Coercivity mechanism of nanocomposite Sm-Co/Fe multilayer films

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The coercivity mechanism of nanocomposite Sm-Co/Fe multilayer films was investigated. It was concluded that the magnetization reversal mechanism of Sm-Co/Fe multilayer films is dominated mainly by domain wall pinning instead of nucleation. The addition of Cu layer between the hard and soft layers causes the change in grain boundary as well as the large variation in anisotropy in the hard layer after annealing, which leads to a strong increase in domain wall pinning, and therefore a significant improvement of the coercivity. This study indicates the potentials of this method to control and improve the magnetic property of nanocomposite multilayer films.
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I. INTRODUCTION

Nanocomposite magnets compose of the hard and soft magnetic phases. The hard magnetic phase provides a high coercive force while the soft phase offers a high magnetization. Therefore a very high energy product (BH)max could be possible for the nanocomposite permanent magnets. However, only when the hard and soft magnetic phases goes into a scale of nanometer and the exchange coupling between these two phases becomes extremely strong, a nanocomposite magnet with a high (BH)max can be achieved.[1] The nanocomposite magnets can be prepared by the techniques such as melt spinning,[2–4] mechanical milling,[5–7] sputtering,[8–11] nanoparticle self-assembly,[12] etc. Nanocomposite multilayer films prepared by sputtering are of the state of the art in permanent magnets and of high importance not only for understanding the mechanism of nanocomposite magnets but also for developing the next generation of magnets. A theoretical study shows that the anisotropic nanocomposite multilayer films with a high fraction of soft phase can achieve a giant (BH)max as high as 120 MGOe.[13] However, it is extremely hard to fabricate this kind of films with such high performance from the experimental points of view. Fullerton *et al.*[8] have successfully prepared the epitaxial Sm-Co/Co superlattice films via magnetron sputtering with well aligned hard-phase layers. Due to a low fraction of soft phase, their (BH)max was still not very high. In comparison with Sm-Co/Co layer films,[14] Sm-Co/Fe layer films[10,15] with aligned thinner hard layers and high fraction of soft phase can be achievable after postannealing due to a comparatively hard interlayer mixture in Sm-Co/Fe multilayers during annealing. Therefore, a higher (BH)max has been obtained in Sm-Co/Fe double-layer and multilayer films.[10,15] For anisotropic Sm-Co/Fe multilayer films, a high coercivity of 7.24 kOe and a large (BH)max of 32 MGOe has been achieved, which experimentally demonstrates that a high-performance magnets could be achievable by fabricating the well textured nanocomposite multilayer films.[10]

The understanding of the coercivity mechanism of permanent magnets is of importance not only for basic research but also for technological application. Basically, there are two models used to explain the reversal mechanism of the permanent magnets: the nucleation model and the pinning model. The coercivity mechanism reveals the origin of the coercivity and provides the basic knowledge and methods for further improving the magnetic properties of the magnets. The coercivity mechanism is dependent on the phase composition, sample preparation method, microstructure, etc. For example, the coercivity of the conventional $SmCo_5$ magnets is controlled by nucleation,[16] while the coercivity of the Sm_2Co_{17}-type magnets is determined by domain wall pinning.[17] Although the nanocomposite Sm-Co/Fe multilayer and double-layer films with desirable properties have been successfully prepared and extensively studied,[10,15,18–21] their coercivity mechanism is still an open question. The coercivity mechanism of Sm-Co/Fe nanocomposite films may be different from the Sm-Co single-phase magnet and the conventional Sm-Co magnets due to the presence of the soft phase. Moreover, it has been also found that the addition of Cu layer can significantly improve the coercivity of Sm-Co/Fe films. The reason for such improvement is not clear. The addition of Cu has been found to increase the pinning coercivity in Sm_2Co_{17}-type magnets.[17] However, the hard-soft nanocomposite magnet with high performance is a different system and its coercivity, generally, is believed to be controlled by the nucleation of the reversed domain.[22] Therefore, it is crucial to clarify the coercivity mechanism of the Sm-Co/Fe films.

In this paper, we report the systematic study of the coercivity mechanism of anisotropic Sm-Co/Fe nanocomposite multilayer films. We concluded that the coercivity mechanism of the films is mainly controlled by the domain wall pinning instead of the nucleation. The inserting Cu layer be-

