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ScienceDirect

Acta Materialia 84 (2015) 405–412

Acta MATERIALIA

www.elsevier.com/locate/actamat

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

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Received 1 July 2014; revised 5 October 2014; accepted 7 October 2014
Available online 26 November 2014

Abstract—Anisotropic Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(y nm)/Ti(20 nm) 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 thicknesses 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 thicknesses of Ta spacer layer, the demagnetization process was analyzed, and the coupling energy was estimated. This work also indicated that, using the 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. Introduction

Nd–Fe–B-based permanent magnets have attracted a lot of attention recently, owing 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 its high remanence and a 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 ~400 kJ m^{-3} with a coercivity of 1.2 T. A higher $(BH)_{max}$ of 475 kJ m^{-3} was reported for a laboratory-scale Nd–Fe–B sintered magnet with highly oriented grains and minimized non-ferromagnetic Nd-rich phases, but its coercivity was 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 has stimulated the development of permanent magnets using fewer RE elements. In 1988, Coehoorn et al. [2] developed isotropic $Nd_2Fe_{14}B/Fe_3B$ nanocomposite magnets with moderate coercivity and enhanced remanence by melt-spinning. The RE content of the alloy was only 4.5%, much less compared with ~12.5–14% for 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 the soft-magnetic (SM) phase. In one dimension, the grain size of a SM phase must be less than the twice 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 $Nd_2Fe_{14}B$ as the HM phase, $A_h \approx 10^{-11}$ J m^{-1}, $K_h = 4.5 \times 10^6$ J m^{-3} [4], $2\delta_h$ is ~8.4 nm. Based on a three-dimensional calculation, in exchange coupled $Sm_2Fe_{17}N_3/Fe_{67}Co_{33}$ anisotropic nanocomposite magnets, a large $(BH)_{max}$ of 1 MJ m^{-3} has been optimistically predicted [5].

Many experiments on anisotropic nanocomposite magnets [6,7] and their exchange coupling mechanism [8,9] have 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, owing 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, attention was paid to how to improve the coercivity in NdFeB-based nanocomposite magnets. A coercivity of 0.8 T was reported for Mo-inserted NdFeB/Fe multilayer films [14], and the coercivity mechanism was studied as a result of the modified interfaces [15,16]. The importance of the interface between SM/HM layers for coercivity was further demonstrated by the insertion of a thin Ta layer between $Nd_2Fe_{14}B$ and FeCo layers. By controlling the interlayer, a high coercivity of 1.38 T and $(BH)_{max}$ of

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http://dx.doi.org/10.1016/j.actamat.2014.10.008
1359-6462/

486 kJ m^{-3} were achieved in a Nd–Fe–B/Ta/FeCo nanocomposite multilayer film [17]. The superior performance in the Ta-inserted $Nd_2Fe_{14}B$/Ta/FeCo nanocomposite enabled a feasible approach for a high-coercivity SM/HM nanocomposite. The direct contact between SM and HM layers will cause strong exchange coupling, which causes a dramatic reduction in the coercivity of a hard phase. However, the coercivity of $Nd_2Fe_{14}B$/FeCo films could be kept comparable with that of single-layer (SL) Nd–Fe–B film by introducing a 2-nm-thick Ta spacer layer as a result of the weakened SM/HM exchange coupling [17]. These results indicate that the Ta spacer layer plays a critical role between SM/HM layers, possibly leading to a different coupling mechanism. In the present work, simplified $Nd_2Fe_{14}B$/Ta/Fe trilayer model films with various Ta and Fe thicknesses were grown, to gain a better understanding of the coupling mechanism and demagnetization process in spacer-layer-tuned SM/HM multilayer systems.

2. Experiments

An alloy target with a composition of $Nd_{13}Fe_{77}B_{10}$ 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(20 nm)/NdFeB(100 nm)/Nd(10 nm)/Ta/Fe/Ti(20 nm). The Ti(20 nm) underlayer and cover layer were sputtered at room temperature to suppress oxidation. These two layers are omitted in the expression of the thin film structure for simplicity. The NdFeB(100 nm) layer was deposited at 600 °C to get a textured structure. In order to enhance the coercivity of the polycrystalline NdFeB layer, thin Nd(10 nm) layer was deposited for the grain boundary diffusion process at 600 °C for 30 min [18]. After cooling to room temperature, the Ta layer and the Fe layer with various thicknesses 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 (TEM) using a Titan G2 80–200 instrument.

