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Rapidly annealed exchange-coupled Sm-Co/Co multilayers

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In exchange-coupled two-phase permanent magnets, the length scale of soft phase is limited to about twice of the domain-wall width of the hard phase. To optimize the energy product, it is important to realize this length experimentally. In this work, we investigate the Sm–Co/Co hard/soft multilayers with varying thickness of soft phase layers. On rapidly annealing, the multilayered hard-soft structure forms. Transmission electron microscopy micrography confirms that the multilayer structure is retained after the annealing. Single-phase-like hysteresis loops are obtained for samples with Co layers up to 13 nm thick. This behavior indicates that the soft phase is well exchange coupled to the neighboring $SmCo_z$ hard phase. An optimal energy product of 16.6 MGOe has been obtained. Longer annealing time results in more diffusion at the interface and yields two-phase-like hysteresis behavior. Direct current demagnetization measurement shows exchange-spring behavior of the samples annealed for longer time. Micromagnetic simulations with varying interface exchange coupling have been performed to compare with the experimental results. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850814]

I. INTRODUCTION

Since the concept of exchange-spring magnets was developed in 1991,[1] the exchange-coupling interaction between magnetically hard and soft phases has attracted much attention in searching for novel permanent magnets.[2,3] Usually, Nd–Fe–B, Pr–Fe–B, $SmCo_5$, $Sm_2Fe_{17}N_3$, $L1_0$–FePt are considered as candidates for hard phases, while Fe, Co, Fe–Co and other soft phases are used to improve the magnetization.[4,5] Much effort has been given to investigating exchange-coupling between hard and soft phases in the forms of multilayers,[6–8] mechanically alloyed powders,[9] chemically systhesized nanoparticles,[10] and hard-soft nanocluster composites.[11]

With the highest bulk anisotropy constant ($K_1 = 17\ \mathrm{MJ/m^3}$), $SmCo_5$ is among the most investigated hard phases in preparing exchange-coupling magnets. Its large anisotropy can help retain the needed coercivity value while the soft phase improves the magnetization. Several groups have performed studies of sputtered SmCo/Co multilayers.[6–8,12,13] However, there has not been a report on the energy product in Sm–Co/Co multilayer thin films, and neither has there been a report on the effects of rapid annealing. In this article, we used rapid thermal annealing to control the hardness of hard phase, as well as the diffusion at interfaces in an attempt to understand the influence of interfaces on exchange coupling. Micromagnetic simulations show that exchange coupling between hard and soft layers depends on the interface, magnetic hardness and soft layer thickness.

II. EXPERIMENTAL METHODS

The multilayer thin films are prepared by magnetron sputtering. Si (100) wafers are used as substrates. $SmCo_4$ and $SmCo_5$ films are prepared by setting different sputtering rates. Cr and Ti under- and cover layers are deposited. A typical layer structure is Cr(Ti)40 nm/(SmCo14 nm/Co7 nm)$\times$7/Cr(Ti)10 nm. Co layers range from 4 to 14 nm. The total thickness of magnetic layers is around 150 nm. The samples are thermally processed by rapid thermal annealing (RTA) at 525 °C for 30 s–5 min, or by furnace for 30 min. The crystal structure of the samples is characterized by x-ray diffraction and transmission electron microscopy (TEM). The sputtering rate is determined by weighing method and the sample thickness is confirmed by the cross-section view of TEM. A superconducting quantum interference device (SQUID) magnetometer with highest field of 70 kOe is used to measure magnetic properties.

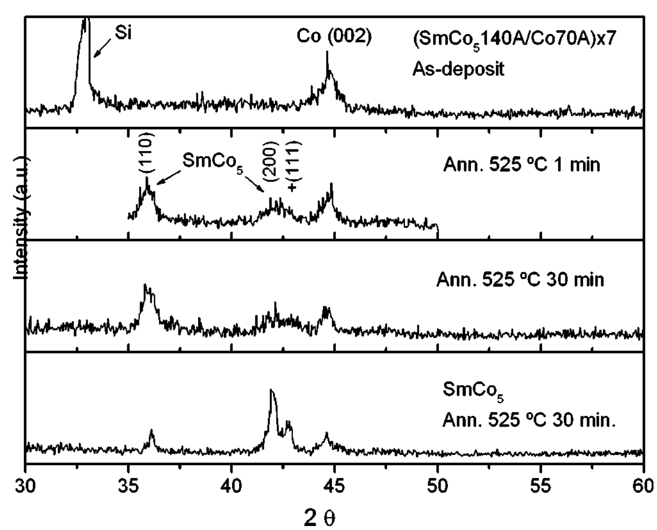

FIG. 1. XRD of [$(SmCo_5)$14 nm/Co7 nm]$\times$7 and $SmCo_5$ films.