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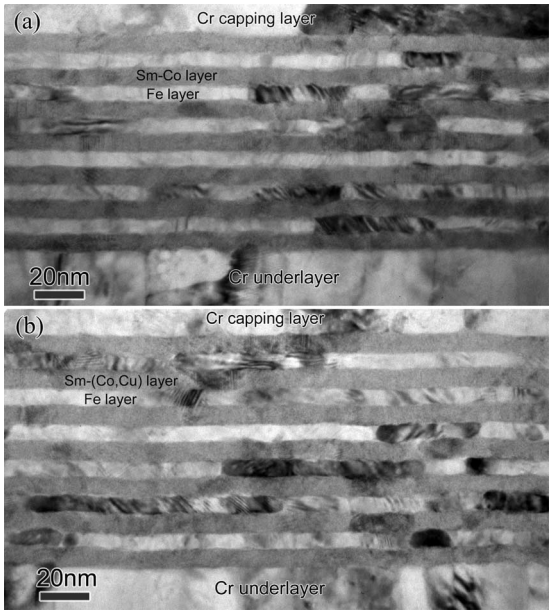

FIG. 1. TEM bright images of the cross sections of the films with x=0 (a) and 0.5 (b) annealed at 500 °C.

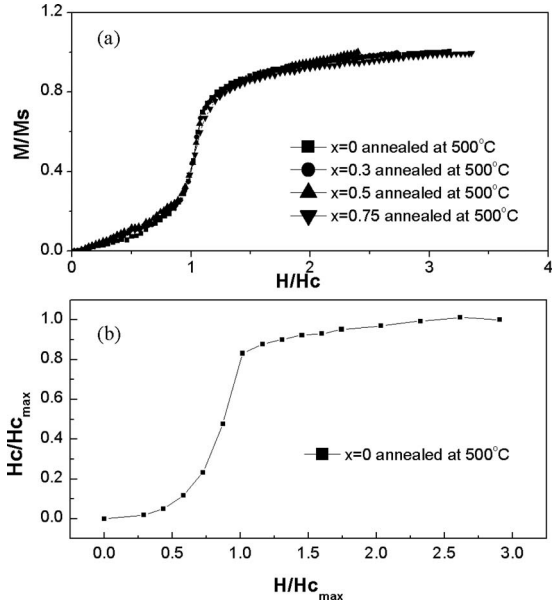

FIG. 2. Normalized initial magnetization curves for the films with x=0, 0.3, 0.5, and 0.75 annealed at 500 °C (a) and normalized dependence of the coercivity on the maximum applied field for the film with x=0 annealed at 500 °C (b) measued at RT. The applied field is along the film plane (the easy axis).

tween the hard and soft layers changes the grain boundary and increases the variation in anisotropy in the hard layer after annealing, leading to a strong increase in domain wall pinning and therefore, a significant improvement of the coercivity in the films. Our results not only clarify the coercivity mechanism of such high-performance nanocomposite films, but also provide a guide for further improving their magnetic property.

II. EXPERIMENTS

The samples Cr(50 nm)/$SmCo_6$(9 nm)/Cu(x nm)/Fe(5 nm)/Cu(x nm)/Cr(100 nm)/a-SiO_2(x=0–0.75) with varying Cu thicknesses were prepared by magnetron sputtering and subsequent annealing for 30 min. The experiments were described in detail in Ref. 10. The data of the angular dependency of hysteresis loops were measured using a Lakeshore vibrating sample magnetometer.

III. RESULTS AND DISCUSSION

Figure 1 shows the transmission electron microscopic (TEM) bright images of the cross section of the films with x=0 and 0.5 annealed at 500 °C. Both samples show the distinct multilayer structure even after annealing at 500 °C. This is the main reason why our samples have the very good magnetic properties.[10] Our films contain a very large amount of the soft phase. The thickness of the hard and soft layer can be corrected accurately by the cross-sectional TEM images shown in Fig. 1. Based on these images, we calculated the volume fraction of the soft phase in our films and it is about 40 vol % (≈39 wt %). Despite such a high content of soft phase, the very high coercivities of 6.3, 7.2, and 8.3 kOe are still achieved for the films with x=0 annealed at 500 °C, x =0.5 annealed at 450 and 500 °C, respectively. If we compare the multilayer films with the nanocomposite magnets prepared by mechanical milling (MM),[7] we found that even for the magnet with a fraction of 35 wt % soft phase prepared by MM, their coercivity is already very low (3 kOe). This demonstrates that the microstructure is of very importance for fabricating the nanocomposite magnets with desirable properties and the films with a multilayer structure are potentially beneficial for developing the high-performance magnets.

We investigated the coercivity mechanism of our multilayer films. Figure 2(a) shows the normalized initial magnetization curves for the films with x=0, 0.3, 0.5, and 0.75 annealed at 500 °C measured at room temperature (RT). Although the initial curves of these samples are different, their normalized curves show the same behavior and match each other very well. A jump appears when the applied field reaches the coercive field. This is a characteristic of the domain wall pinning. Figure 2(b) shows the normalized dependence of the coercivity on the maximum applied field, determined from the minor hysteresis loops for the film with x=0 annealed at 500 °C measured at RT. The coercivity changes slowly at the low applied fields but increase sharply at the applied field with around the maximum coercivity *H*max. In addition, in order to reach the maximum coercivity *H*max, the applied field is much larger than *H*max. Such behaviors are characteristics of domain wall pinning controlled magnets.

