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Influence of surface/interface roughness and grain size on magnetic property of Fe/Co bilayer

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In this work, we report the influence of surface roughness and cluster size on coercivity of Fe/Co bilayer. Coercivity was tuned by thermal annealing. No systematic trend was found for temperature dependent annealing. However, after annealing at 350 °C, we find systematic increase in coercivity with anneal time. For as-deposited film, we find unusually low coercivity (0.39 Oe). By increasing annealing time, coercivity was tuned to values as high as 600 Oe. Surface characterization using atomic force microscopy showed uniform clusters at this temperature after 2 h of annealing. The observed magnetic properties are discussed in terms of cluster size and surface/interface roughness. © 2005 American Institute of Physics. [DOI: 10.1063/1.2119427]

Magnetic thin films, in particular nanometer scale structures have drawn a lot of attention both for technological application and fundamental research. Magnetic properties of multilayer thin films are strongly influenced by microstructure such as thin film thickness, interface roughness and crystallinity.¹⁻³ Crystallographic modification is therefore an excellent tool for tuning properties.

Ion radiation¹ and thermal annealing⁴ are found to be the most effective ways to change surface and interface microstructure. Change in the ordering of the bilayers is also reported to influence magnetic properties.⁵

Previously, a large number of studies were completed on Fe/Co multilayer system. Dirne *et al.*⁶ showed that an interdiffused layer can be form during deposition of Fe/Co multilayer. Several thickness dependent studies^{5,7,8} are also available where shape and magnetocrystalline anisotropy are rigorously studied. However, we find no studies on anneal induced behavior of Fe/Co multilayer. Especially a systematic increase in coercivity induced by interface roughness and grain size has never been reported. In this letter, we report systematic time-dependent study on magnetic property of Fe/Co bilayer. We find that the coercivity can be nicely tuned from soft magnet to hard magnet. We also observe unusually low coercivity for as deposited Fe/Co bilayer.

Fe/Co films were deposited on Si(111) with native oxide using a thermal evaporator at the rate of $\sim 2 \text{ \AA/s}$ and a base pressure of 10^{-7} Torr. The layer thickness was 25 nm for each case. The total thickness was 100 nm. Order of deposition from substrate level is Fe/Co/Fe/Co. Annealing was performed in a convection furnace with 20 sccm nitrogen flow. Films were annealed at 350 °C with anneal time ranging from 5 to 300 min. Several high temperature annealing was also performed for comparison. However, no systemic trends were observed for all those cases. The post anneal samples were characterized with an alternating gradient field magnetometer (AGFM) (Princeton Measurements Corporation) and atomic force microscopy (AFM)-MFM (Veeco).

Figure 1 shows representative hysteresis curve for some of the samples with applied field in the parallel direction, that

is, along the film plane. Figure 2 shows the corresponding hysteresis curves for the same samples with applied field along the perpendicular direction. For the as-deposited sample, coercivity is very low ~ 0.39 Oe. Earlier reported value for a sample with comparable thickness, was somewhat higher, ~ 7 Oe.⁵ The corresponding AFM image shows substantial clustering at the surface. The rms surface roughness is quite low ~ 1 nm (Fig. 3) though the sample is not perfectly smooth. Normally increase in surface roughness increases coercivity of thin films.⁹ Therefore, reduced coercivity in our samples is most probably due to demagnetization factor arising from rough boundaries due to multiple interface. For a perfectly smooth interface demagnetization factors at least along the in-plane direction should be zero. However, a rough substrate will induce some interface roughness at the interface of two materials. This has already been observed for other systems for both single layer¹⁰ and multilayer.⁴ In our samples, interface roughness should be at least of the order of a nanometer. Due to existence of multiple rough boundary, the demagnetization factor along in-plane and out of plane direction will be nonzero. This effect will reduce the coercivity substantially. The actual process of interaction between the layers may be very complicated, however, for a single layer, local roughness features induce

