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Exchange coupling and remanence enhancement in nanocomposite Nd–Fe–B/FeCo multilayer films

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Abstract

Nd–Fe–B-type hard phase single layer films and nanocomposite $Nd_{28}Fe_{66}B_6/Fe_{50}Co_{50}$ multilayer films with Mo underlayers and overlayers have been fabricated on Si substrates by rf sputtering. The hysteresis loops of all films indicated simple single loops for fixed Nd–Fe–B layer thickness (10 nm) and different FeCo layer thickness ($d_{FeCo} = 1$–50 nm). The remanence of these films is found to increase with increasing d_{FeCo} and the coercivity decrease with increasing d_{FeCo}. It is shown that high remanence is achieved in the nanocomposite multilayer films consisting of the hard magnetic Nd–Fe–B-type phase and soft magnetic phase FeCo with $20\,nm \geqslant d_{FeCo} \geqslant 3\,nm$. The sample of maximum energy product is 27 MG Oe for $d_{FeCo} = 5$ nm at room temperature. The enhancement of the remanence and energy products in nanocomposite multilayer films is attributed to the exchange coupling between the magnetically soft and hard phases.
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1. Introduction

Exchange-spring nanocomposite magnets have attracted a significant amount of attention in the last decade [1–5]. The exchange-spring magnets are composed of suitably nanodispersed hard and soft phases that are exchange coupled at the interface. According to the micromagnetic theory of exchange coupling [1,2], it is expected that a high remanence and large values of coercivity would be obtained if grains of the soft and hard phases could be coupled well in nanocomposite magnets. However, appropriate control of the nanoscale microstructure is required to take full advantage of the attributes of both phases, since the exchange coupling depends strongly on microstructural

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factors. Generally, in contrast to bulk magnets, the structure of the multilayer materials can be easily controlled during the preparation process by properly arranging different layers, adjusting the thickness of the layers of the hard and soft phases, and annealing at the proper temperatures. This leads to an optimized average grain size and grain-size distribution. Up to now, the energy products of nanocomposite magnets prepared by means of rapid quenching and mechanical alloying have been much lower than theoretically expected, because of difficulties in controlling the nanostructures in nanocomposite bulk magnets [5–8]. Various methods of film deposition such as rf sputtering [9,11–13], dc sputtering [10,14,15], molecular beam epitaxy (MBE) [16], as well as laser ablation [17] have opened up possibilities for realization in nanocomposite magnets such as micromechanical devices [18], magnetic recording and magnetoelectronic devices [19], where films of a micron or more in thickness are required.

Recently, some studies on exchange coupling were carried out for nanostructured (Nd,Dy)(Fe,Co,Nb,B)$_{5.5}$/M (M = $Fe_{65}Co_{35}$,α-Fe) multilayer films prepared by sputtering and subsequent annealing [20,21]. Magnetic properties of exchange-coupled α-Fe/Nd–Fe–B multilayer film magnets were reported by Shindo and Ishizone [22], the observations for the Nd–Fe–B/Fe/Nd–Fe–B trilayer system were carried out by Parhofer et al. [23] and Yang et al. [24]. It is found that further studies are necessary in developing nanostructured multilayer magnets.

In this paper, we report on the magnetic and structural properties of $Nd_{28}Fe_{66}B_6/Fe_{50}Co_{50}$ multilayer films with Mo underlayers and overlayers, where Nd–Fe–B is the "hard" phase with a coercivity of $H_c \approx 20$ kOe and a magnetization of $M_r \approx 955$ emu/cc. The FeCo was chosen as the "soft" phase with $M_r \approx 1400$ emu/cc and negligible coercivity.

2. Experimental

$Nd_{28}Fe_{66}B_6/Fe_{50}Co_{50}$ multilayer films with different FeCo layer thickness, with Mo underlayers and overlayers, were fabricated in a multiple-gun sputtering system. All the films were sputtered on Si substrates by dc (Nd–Fe–B, Mo) and rf (FeCo) sputtering guns. The FeCo target was made by sintering pressed Fe and Co powders in vacuum ($\approx 10^{-6}$ Torr) at a temperature of 900 °C for ≈1 h. The Nd–Fe–B target was made by sintering pressed Nd, Fe and B powders in vacuum ($\approx 10^{-6}$ Torr) at a temperature of 1100 °C for ≈1.5 h. The Mo target was commercially obtained and had 99.95% purity. The base pressure of the sputtering system was 6×10^{-7} Torr and the Ar pressure during sputtering was 2×10^{-4} Torr. The $Nd_{28}Fe_{66}B_6/Fe_{50}Co_{50}$ multilayer films have the form Mo(5 nm)/[$Nd_{28}Fe_{66}B_6$ (10 nm)/$Fe_{50}Co_{50}$(d_{FeCo})] × 20]/Mo(40 nm)/Si, with $d_{FeCo} = 1 - 50$ nm. All the as-deposited films were annealed in a furnace at 873 K for 15 min at a pressure below 4×10^{-4} Torr. The crystalline structure of the phases in the films were determined by X-ray diffractometry (XRD) with Cu–Kα radiation. The thickness of the films was measured by means of Auger electron spectroscopy (AES). Magnetic properties of the films were measured by a superconducting quantum interference device (SQUID) magnetometer.

3. Results and Discussion

Fig. 1 shows the XRD patterns of the Mo(5 nm)/[$Nd_{28}Fe_{66}B_6$(10 nm)/$Fe_{50}Co_{50}$(d_{FeCo} = 5–50 nm)] × 20/Mo(40 nm)/Si multilayer annealed at 873 K for 15 min. Apart from the diffraction peak of the FeCo (1 1 0) at $2\theta = 44.87°$, the most intense peaks of the diffractogram can be attributed to the $Nd_2Fe_{14}B$ phase. The diffraction pattern shows clearly that the preferred orientation of the Nd–Fe–B crystals is the *c*-axis, with some of the (2 1 4), (2 0 4) and (1 0 5) peaks and trace quantities of Nd-rich phase. The FeCo phase increases quickly with increasing thickness of FeCo layers. The Nd-rich phase decreases with increasing thickness of FeCo layer. As for the thinner films ($d_{FeCo} \leqslant 3$ nm), however, it is difficult to analyze the FeCo phase with XRD because of the low signal intensity.

In order to directly check the composition in the annealed samples, AES experiments have been performed during depth profile sputtering in