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Tailoring exchange bias with magnetic nanostructures

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Exchange bias in antiferromagnet/ferromagnet bilayers can be tuned by their lateral dimensions when they are below typical magnetic domain sizes. Bilayers of uniform-FeF_2/nanoporous Fe-network have been successfully fabricated by a copolymer nanolithography technique. Feature sizes of about 200 Å have been achieved over a macroscopic 1 cm^2 area. Exchange bias larger than that in uniform FeF_2/Fe film has been observed, with a similar temperature dependence. Magnetic hysteresis loops and anisotropic magnetoresistance both show pronounced asymmetry that vanishes at the FeF_2 Néel temperature. The asymmetry is due to a magnetization rotation process that builds up moments perpendicular to the saturation magnetization directions. The characteristics of the exchange bias originates from the nanostructure of the bilayer.

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Magnetism in nanostructured materials has attracted a great deal of interest in recent years, as the physical dimensions involved are comparable to certain characteristic length scales.[1] In the exchange bias phenomenon in antiferromagnet/ferromagnet (AF/FM) bilayers,[2,3] one characteristic length scale is the size of the magnetic domains in both FM ($\sim 1\ \mu$m) and AF (much smaller), which are believed crucial to the mechanism.[4–10] Different types of domain models, with domain walls perpendicular or parallel to the interface, have been proposed.[4,5,9] Studying exchange bias in nanostructured AF/FM bilayers with nanoscopic feature sizes has the unique advantage of probing the role of domain size and morphologies. It is also technologically important as the exchange bias in AF/FM nanostructures provides an additional, tunable source of anisotropy to stabilize the magnetization, therefore possibly reducing the length scale that determines the superparamagnetic limit in magnetic recording. However, the ease of fabrication and characterization of any nanostructure often scales inversely with the physical dimensions. One convenient method to achieve nanometer-scale features over macroscopic area is by using self-assembled synthetic materials.[11] In this work, we report the successful fabrication of uniform-FeF_2/nanoporous Fe-network with feature size of about 200 Å, over 1 cm^2 area, using a diblock copolymer nanolithography technique.[11,12] We have studied exchange bias in this system by both magnetometry and anisotropic magnetoresistance (AMR) measurements. The exchange field in networked bilayers exhibits similar temperature dependence and a larger magnitude than in uniform FeF_2/Fe bilayers. Moreover, the magnetic hysteresis loops exhibit a pronounced asymmetry that vanishes at the FeF_2 Néel temperature T_N. AMR measurements further confirm that the asymmetry is due to a coherent magnetization rotation process that exists primarily in the increasing-field branch of the hysteresis loop.

Uniform thin films of $FeF_2(200\,\text{Å})/Fe(150\,\text{Å})$ were deposited by *e*-beam evaporation onto 1×1 cm^2 single crystal MgO (100) substrates, capped with a 40 Å Al layer. The substrate was first annealed at 450 °C for 0.5 h, followed by the deposition of FeF_2 at 200 °C, and Fe and Al at 150 °C. The deposition rates of all layers were 1 Å/s. Further details of the growth parameters have been published elsewhere.[13] Structural characterizations of the thin films have been performed by x-ray diffraction and atomic force microscopy (AFM). The AF FeF_2 ($T_N \sim 80$ K) grows as twinned quasiepitaxial layer along the compensated (110) surface (twin domain size $\sim$100 Å),[14] while the Fe layer is polycrystalline. The root-mean-square roughness determined by AFM is about 16 Å.

Diblock copolymers of polystyrene and polymethylmethacrylate, denoted PS-*b*-PMMA, were used as templates on top of the FeF_2/Fe/Al structure. A 50 Å thick neutral brush layer of a hydroxy-terminated random copolymer PS-*r*-PMMA (volume ratio 58:42) was anchored to the native aluminum oxide on top of the Al layer in a manner described previously.[15] This layer eliminated any preferential orientation of the copolymer caused by the working surface. A 300 Å thick layer of an asymmetric PS-*b*-PMMA volume ratio 70:30) was then spin-coated onto the neutral brush layer and annealed in vacuum at 150–170 °C for 24–78 h. The two blocks of the copolymer are chemically distinct and spontaneously self-assemble into microdomains upon annealing.[11,12] At this composition of the copolymer, the minor component, PMMA, forms an array of cylinders.[16] Under UV exposure and sonication in glacial acetic acid and deionized water, the PMMA blocks were degraded and removed, while the PS blocks were crosslinked and immobilized.[17] This produces a nanoporous template that can be used to transfer the pattern into the FeF_2/Fe film by ion milling. This process has been analyzed by ellipsometry and atomic force microscopy (AFM). Figure 1 shows a top view AFM phase image of a resultant Fe network. The dark and bright areas are, respectively, the pores and the cross-linked network. The transfer of the template to the film was excellent. The characteristic dimensions of the network, the pore size and pore separation, are both about 200 Å. By stopping ion milling at the interface of FeF_2/Fe, we have fabricated a network of Fe on top of a uniform FeF_2 layer. In the follow-