In order to further clarify the coercivity mechanism, the angular dependency of magnetic properties of the films at RT was investigated. Figure 3(a) shows the hysteresis loops for the sample with x=0 annealed at 500 °C measured at different angle θ. θ is the angle between the applied field and the film plane. The switching field *Hs* is defined as the maximum irreversible susceptibility in the magnetization reversal process. At the low angles, the switching field *Hs* is equal to the coercivity *Hc*, while at the high angle, *Hs* becomes much

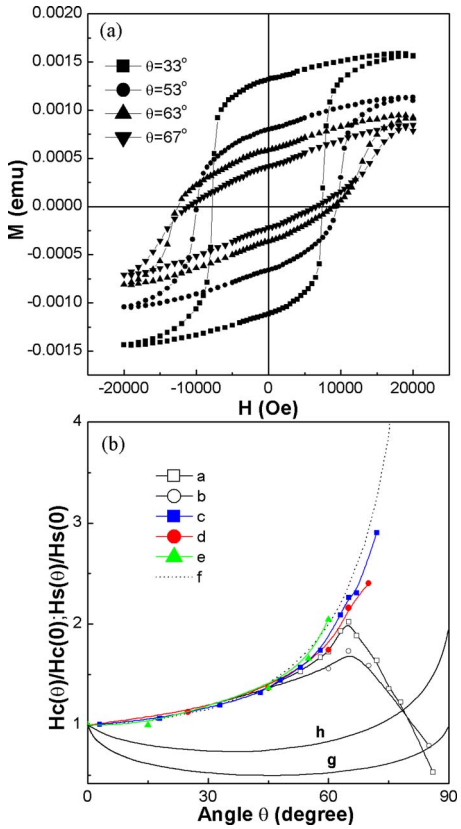

FIG. 3. (Color online) (a) Hysteresis loops at RT for the sample with x=0 annealed at 500 °C measured at different angle θ; θ is the angle between the applied field and the film plane. (b) a and b are the coercivity *Hc* as function of angle θ for the films with x=0 annealed at 500 °C and x=0.3 annealed at 450 °C, respectively; c, d and e are the switch fields *Hs* as function of angle θ for the films with x=0 annealed at 500 °C, x=0.3 annealed at 450 °C and x=0.5 annealed at 500 °C, respectively; curve f is the Kondorsky relation $1/\cos\theta$; curve g corresponds to the nucleation mechanism based on Stoner–Wohlfarth rotation; curve h corresponds to the modified nucleation process according to the Ref. 25.

different from *Hc*. The switching field can be accurately determined from the field derivative of the hysteresis loop. Figure 3(b) gives the *Hs* and *Hc* as function of the angle θ for some samples. The coercivity *Hc* increases with angle θ, reaches a maximum at 65°, then decreases. The switching field *Hs*, however, increases with θ continuously. These behavior are similar to what has been observed in single Sm-Co layer films.[23] The switch field *Hs* is attributed only from the irreversible process. It is of interest to note that the angular dependent switch field *Hs* for all measured samples fits quite well with an inverse cosine dependence (Kondorsky relation) which is associated with domain wall pinning,[23,24] but deviates far from the curves related to the nucleation process.[25] This gives the definite evidence that the coercivity of our films is mainly controlled by domain wall pinning instead of the nucleation.[23–25]

Based on the initial curves, the normalized dependence of the coercivity on the maximum applied field and, in particular, the angular dependency of the switching field *Hs*, it is clear that the coercivity of Sm-Co/Fe multilayer films is dominated mainly by domain wall pinning instead of nucleation. As the preparation procedure for both Sm-Co/Fe multilayer and double-layer films is almost the same, the pinning mechanism should be also suitable for the Sm-Co/Fe double-layer films.

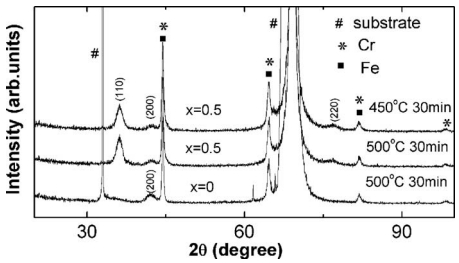

FIG. 4. XRD patterns for the annealed films with x=0 and 0.5.

