Microstructure and magnetic properties of multilayer [(Nd$_2$Fe$_{14}$B)$_x$/M]$_n$ films (M=Nb or Cr)

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Multilayer [(Nd$_2$Fe$_{14}$B)$_x$/M]$_n$ films (x being thickness in nm; M=Nb or Cr, with a fixed thickness 5 nm; $n$ being the number of layers) have been deposited by ion-beam sputtering. Spacer layer was found to have substantial effects on the multilayers to exhibit different magnetic properties and microstructure. The average grain size of the multilayer films can be well controlled by both annealing temperature and thickness of the Nd$_2$Fe$_{14}$B layer. From a cross-sectional HRTEM study, the average grain size of [(Nd$_2$Fe$_{14}$B)$_x$/M]$_n$ films is 50 nm for $x=40$ nm, $n=10$, that is smaller than the grain size of single-layer Nd$_2$Fe$_{14}$B films with 200 nm thickness. The average grain size of single-layer film is 160 nm. From selected area diffraction patterns, the structure of Nb spacer is amorphous, while that of Cr is columnar, crystalline, and with some soft magnetic Fe–Cr phase in the broken layer structure around the hard magnetic Nd$_2$Fe$_{14}$B phase. Therefore, the multilayer films with Nb spacer exhibit good hard magnetic properties. While for those with Cr spacer, random exchange-coupling effect was observed in demagnetization curves. © 2002 American Institute of Physics. [DOI: 10.1063/1.1449448]

INTRODUCTION

Nanophase hard magnetic material especially the rare-earth transition (RE-TM) metal and intermetallic compounds have been extensively studied due to their potentially wide applications. The motivation in research comes from size dependence of coercivity, $H_c$, highly magnetocrystalline anisotropy of hard magnets, and complicated microstructure. Hard and soft phases exchange-coupled nanocomposites have been emphasized in bulk magnets and ribbons. Therefore, microstructure variation particularly on interface and grain boundary of thin film magnets is an interesting subject in the processing of RE-TM permanent magnets. Different underlayers have been used to control the texture of hard magnetic films. In RE-TM thin films the grain size has been controlled by annealing temperature and film thickness. Granular multilayer NdFeB/W and SmCo/W have also been reported.

Textured Nd–Fe–B films prepared at high temperature or subsequently annealing on different underlayers have been published in our previous works. In this paper, spacer layer effect on multilayer [(NdFeB)$_x$/M]$_n$ (M=Nb or Cr) films will be discussed, especially on the magnetic properties and microstructure and the role of either Nb or Cr spacer layer structure.

EXPERIMENT

Multilayer [(Nd$_2$Fe$_{14}$B)$_x$/M]$_n$ (x being the thickness in nm; M=Nb or Cr, with a fixed thickness 5 nm; $n$ being the number of layers; and $xn=400$ nm) films were prepared by an ion beam sputtering method. The base pressure of the vacuum chamber is $1.5 \times 10^{-8}$ Torr. The working pressure was $7.5 \times 10^{-5}$ Torr using a high-purity argon source (99.999%). The thickness of hard magnetic layer Nd$_2$Fe$_{14}$B was varied from 25 nm to 200 nm. The total thickness of the NdFeB hard layer was fixed at 400 nm. Commercially available sintered Nd$_{15}$Fe$_{77}$B$_8$ magnet was used as the target. Si(111) wafers cleaned with a standard IC process were used as the substrates. All the films were deposited at room temperature and then subsequently annealed. As-sputtered films were put in quartz tubes and pumped to a vacuum $2.3 \times 10^{-7}$ Torr. The annealing temperature was varied from 823 K to 973 K for 20 min.