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($100 \times 100 \times 100$ nm^3)/Ta($100 \times 100 \times t$ nm^3)/Fe($100 \times 100 \times 10$ nm^3) layers in which the thickness of the Ta layer was changed from 0 to 50 nm. The Nd–Fe–B layer was modeled with cubic $Nd_2Fe_{14}B$ polycrystals with a mean grain size of 50 ± 10 nm, separated from each other by the 5-nm-thick grain boundary phase that was formed by the grain boundary diffusion process. The $Nd_2Fe_{14}B$ grains were aligned parallel to the z-axis direction with $\pm 10°$ 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 m^{-3} and 8 pJ m^{-1} for the $Nd_2Fe_{14}B$ phase, and 1.0 T, 0 MJ m^{-3} [4,19] and 8 pJ m^{-1} for the grain boundary phase, respectively. In fact, there are no reports regarding the magnetic behavior of the grain boundary phase in Nd–Fe–B thin films. Here, it is assumed that the grain boundary phase has chemistry and magnetic behavior similar to that 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 m^{-3} and 0.0 pJ m^{-1} for the Ta layer, and 2.15 T, 0.048 MJ m^{-3} and 22.0 pJ m^{-1} for the Fe layer, respectively [19]. Tetrahedron meshes were applied with a size of 1.5 nm, and the Landau–Lifshitz–Gilbert equation at each node was solved by FEMME software [22].

3. Results

Fig. 1 shows the X-ray diffraction (XRD) patterns of NdFeB(100 nm)Nd(10 nm) SL film compared with those of NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm) films ($x = 2$ nm, 10 nm, 100 nm, 250 nm). Strong (004) and (006) peaks are observed in the XRD of the SL film. A peak $\sim 2\theta = 31°$ is from the Nd-rich phase, which is of double hexagonal close-packed structure. In the thin films with a Ta(2 nm) spacer layer and a Ta(10 nm) spacer layer, besides the (00 2l) peaks, a minor peak corresponding to (1 1 4) of $Nd_2Fe_{14}B$ phase is observed. In the thin films with Ta(100 nm) and Ta(250 nm) spacer layers, the (1 0 5) peak of $Nd_2Fe_{14}B$ phase appears, along with some additional peaks from the Ta spacer layer.

Fig. 2 compares the hysteresis loops of the NdFeB (100 nm)Nd(10 nm) SL film and those of the NdFeB (100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm) films ($x = 0$ nm, 2 nm, 10 nm, 50 nm, 250 nm and 400 nm). In the SL film, good squareness and a high remanence ratio are achieved along the out-of-plane (OOP) direction, as shown in Fig. 2a. Along the in-plane (IP) direction, the magnetization is hard to saturate, and a wide hysteresis is observed, indicating the strong perpendicular anisotropy, consistent with the XRD results. The coercivity of the SL film is ~1.2 T. When the Fe(10 nm) layer is deposited on the NdFeB film directly, the saturation magnetization is increased, while keeping the perpendicular anisotropy (Fig. 2b). However, owing 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 the Ta(2 nm) spacer layer was inserted between the SM and HM layers, the

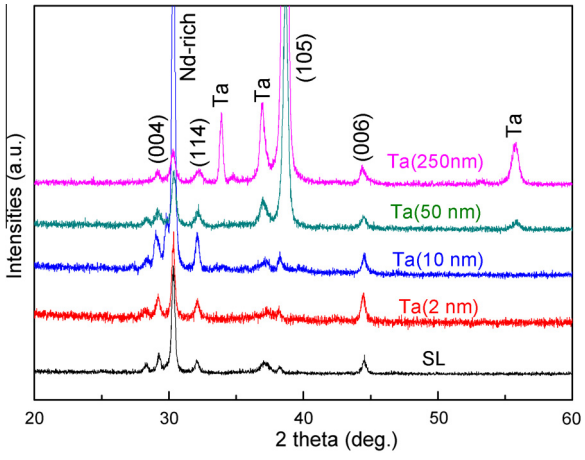

Fig. 1. XRD patterns of Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ti(20 nm) SL film and Ti(20 nm)/NdFeB(100 nm)Nd(10 nm)/Ta(x nm)/Fe(10 nm)/Ti(20 nm) nanocomposite films ($x = 2$ nm, 10 nm, 50 nm and 250 nm).