It has been found that inserting a Cu layer between the hard and soft layers can significantly improve the coercivity of Sm-Co/Fe multilayer films at RT.[10] We also measured the magnetic properties of the films at the low temperatures. We found that the coercivity of the Cu doped films is much improved at the low temperatures in comparison with that at RT (at 10 K, *Hc*=12 kOe for x=0 annealed at 500 °C, *Hc* =18 and 21 kOe for x=0.5 annealed at 450 °C and 500 °C, respectively). We know that Cu goes totally into the Sm-Co hard layers after annealing.[10] It was calculated that Cu(0.5 nm)/$SmCo_6$(9 nm)/Cu(0.5 nm) corresponds to a nominal composition of $Sm(Co_{0.867}Cu_{0.133})_{6.92}$. Such high amount of Cu substitution basically decreases the anisotropy of the hard layer,[26,27] which, therefore, should not be responsible for the improvement of the coercivity. In addition, the coercivities of the films with x=0 annealed at 500 and 525 °C are almost the same value, so that the crystallization of the film with x=0 after annealed at 500 °C is almost complete. There must be other reasons for the significant improvement of the coercivity after the addition of Cu layer. As the coercivity mechanism of the multilayer films is mainly determined by domain wall pinning, the significant improvement of the coercivity must be ascribed to the efficient pinning of domain walls. Figure 4 shows the x-ray diffraction (XRD) patterns for the annealed films with x=0 and 0.5. The film with x =0 has only (200) texture, while the films with x=0.5 exhibits both (110) and (200) texture. It is highly likely that the variation in A and K in the grain boundary between (110) and (200) grains is larger than that between (200) grains, leading to a stronger pinning of domain wall in the film with x =0.5. Figure 5 presents the recoiling loops of the films with x=0 and 0.5 measured at 10 K. It is interesting to note that the opening of the recoiling loops for the Cu doped films is much larger than that of the film with x=0. It has been proved that for the nanocomposite layer films, the open recoiling loop is originated from the anisotropy variation in the hard phase.[18] The soft phase content and the grain boundary may also influence the opening of the recoiling loop.[28,29] As the thickness of the soft layer for both films with x=0 and 0.5 is the same, we can ignore the influence of the soft layer. The large opening of the recoiling curve for the sample with x=0.5 at 10 K must be attributed to the large variation in anisotropy in the hard layer[18] or the change in grain boundary[28] (the increase or property change in the boundary phase) which may also causes a strong pinning of the domain

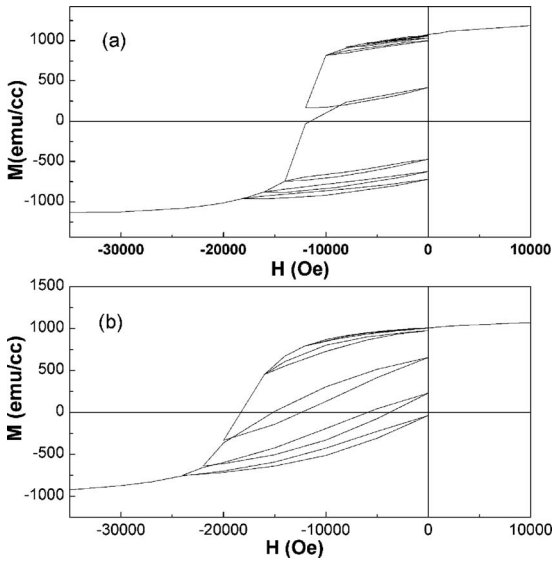

FIG. 5. Recoiling loops of the films with x=0 annealed at 500 °C (a) and x=0.5 annealed at 450 °C (b) measured at 10 K.

wall or an increase in pinning sites, and therefore a larger coercive field. It was also noticed that the squareness of the demagnetization curve of Cu doped films become worse (see Fig. 5), which also reflects the inhomogeneity in the hard layer. We add the Cu by inserting a Cu layer. During annealing, Cu will diffuse into the hard layer. This method can make an inhomogeneous distribution of Cu in the hard layer along normal direction of the film, leading to a large variation in the anisotropy in the hard layer and a high coercivity. By choosing the inserting layer with other materials, it can be also possible to realize a large variation in anisotropy and high coercivity. This represents a novel method for controlling the property of the nanocomposite films. The addition of Cu has been found to enhance the pinning coercivity of Sm_2Co_{17} magnets. The microstructure origin of the pinning in Sm-Co/Fe films, however, is different from that in Sm_2Co_{17} magnets because the microstructure of these two systems is different. In Sm_2Co_{17} magnets, Cu mainly goes into the cell wall for increasing the pinning coercivity.[17]

IV. CONCLUSION

It was concluded that the coericivity of the films is mainly controlled by the domain wall pinning instead of the nucleation. The inserting Cu layer leads to the change in grain boundary as well as the large variation in anisotropy in the hard layer, resulting in a strong pinning field of domain wall and therefore a large coercivity. Our study clarified the coercivity mechanism of nanocomposite Sm-Co/Fe layer films and gives the answer why the inserting Cu layer can significantly improve the coercivity of the films. It will be beneficial for tuning and improving the magnetic properties of the nanocomposite layer films.

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