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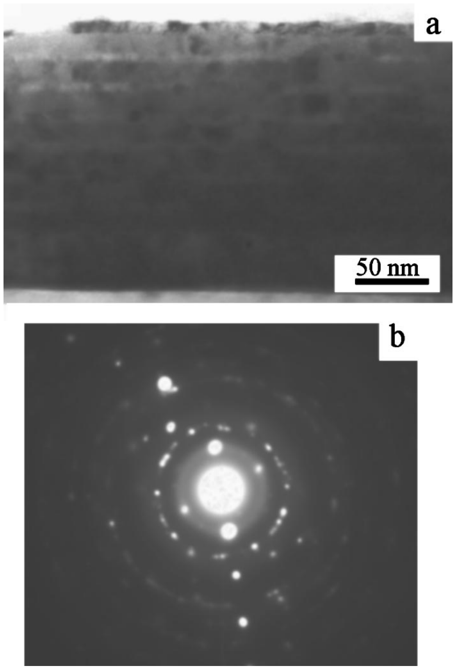

FIG. 2. (a) TEM image and (b) diffraction pattern of Si/Cr40 nm/[$(SmCo_4)$14 nm/Co8 nm]$\times$6. The 8 nm Co layer remains after heat treatment. Diffraction pattern shows the film is polycrystalline.

III. RESULTS AND DISCUSSION

The as-deposited samples are amorphous under x-ray diffraction (XRD). After annealing for 1 min at 525 °C, the Sm–Co hard phase forms. Figure 1 shows the XRD of a typical film. In Fig. 1(a) for the as-deposited film only the Co (111) peak appears. After 1 min rapid annealing at 525 °C, the Sm–Co phase appears, as shown in Fig. 1(b). Figure 1(c) shows the long-term annealing indicating both Sm–Co and Co phases still coexist. As a comparison, single-phase SmCo is shown in Fig. 1(d). The missing (101) peak of $SmCo_5$ at 30.5° and the strengthened (200) peak at 42° indicate that the hard phase $SmCo_5$ easy axis (c axis) is partially in the film plane. This result is consistent with our previous report.[6]

Further investigation by TEM shows the layer structure in rapidly annealed sample. Figure 2(a) shows the cross-section view of the annealed multilayer Cr40 nm/[$(SmCo_4)$14 nm/Co8 nm]$\times$6/Cr10 nm. It is clearly seen that the hard and soft layers remain distinct after the short-term annealing, with interfaces that are sharp to within about 1 nm. The diffraction pattern [Fig. 2(b)] shows the film is polycrystalline. Since the annealing time is short and annealing temperature is moderate, the layers have relatively sharp interfaces because the diffusion of atoms between layers is limited.

Magnetic hysteresis loops measured by SQUID magnetometer show the effect of annealing and layer thickness on the magnetic properties. Figure 3 shows hysteresis loops of two annealed films, [$(SmCo_4)$14 nm/Co8 nm]$\times$6 and [$(SmCo_4)$14 nm/Co13 nm]$\times$5. Both samples are annealed at 525 °C for 1 min. Both loops are measured in the film plane. It can be seen that the two hysteresis loops show single-phase-like behavior which indicates strong exchange coupling between hard and soft phases. All our samples annealed under this condition with Co layer thickness less than 13 nm show single-phase behavior. However, the sample with 14-nm-thick Co layer begins to show a shoulder kink at a small reversed field which indicates the exchange coupling is poor because the Co thickness is beyond the exchange-coupling range, which in this situation is 13 nm of Co. It is shown in the figure that the increased soft phase Co (13 nm layer) enhances magnetization compared to the film containing less Co (8 nm Co layer). Even though the coercivity is only 6.7 kOe, a higher energy product of 16.6 MGOe is achieved.

