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The magnetic properties of nanocomposite multilayer magnets of the $(Nd,Dy)(Fe,Co,Nb,B)_{5.5}/M$ (M=Co,FeCo) on Ti-buffered Si substrates have been investigated. X-ray diffraction results reveal that the $Nd_2Fe_{14}B$-type phase in almost all the films is randomly oriented. Different thickness x (nm) for Co layers and y (nm) for FeCo layers were adopted in the multilayers. It is found that high remanence is achieved in the nanocomposite multilayer films consisting of the $Nd_2Fe_{14}B$-type phase and soft magnetic phase for Co with $6 \geqslant x \geqslant 4$ and for FeCo with $10 \geqslant y \geqslant 6$. The enhancement of the remanence in the nanocomposite multilayer films is attributed to the exchange coupling between the magnetically soft and hard phases. Increasing the soft magnetic components results in a continuously decreasing coercivity. © *2002 American Institute of Physics.*
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I. INTRODUCTION

Many experimental investigations of nanocomposite magnets prepared by rapid quenching and mechanical alloying have been reported in the past ten years.[1–5] Because of the difficulties in controlling the morphology in nanocomposite magnets, the maximum energy products achieved have been much smaller than those expected from the calculations. On the other hand, it is known that the morphology of nanocomposite films can be relatively easily controlled, by adjusting parameters like the thickness of hard and soft phases, and by changing annealing temperature and time in nanostructured multilayer magnets. Recently, some studies on exchange coupling were carried out for nanostructured CoSm/FeCo and PrCo/Co multilayer films prepared by sputtering and subsequent heat treatment.[6,7] Magnetic properties of exchange-coupled α-Fe/Nd–Fe–B multilayer magnets were investigated by Shindo[8] and the observations for the Nd–Fe–B/Fe/Nd–Fe–B trilayers were reported by Parhofer and Yang *et al.*[9–11] It is found that further studies are necessary in developing nanostructured multilayer magnets. In the present article, we report the structural and magnetic properties of nanocomposite $(Nd,Dy)(Fe,Co,Nb,B)_{5.5}/M$ $(M=Co,Fe_{65}Co_{35})$ multilayer magnets synthesized by sputtering and subsequent annealing.

II. EXPERIMENT

$(Nd,Dy)(Fe,Co,Nb,B)_{5.5}/M$ $(M=Co,Fe_{65}Co_{35})$ multilayer films were prepared with a multiple-gun dc- and rf-sputtering system by depositing the $(Nd_{0.9}Dy_{0.1})(Fe_{0.77}Co_{0.12}Nb_{0.03}B_{0.08})_{5.5}$ alloy and Co or FeCo targets onto silicon substrates covered with a 20 nm Ti buffer. The target of hard magnetic phase alloy was prepared by sintering powders of the alloys and the others were commercial products. Purities of all the materials of targets were higher than 99.9%. The base pressure of the sputtering system was $2-3\times10^{-7}$ Torr, and the Ar pressure during the

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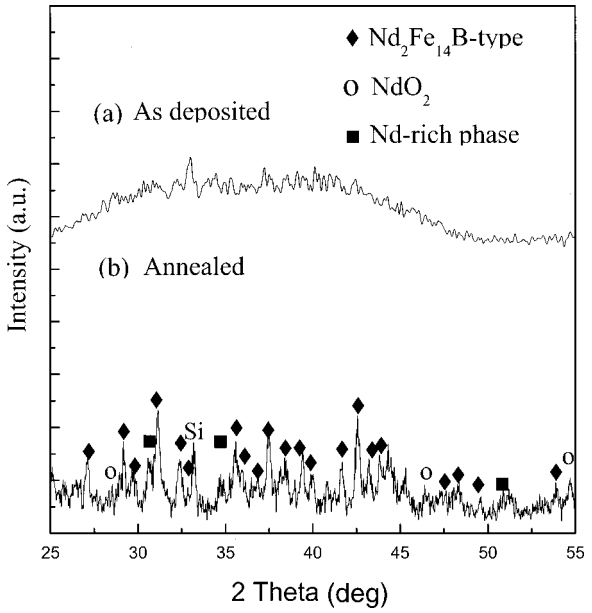

FIG. 1. XRD patterns for the Ti(20 nm)/NdDyFeCoNbB(300 nm)/Ti(20 nm)/(Si substrate) thin film as-deposited and annealed at 625 °C for 5 min.

