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Remanence Enhancement and Exchange Coupling in PrCo/Co Films

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Abstract—PrCo/Co multilayer thin films have been prepared by sputtering. After heat treatment, high coercivity is developed and the remanence is enhanced for the insertion of the Co layers and the exchange coupling between the hard phase and the soft phase, which leads to high energy products of the composite magnets. The nanostructure of the films has been studied by TEM. It is found that the grain size of the films with high energy products is in the range from 10 to 25 nanometers.

I. INTRODUCTION

Since the discovery of the experimental evidence for the intergrain exchange coupling between magnetically hard phase and soft phase[1], many experimental studies have been done in mechanically alloyed and rapid quenched rare-earth transition-metal systems with the purpose of finding high-performance permanent-magnet materials. However, these two methods have the technical difficulty in controlling the required nanostructures. This difficulty stimulated us to try to realize the ideal nanostructure and therefore better intergrain exchange coupling by preparing such materials by sputtering and subsequent heat treatment.

Recently, Malhotra et al. [2, 3] have investigated the magnetic properties of PrCo single-layer films with Cr underlayer and cover layer. Magnetic hardening has been observed in the films after a heat treatment and the coercivity value reached 8 kOe. However, the saturation magnetization is low after the heat treatment.

In this paper we report the magnetic properties of the PrCo/Co multilayer films after appropriate thermal processing.

II. EXPERIMENT

The PrCo/Co multilayer thin films with Cr underlayer and cover layer were prepared with a multiple-gun DC- and RF- sputtering system by depositing the targets onto high-temperature operational glass or silicon substrates. The underlayer and the cover layer are 500 angstrom and 100 angstrom thick, respectively. The PrCo target was made by compressing Co and Pr powders with the Pr:Co atomic ratio 2:7 and sintered at 1050°C for 1 hour. The Co and Cr targets are commercial products with purities higher than 99.9 at.%. The substrates were attached to a rotating table that was cooled by water during sputtering. The base pressure in the chamber is 2×10⁻⁷ Torr. Flowing high-purity argon gas was used for sputtering and the pressures were varied from 5 to 20 mTorr. The thicknesses of the films were measured by weighing the mass of the films and by low-angle X-ray diffraction. The as-deposited films were then heat treated in a furnace with a pressure of 2×10⁻⁷ Torr. The crystal structures of the films were studied by using high-angle X-ray diffraction with Cu-Kα radiation. The microstructures of the films were observed by a JEOL 2010 Transmission Electron Microscope. Magnetization loops were measured by a Micromag-2900 Alternating Gradient Force Magnetometer and a Quantum SQUID Magnetometer.

The sputtering parameters in this investigation (including the argon pressure, the power rates of sputtering targets and the distance between the targets and the substrates) were chosen to obtain a deposition rate of the PrCo target around 1 Å/s and the Co target around 2.5 Å/s. The thicknesses of PrCo and Co layers in each period of the multilayer were adjusted according to the desired compositions.
III. RESULTS AND DISCUSSION

X-ray diffraction patterns of the as-deposited films showed only peaks from the Cr cover layer. This is consistent with the result in reference [3] that the as-deposited PrCo layer is amorphous. The coercivity \( H_c \) in these films is less than 100 Oe. The films were then annealed at 500°C for 40 minutes in vacuum. Considerable coercivity development after the heat treatment. Figure 1 shows an example of the hysteresis loops. From the loop measured in the parallel direction we can see that the ratio \( M_p/M_s \) is larger than 0.5 (around 0.8), indicating an intergranular exchange coupling. On the other hand, the films are magnetically anisotropic with the easy direction in the film plane, which may be due to the magnetocrystalline anisotropy of the PrCo grains.

![Fig. 1. Hysteresis loops measured in different directions of the film (PrCo30nm/Co5nm)x10.](image)

It is interesting to note that the coercivity of the films with the Co layers is even higher than the single-layer films reported in reference [3], if the thickness of Co layer is not large. The maximum coercivity is 10 kOe. The previous work [3] has shown that the magnetic hardening originates from the crystallization of the PrCo grains with a hexagonal structure. It appears that the insertion of Co layers does not have any negative influence on the magnetic hardening if their thickness is small enough. Figure 2 shows an example of the dependence of the coercivity on the Co layer thickness. It is seen that when the Co layer thickness is less than 6 nm, the coercivity is relatively high, which may be connected to the theoretical prediction that the intergranular exchange coupling remains strong if the dimension of the soft phase is less than twice that of the domain-wall thickness in the hard phase[5]. The typical wall thickness in a hard rare-earth transition-metal phase is about 3 nm. In this experiment, it can be easily realized to put more than 20% soft Co phase into the film without leading to the Co grain size larger than 6 nm, because the thickness of the PrCo layers can be adjusted.

Yet the insertion of the Co layer enhances the magnetization remarkably. As shown in Fig. 2, the saturation magnetization nearly doubles when the thickness of Co layer increases to 8 nm. The same trend was found for the remanent magnetization. Considering the decrease of the coercivity, the Co layers with thickness near to 6 nanometers bring optimal magnetic properties.

![Fig. 2. Dependencies of coercivity and the saturation on Co layer thickness in the film (PrCo28nm/Co)x8.](image)

Transmission Electron Microscopy (TEM) and Electron Diffraction have been performed to investigate the microstructure and the crystalline structure. Figure 3 shows the dark field TEM picture of the sample whose hysteresis loops have been shown in Fig. 1. From the picture one can find that the grain size in the annealed film is in the range of 10-25 nm. The electron diffraction and the energy dispersive X-ray spectroscopy (EDX) studies indicate that the crystal structure of the majority grains in the film is hexagonal close-packed structure with ordering of Pr in Co lattice[4]. This result is consistent with the result in reference [3]. The EDX study has also indicated the existence of Co phase.
From the magnetization measurement (see Fig. 2), we can also infer the existence of the Co phase or a Co-rich phase.

Fig. 3. Dark field TEM picture of the annealed film (PrCo30nm/Co5nm)x10.

The coexistence of the magnetically hard phase(PrCo) and soft phase(Co) may result in exchange coupling if the grain size of the soft phase is in a suitable range. Calculations[5] have shown that the exchange coupling may provide the possibility to produce the composite magnets with very high energy products. In best situation in this investigation, we obtained the maximum energy product of the film up to 20 MGOe at room temperature.

IV. CONCLUSIONS

By sputtering and proper heat treatment, PrCo/Co composite films with energy product up to 20 MGOe have been successfully produced. The intergrain exchange coupling has been observed in the films in which the grain size is well controlled. The results presented in this paper provide further evidence that nanostructural multilayer plus appropriate thermal processing can lead to novel structure with favorable magnetic properties.

REFERENCES