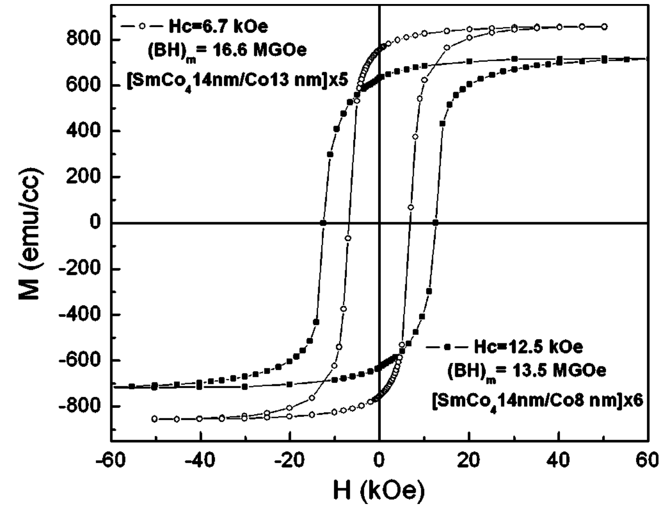

FIG. 3. Hysteresis loops of two annealed samples at room temperature.

The exchange-spring magnets limit the soft grain size to twice the hard phase domain wall width, which is $\pi(A/K_1)^{1/2}$.[14] For $SmCo_5$, the soft grains should be limited to 7.2 nm. In our situation, since the annealing time is short, very likely the magnetic hardening is not fully realized in the Sm–Co hard phase. The coercivities of 150 nm single layer $SmCo_4$ and $SmCo_5$ are 32 and 23 kOe, respectively, much lower than that of the long-term annealed samples (39.3 and 37.5 kOe, respectively[6]). Thus the anisotropy constant K_1 is not as large as that of bulk $SmCo_5$. Assuming the exchange

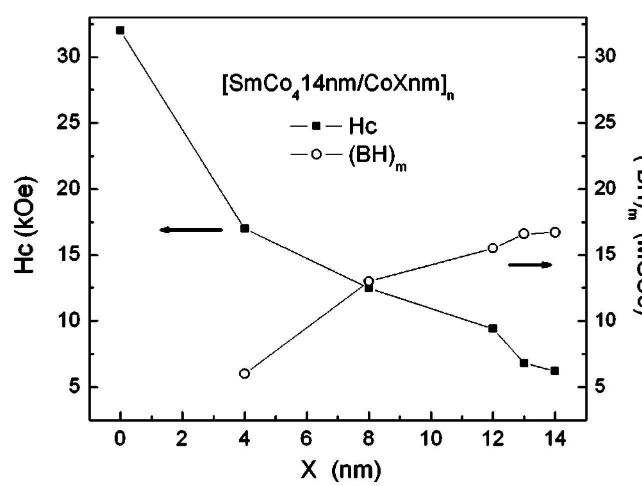

FIG. 4. Soft phase dependence of coercivity and energy product in annealed [$(SmCo_4)$14 nm/CoX nm]$\times n$ film.

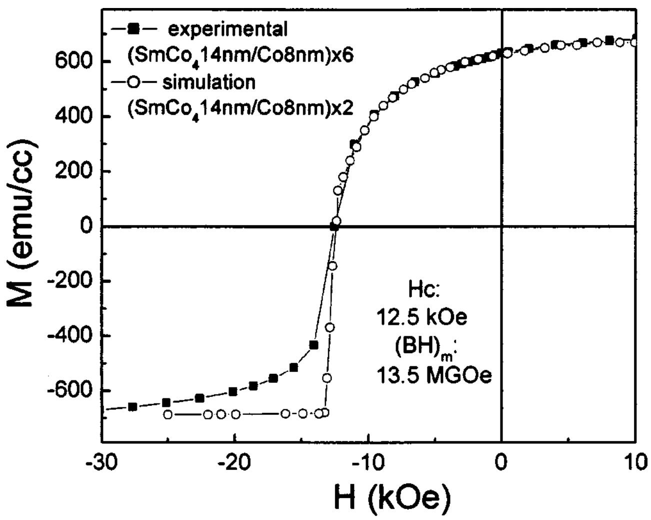

FIG. 5. Simulated and experimental demagnetization curves of rapidly annealed [$(SmCo_4)$14 nm/Co8 nm]$\times n$ film.