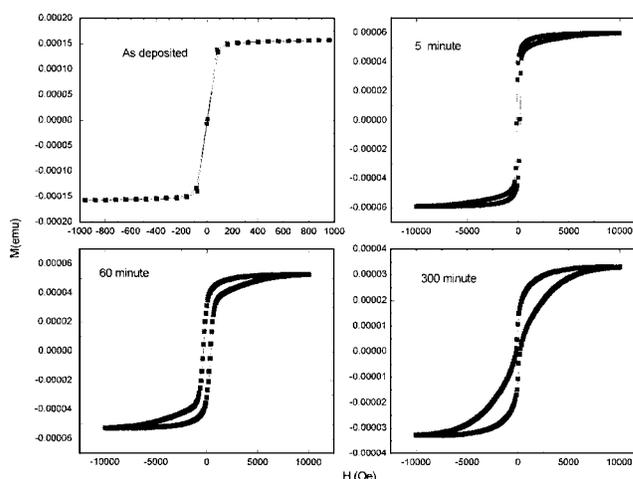


FIG. 1. Hysteresis curve for the samples with applied field along the samples plane.

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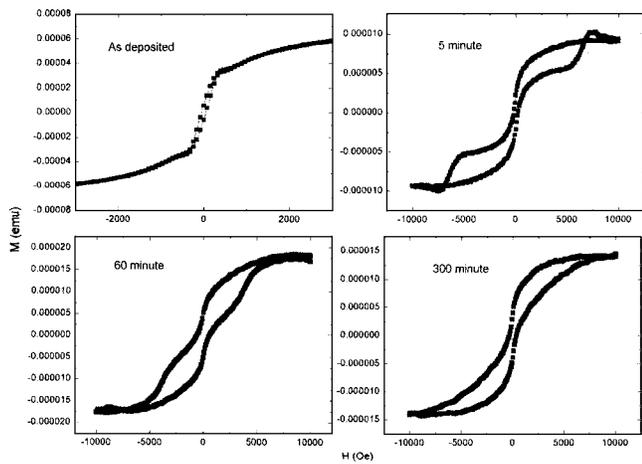


FIG. 2. Hysteresis curve for the samples with applied field perpendicular to film plane.

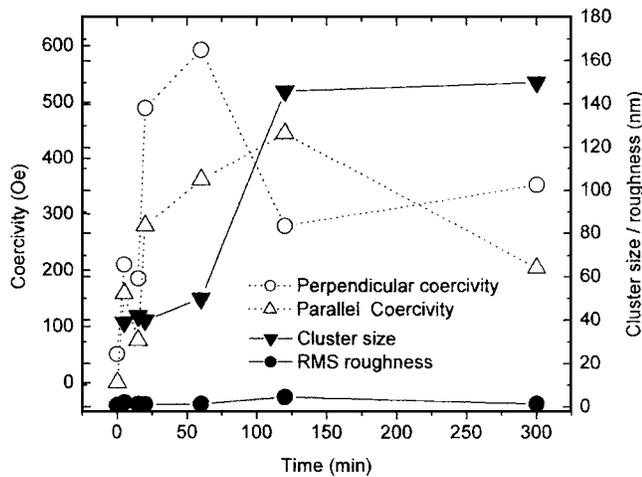


FIG. 3. Coercivity, rms roughness and cluster size as a function of anneal time.

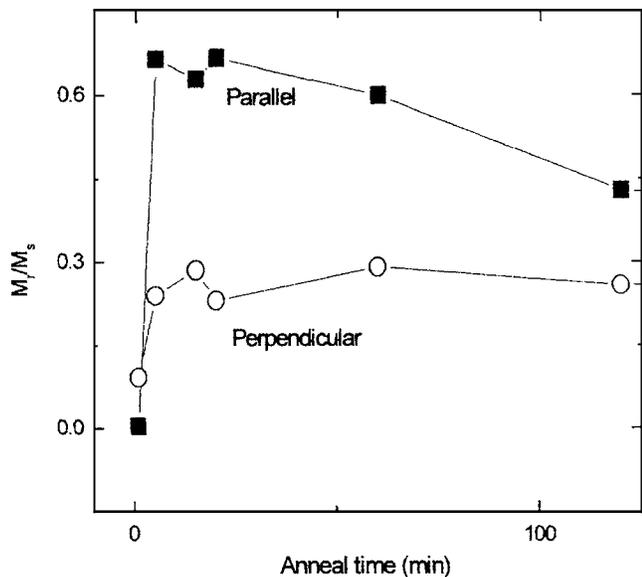


FIG. 4. M_r/M_s as a function of anneal time.

