diffraction patterns of Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ti(20nm) single-layered (SL) film and Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ta(x nm)/Fe(10nm)/Ti(20nm) nanocomposite films ($x = 2$ nm, 10 nm, 50 nm and 250 nm).
- Fig. 2 Hysteresis loops of Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ti(20nm) single-layer (SL) film (a) and Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ta(x nm)/Fe(10nm)/Ti(20nm) nanocomposite films ($x = 0$ nm (b), 2 nm(c), 10 nm (d), 50 nm (e), 250 nm(f) and 400 nm(g)). The coercivity dependence of Ta thickness is shown in Fig. 2h.
- Fig. 3 Cross-sectional bright-field (BF) TEM images of Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(10 nm)/Ti(20 nm) (a) and Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(50 nm)/Fe(10 nm)/Ti(20 nm) (b) films. The energy disperse spectrums (EDS) of Nd, Fe and Ta elements are also shown.
- Fig. 4 Hysteresis loops of Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ta(2nm)/Fe(y)/Ti(20nm) films ($y = 5$ nm (a), 20 nm(b), 100 nm (c), 120 nm (d)) along out-of-plane (OOP) direction and in-plane (IP) direction.
- Fig. 5 Recoil loops of Ti(20nm)/NdFeB(100nm)Nd(10nm)/Ti(20nm) single-layer (SL) film (a) and Ti(20nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(y)/Ti(20nm) films ($y = 5$ nm (b), 20 nm (c) and 100 nm(d)). The different demagnetization fields of each recoil curve are indicated in each figure. The marked δm_{rev} stands for the magnetization changes in recoil curves.
- Fig. 6 Modeled geometry of NdFeB (100nm)/Ta(X)/Fe(10nm) thin film with variant Ta thickness. In such geometry, Nd₂Fe₁₄B grains are separated with 5nm thick grain boundary phase to mimic the grain boundaries in real films. The Fe layer on top has a constant thickness of 10 nm while the thickness of Ta layer can be changeable.
- Fig. 7 Simulated hysteresis loops of Ti(20 nm)/NdFeB(100 nm)/Ta(x)/Fe(10 nm)/Ti(20 nm) films ($x = 0$ and 2 nm) with considering (a and b) and without considering (c and d) magnetostatic coupling along out-of-plane (OOP) direction and in-plane (IP) direction. Fig.7a and Fig.7c are simulated hysteresis loops with Ta(0 nm). Fig.7b and Fig.7d are simulated hysteresis loops with Ta(2 nm).

The area marked by red corresponds schematically to the coupling strength between soft/hard-magnetic layers.

Fig. 8 Magnetization configurations of modeled NdFeB (100nm)/Ta(X)/Fe(10nm) thin films with (a) Ta thickness of $X = 0$ nm and (b) Ta thickness of $X = 2$ nm during magnetization reversal from saturated state to their respective coercive field. The corresponding applied magnetic field is shown in the inset of figures. The dynamic process can be referred to Suppl. Video 1.

Fig. 9 (a) Schematics of demagnetization process of thin films with different Ta thickness. (b) δM as a function of the thickness of Ta spacer layer in Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm)/Ti(20 nm) films ($x = 0 \sim 400$ nm). The inset figure shows the magnetization curve of 10 nm of Fe layer. (c) Schematic of the estimation of coupling energy; (d) coupling energy σ as a function of the thickness of Ta spacer layer along out-of-plane direction.

Fig.10 $(BH)_{\max}$ as a function of the thickness of Ta spacer layer in Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm)/Ti(20 nm) films ($x = 0 \sim 50$ nm) and the thickness of Fe layer in Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(2 nm)/Fe(y nm)/Ti(20 nm) films ($y = 0 \sim 20$ nm). The dashed line stands for the $(BH)_{\max}$ benchmark of single-layered (SL) Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ti(20 nm) thin film.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10