The structure of films was identified by an x-ray diffractometer (XRD), the SIMENS D5000. The microstructure of films was investigated by high-resolution electron microscopy (HRTEM) (JEOL400FX). Chemical analysis of films was performed by inductively coupled plasma spectroscopy (ICP, Spectroflame). Magnetic properties were measured by a vibration sample magnetometer (VSM, EG&G Model-PAR 4500), and by a SQUID (Quantum Design MPMS SQUID Magnetometer) in the range 5–400 K with the maximum applied field 2 T, 5.5 T, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the hysteresis loops of [NdFeB$_{200}$ nm/M]$_2$ films annealed at 903±3 K for 20 min. The coercivity ($H_c$) and remanence ratio ($M_r/M_s$) of...
Nd$_2$Fe$_{14}$B film with Nb spacer layer were 9.0 kOe and 0.78, respectively. High crystalline-anisotropy Nd$_2$Fe$_{14}$B grains dominate the magnetization reversal process and performed the near square loops. However, the shoulder was found significantly affected in the near square loops. However, the shoulder was found significantly effected in the [NdFeB$_{200}$mm/Nb, Cr]$_2$ (M=Nb or Cr) measured at room temperature. 

![FIG. 1. Hysteresis loops for the multilayer [(NdFeB)$_{200}$mm/M]$_2$ (M=Nb or Cr)] measured at room temperature.]

Nd$_2$Fe$_{14}$B film with Nb spacer layers will inhibit the grain growth of Nd$_2$Fe$_{14}$B grains and enriches the grain boundary. 

The average grain size of [Nd$_2$Fe$_{14}$B$_x$/M]$_n$ (M=Nb or Cr) multilayers was found to be well controllable by both annealing temperature and thickness of the Nb$_2$Fe$_{14}$B layer. From a TEM cross-section view observation as shown in Figs. 3(a) and 3(c), the average grain size of [Nd$_2$Fe$_{14}$B$_x$/M$_{5}$nm]$_n$ films was 50 nm for $x=40$ nm, $n=10$. It is smaller than the grain size of single-layer films with thickness 200 nm. The average grain size of the single-layer film is 160 nm. The various gray regions are originated from the different crystallographic orientations of the isotropically distributed grains. The difference of the multilayer

![FIG. 2. $H_c$ and $M_r/M_s$ vs number of Nb spacers ($n$).]

Table I lists the magnetic properties and existing phases of [NdFeB$_{x}$/Cr$_{5}$nm]$_n$ films annealed at 883±3 K and 903±3 K for 20 min. respectively. The coercivity enhancement can be understood from the fact that for an NdFeB film and Nb spacer layer with fixed thickness, the increasing number of Nb spacer layers will inhibit the grain growth of Nd$_2$Fe$_{14}$B grains and enriches the grain boundary.

The average grain size of [Nd$_2$Fe$_{14}$B$_x$/M$_{5}$nm]$_n$ films was 50 nm for $x=40$ nm, $n=10$. It is smaller than the grain size of single-layer films with thickness 200 nm. The average grain size of the single-layer film is 160 nm. The various gray regions are originated from the different crystallographic orientations of the isotropically distributed grains. The difference of the multilayer

![FIG. 3. Cross-sectional HRTEM micrographs of [(Nd$_2$Fe$_{14}$B)$_{10}$mm/M]$_{10}$] films. The annealing temperature of films are at 903 K for 20 min. (a) M=Nb, (b) SAD pattern of (a), (c) M=Cr, (d) SAD pattern of (c).]
Nd$_2$Fe$_{14}$B films with Nb or Cr spacer layer lies in the structure of spacer layer and the precipitation of second phases.

From selected area diffraction pattern as shown in Fig. 3(b), one amorphouslike ring can be identified that is very close to Nb (110), and some small spots on ring can be indexed belonging to (313) of the Nd$_2$Fe$_{14}$B phase. The precipitation of the Nb-type second phase is also indexed around and enriched the grain boundary of Nd$_2$Fe$_{14}$B grains. Therefore, the layer by layer structure of [(Nd$_2$Fe$_{14}$B)$_x$/Nb$_5$nm]$_n$ film exhibits good hard magnetic behaviors. However, the structure of the Cr spacer layer is columnar and crystalline that can be proved in the lattice fringe in the HRTEM pattern. From the diffraction pattern in Fig. 3(d), complete grain growth without amorphous ring is manifest. The layer structure of [(Nd$_2$Fe$_{14}$B)$_x$/Cr$_5$nm]$_n$ films was partially deteriorated as observed in Fig. 3(c). As a result, soft magnetic Fe–Cr grains were formed around Nd$_2$Fe$_{14}$B grains or between interfaces. The volume fraction of the soft Fe–Cr phase was increased as being responded on the shoulder of the M–H curve in Fig. 1.

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