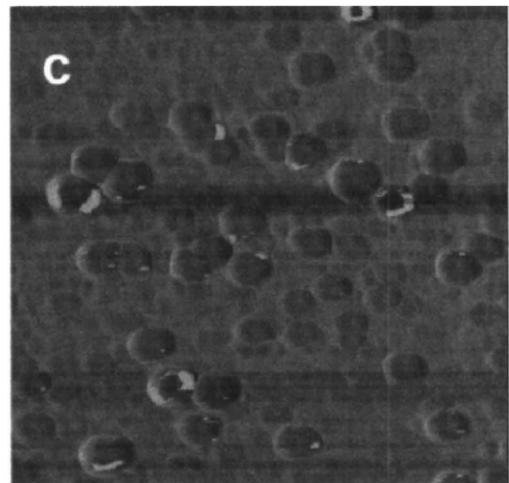
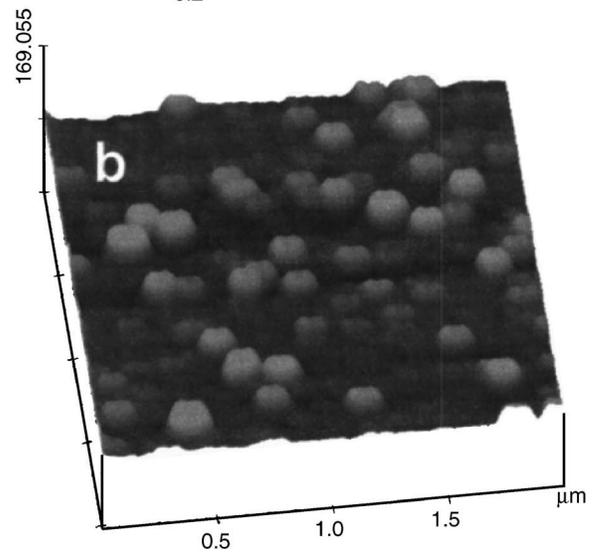
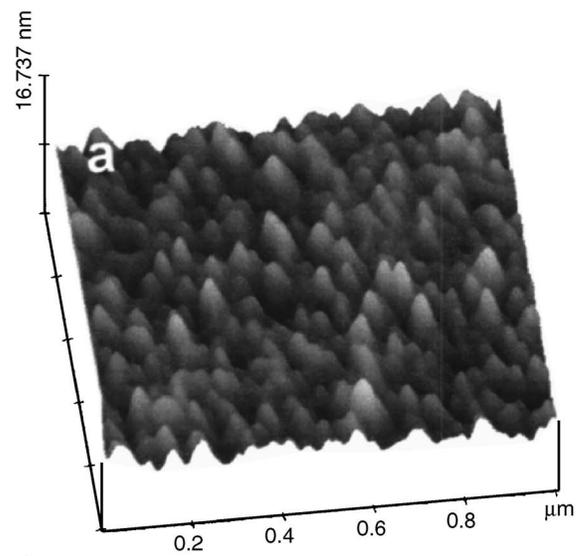


FIG. 5. AFM image for the samples with (a) 1 h ($1 \mu\text{m} \times 1 \mu\text{m}$ scan), (b) 2 h anneal ($2 \mu\text{m} \times 2 \mu\text{m}$ scan) at 350°C , (c) MFM image ($2 \mu\text{m} \times 2 \mu\text{m}$) for the sample annealed for 2 hs at 350°C , showing domain formation.

local in-plane magnetic poles¹¹ resulting in demagnetization. One effect of such local poles will be incoherent rotation of the spins. From Figs. 1, 2, and 4 we find very low value of M_r/M_s for both parallel (0.0042) and perpendicular direction (0.092). This supports the fact that domains/spins do not respond to the applied field uniformly. Also, from Figs. 1 and

2, the saturation field is found to be very small for the as-deposited case. This further supports the fact that local interface roughness is an important factor dictating switching behavior of the films.