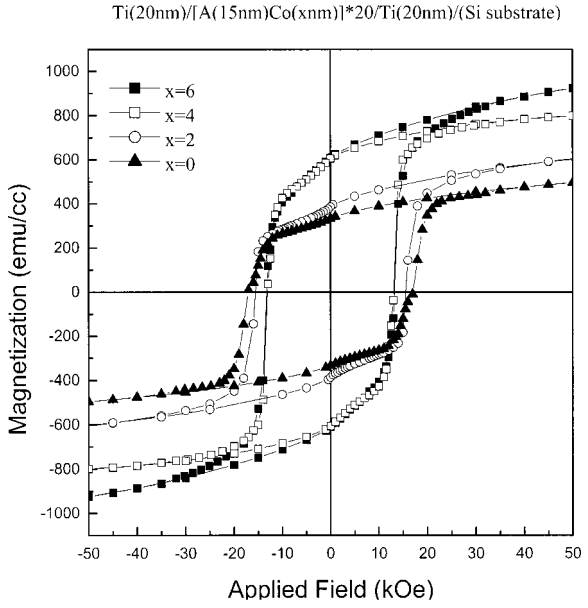

FIG. 2. Hysteresis loops at room temperature for the thin film of Ti-buffered NdDyFeCoNbB and NdDyFeCoNbB/Co multilayers on the Si substrate annealed at 625 °C for 5 min. Magnetic measurements are in the film plane.

sputtering was 5 mTorr. The thickness of the films was measured by weighting the mass of the films. The as-deposited films were heat-treated in a furnace with a vacuum of 2×10^{-7} Torr. The crystalline structure of the phases in the films was determined by x-ray diffractometry (XRD) with Cu $K\alpha$ radiation. The nanostructure of a representative sample was examined by transmission electron microscopy (TEM). Magnetic properties of the films were measured by an alternating gradient force magnetometer and superconducting quantum interference device magnetometer. The hysteresis loops and the magnetic properties were recorded without the demagnetizing correction.

III. RESULTS AND DISCUSSION

To study the structural and magnetic properties of multilayers, initially a hard phase single-layer film with composition of Ti(20 nm)/NdDyFeCoNbB(300 nm)/Ti(20 nm)/(Si substrate) was investigated.

Figure 1 shows XRD patterns for the Ti(20 nm)/NdDyFeCoNbB(300 nm)/Ti(20 nm)/(Si substrate) thin film as-deposited and annealed at 625 °C for 5 min. In the as-deposited film, the amorphous phase is formed. After annealing at 625 °C for 5 min, the hard magnetic phase with $Nd_2Fe_{14}B$-type structure is formed, accompanied with some Nd-rich phase and a trace of NdO_2 in the film.

Figure 2 gives hysteresis loops at room temperature of the thin films of Ti(20 nm)/[NdDyFeCoNbB(15 nm) Co($x$nm)]$\times$20/Ti(20 nm)/(Si substrate) multilayers annealed at 625 °C for 5 min. It can be seen from Fig. 2 that the intrinsic coercivity of the single layer is as high as 17.2 kOe, the remanence is about 333 emu/cc for $x=0$, i.e., the single-layer case. With increasing thickness of Co, the remanence increases and coercivity decreases slightly. It is clearly seen that for $4\leqslant x\leqslant 6$, the existence of Co layers among $Nd_2Fe_{14}B$-type hard-phase layers results in an increase of remanence by about 270 emu/cc and a decrease of the coercivity by about 4 kOe, compared to the case of single layer. The enhanced remanence is due to exchange coupling between hard and soft phase and magnetic softening is caused by the soft magnetic component.