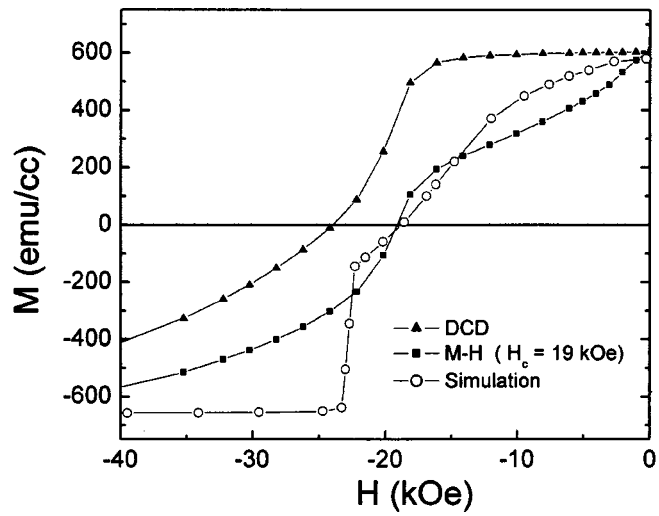

FIG. 6. Simulated and experimental demagnetization curves of annealed (30 min) [$(SmCo_4)$14 nm/Co8 nm]$\times n$ film.

constant remains unchanged, a K_1 value of 5 MJ/m^3 could yield a domain wall width of 6.5 nm, corresponding to the soft grain size of 13 nm.

The Co layer thickness dependence of coercivity and energy product is shown in Fig. 4. The samples have layer structures of [$(SmCo_4)$14 nm/CoX nm]$\times n$. As expected, more soft phase drops the coercivity but increases the magnetization as well as remanence so that the energy product increases with Co layer thickness. The largest energy product is obtained in 13-nm-thick Co. Beyond 13 nm the two-phase kink appears indicating the phases are not well exchange coupled.

Numerical micromagnetic simulations are performed using the National Institute of Standards and Technology OOMMF code to compare to the experimental results. Figure 5 shows the simulated reversal curve compared to the corresponding experimental curve. (For the simulation, parameters are chosen as the following: for $SmCo_4$, K_1 =1.5 MJ/m^3, A=25 pJ/m, Ms=0.45 MA/m; for Co, K_1 =0.3 MJ/m^3, A=10 pJ/m, Ms=1.2 MA/m.) The simulated curve matches the experimental results well.

Longer annealing time will result in further hardening of the Sm–Co hard phase, as well as atomic diffusion at layer interfaces. Annealing at 525 °C for 5 min causes a small shoulder at remanence, while annealing for 30 min in furnace causes the two-phase-like behavior as shown in Fig. 6, suggesting that the soft phase starts to reverse first, due to the reduced anisotropy of the Co-rich interface. As one can see from the DCD curve, the magnetization reversal of the soft phase is reversible before the negative field reaches 18 kOe, indicating the exchange-spring recoil behavior for the soft phase. The moment is reversed back to remanence by the coupling of hard phase when the negative field is removed. A simulated demagnetization curve (circled line) shows similar behavior. The consideration of 1-nm-thick interface layer between hard and soft layers is included in the simulation. Further investigation of the effect of the magnetic hardening and interface on the exchange coupling is under way.

IV. SUMMARY AND CONCLUSIONS

In this article we report our work on rapidly annealed SmCo/Co multilayers. The 1 min annealing at 525 °C retains the layer structure unchanged and the hard phase of Sm–Co forms. The hard and soft layers are exchange coupled. Co layer as thick as 13 nm can be coupled to the hard-phase $SmCo_4$ in the samples. An energy product of 16.6 MGOe is achieved. Micromagnetic simulations results match the experimental hysteresis well. Longer annealing causes a more hardened Sm–Co phase, as well as diffusion between hard and soft phases. The latter effect can harm the permanent magnet properties of the system, whereas the former effect creates restriction on the soft-phase dimension.

ACKNOWLEDGMENTS

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