With increased annealing time, in the parallel direction, we observe single-phase magnetic behavior in all samples due to strong ferromagnetic exchange interaction between the ferromagnetic layers.^{5,12} In contrast, in the perpendicular direction, the magnetization curves suggest possible existence of multiple phases. With increased anneal time, FeCo solid solution is formed at the interface. This will in turn also increase the rms roughness of the interface leading to increased coercivity. It must also be noted that the presence of several nanoscaled layers can decouple the strong ferromagnetic coupling observed in as-deposited sample. Figures 1, 2, and 3 also show that with increased anneal time, the coercivity jumps very quickly to much higher values. Corresponding magnetic force microscopy images of these samples clearly indicate domain formation (Fig. 5(c)). The largest coercivity is obtained for 60 min of anneal time. For this anneal time, the coercivity is about 600 Oe in the perpendicular direction and about 350 Oe in the parallel direction. With further increase in anneal time, coercivity drops to about 300 Oe in the perpendicular direction and about 200 Oe in the parallel direction. It is also interesting to note that after 300 min of annealing, the hysteresis curves in both directions appear similar (compare Figs. 1 and 2). That is, there is no preferential magnetization direction. As will be discussed later, this is indicative of nanocrystal growth at the interface.

The main experimental results are summarized in Fig. 3. As mentioned earlier, the coercivity for both parallel and perpendicular directions increases with anneal time. While we attempted to anneal our samples at higher annealing temperatures up to 800 °C, we observed no systematic increase in coercivity. One of the reasons is thought to be inhomogeneous clustering. At this particular temperature, 350 °C, we find that grain size systematically increases with anneal time (Fig. 5). Systematic substantial increase in grain size does not occur until 60 min of anneal time (Fig. 3). However, coercivity of the thin film sharply increases even after 5 min of anneal time. Therefore, clustering cannot describe this early stage behavior. Also, we cannot rule out rms roughness (Fig. 3) as a major contributing source for such a high jump in coercivity. Fe and Co forms solid solution over a wide range of temperature.¹³ However, over a wide range of compositional distribution they are soft magnetic materials.¹⁴ In this case, we can easily rule out compositional impact. However, local disorder or defects can influence the coercivity. We attribute this early stage of increase in coercivity to nano-

crystal formation at the interface due to diffusion. While we could not find any study on diffusion activation energy of Fe in Co or vice versa, increase in interface roughness due to FeCo nanocrystal formation at the interface due to interdiffusion similar to the Ni/Al (Ref. 4) system is expected. While systematic study of the nanocrystal is not possible without detailed study of the interface, a similar trend is expected as shown in the model. However, in this case the nanocrystals grow with time. After certain time, they will coalesce and shape anisotropy will no longer be effective. At this stage grain growth and surface roughness will influence the coercivity. The saturation of coercivity can be easily correlated to these two points. At earlier stage the coercivity mainly increases due to nanocrystal growth at the interface, therefore, increasing also the interface roughness. At this point, the film becomes homogeneous and the behavior is dictated by cluster growth and surface roughness.

In conclusion, we have studied the magnetic properties of Fe/Co bilayer. We have shown that the coercivity can be easily tuned to values as large as 600 Oe to obtain hard magnetic material. Our analysis suggests that grain growth induced surface roughness is not enough to explain the observed increase in coercivity with thermal annealing. Instead, the initial rise is attributed to FeCo nanocrystal induced interface roughness.

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