Because of the higher saturation magnetization of $Fe_{65}Co_{35}$ alloy than those of pure Co and Fe, the $Fe_{65}Co_{35}$ alloy is also chosen as the soft magnetic layer component of the multilayer films. XRD patterns for Ti(20 nm)/[NdDyFeCoNbB(15 nm)$Fe_{65}Co_{35}$(y nm)] $\times$20/Ti(20 nm)/(Si substrate) multilayers annealed at 625 °C for 1 min are given in Fig. 3. In comparison with the result of the single layer, it is found that there is a large amount of α-Fe(Co) in the films and trace quantity of Nd-rich phase and NdO_2 besides the hard magnetic phase of $Nd_2Fe_{14}B$ type. The Nd-rich phase decreases quickly with increasing thickness of FeCo layers. The XRD profiles of the films reveal a random orientation of $Nd_2Fe_{14}B$-type phases.

Figure 4 shows hysteresis loops at room temperature for the Ti(20 nm)/[NdDyFeCoNbB(15 nm)$Fe_{65}Co_{35}$(y nm)] $\times$20/Ti(20 nm)/(Si substrate) multilayers annealed at 625 °C for 1 min. It can be seen that for a 2 nm thickness of the FeCo layers, the magnetic behavior is similar to that of single layer, because the hard phase is dominant in this multilayer. However, the remanence is higher than that of single layer. The saturation magnetization and remanence enhance with increasing thickness of FeCo alloy, and the remanence is up to 692 emu/cc for $y=10$. It is found that the remanence of the film with $y=6$ is slightly higher than that of the above-mentioned Co-containing series due to the higher magnetization of FeCo than Co. In contrast, the coercivity of the former is only 5.7 kOe, which is remarkably lower than the latter. The ratio M_r/M_s for all the samples is higher than 0.65. The enhancement of the magnetic properties in the nanocomposite multilayer films is attributed to the

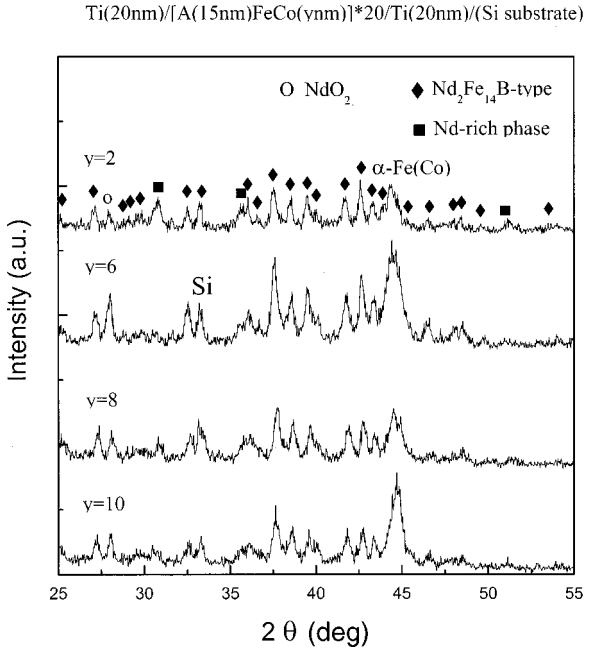

FIG. 3. XRD patterns for the thin film of Ti-buffered NdDyFeCoNbB/$Fe_{65}Co_{35}$ multilayers on the Si substrate annealed at 625 °C for 1 min.

exchange coupling between the magnetically soft and hard phases.

To study the nanostructure of the multilayer magnets, a TEM observation with a plane view was conducted. Figure 5 shows a TEM bright-field image of the Si/Ti(20 nm)/[A(15 nm)$Fe_{65}Co_{35}$(6 nm)]×20/Ti(20 nm) multilayer thin film annealed at 625 °C for 1 min. Two types of grain morphology were observed, i.e., relatively small grains (labeled as A) and large ones with irregular contrasts (labeled as B). Selected-area electron diffraction analyses reveal that the relatively small grains (type A) represent the hard phase with $Nd_2Fe_{14}B$-type structure and the large grains (type B) represent the soft phase of the α-Fe(Co) with the cubic structure in agreement with the results of XRD. Average grain size of the hard phase is between 10 and 15 nm, but the average grain size of the soft phase is larger than that. In order to improve the magnetic properties of the multilayer nanocomposite magnets, it is necessary to effectively control the grain size of the soft phase.

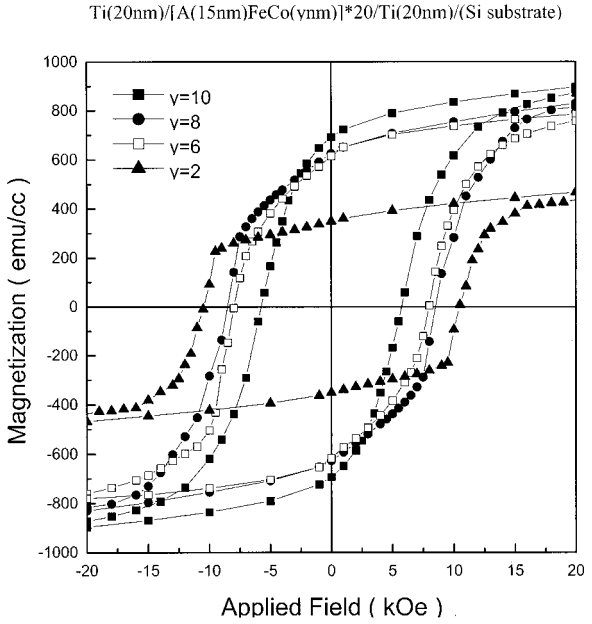

FIG. 4. Hysteresis loops at room temperature for the thin films of Ti-buffered NdDyFeCoNbB/$Fe_{65}Co_{35}$ multilayers on the Si substrate annealed at 625 °C for 1 min. Magnetic measurements are in the film plane.

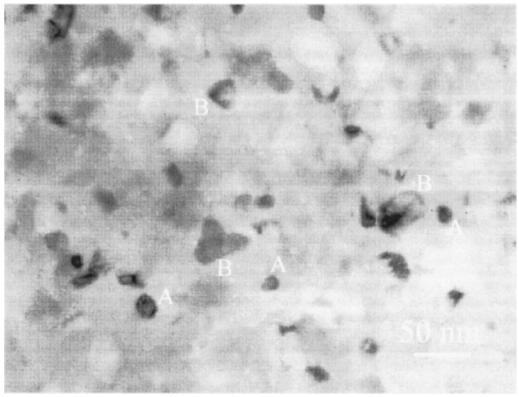

FIG. 5. TEM bright-field image of the Si/Ti(20 nm)/[A(15 nm)$Fe_{65}Co_{35}$(6 nm)]20/Ti/(20 nm) multilayer thin film annealed at 625 °C for 1 min.

In conclusion, the structure and magnetic properties of (Nd,Dy)(Fe,Co,Nb,B)$_{5.5}$/M (M=Co,FeCo) multilayer magnets have been prepared by sputtering and subsequent heat treatments. The hard magnetic phase of $Nd_2Fe_{14}B$-type coexists with a large amount of soft phase (Co phase and FeCo alloy) in the multilayer magnets. In comparison with the case of single layer, the remanence of the multilayer magnets is noticeably enhanced. The enhancement effect has been observed in all of the multilayer magnets due to exchange coupling between the soft and the hard phases.

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