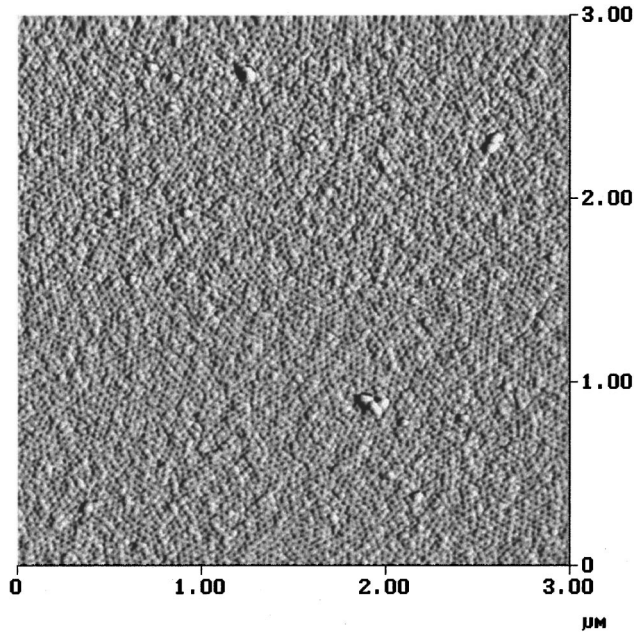

FIG. 1. Top view AFM phase image of a porous Fe(150 Å)/Al(40 Å) network. The dark and bright areas are pores and networks, respectively.

ing, we will compare network structures with uniform films.

Because of the macroscopic area of the Fe networks, magnetic measurements can be easily performed by a conventional superconducting quantum interference device. In simpler cases of a single 150 Å thick Fe network and a uniform Fe film, the hysteresis loops are qualitatively the same in the temperature range studied (10–300 K). However, the coercivity H_C in the network is always slightly smaller than that in the uniform film (e.g., 28 Oe vs 44 Oe at 10 K). This is due to the reduced anisotropy in the network geometry as the physical dimensions approach zero. Note that in the present networks, H_C is smaller than in networks made on porous Al_2O_3 membranes, where the different morphology and pinning from surface roughness played important roles.[18]

The behaviors of FeF_2/Fe bilayers, however, are drastically different in the network and uniform film geometries. Figures 2(a) and 2(b) show the hysteresis loops of a uniform film of FeF_2/Fe and a uniform-FeF_2/Fe-network taken at 10 K, respectively, field-cooled in 5 kOe from room temperature. The uniform FeF_2/Fe film shows a symmetric hysteresis loop, shifted by an exchange field of $H_E = -200$ Oe. In contrast, in uniform-FeF_2/Fe-network, the loop has a pronounced asymmetry, with sharper switching and a larger loop shift. The switching from positive to negative saturation is rather abrupt [leg I in Fig. 2(b)]. Upon returning, the magnetization reversal first gradually proceeds to about -160 Oe (leg II), at which point it quickly switches to positive saturation (leg III). The exchange field is about -260 Oe. The temperature dependence of exchange field is shown in Fig. 3. In the uniform FeF_2/Fe, the exchange field shows a plateau before vanishing at the FeF_2 T_N of 80 K. In the uniform-FeF_2/Fe-network, the exchange field is larger and follows a similar temperature dependence.

To examine more closely the unusual switching characteristics, we define the switching width as the field change needed to switch between $\pm 70\%$ saturation magnetization M_S. Figure 4 shows the temperature dependence of the switching width in the network geometry. In the decreasing-field branch, the width is essentially flat, within a narrow range of 50–80 Oe. In the increasing-field branch, the width

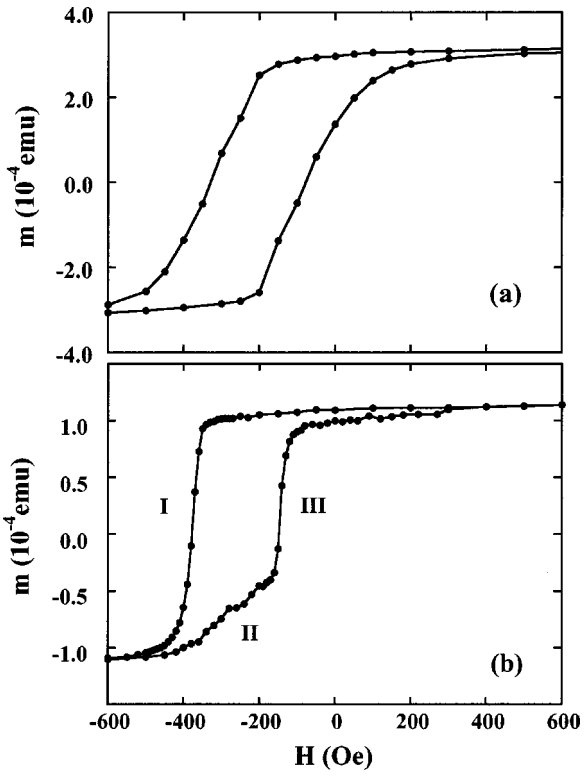

FIG. 2. Magnetic hysteresis loops of (a) a uniform FeF_2(200 Å)/Fe(150 Å)/Al(40 Å) film, and (b) a uniform FeF_2 (200 Å)/networked-Fe(150 Å)/Al(40 Å) at 10 K, after field cooling in 5 kOe from 300 K. Different parts of the reversal process are labeled I, II, and III.

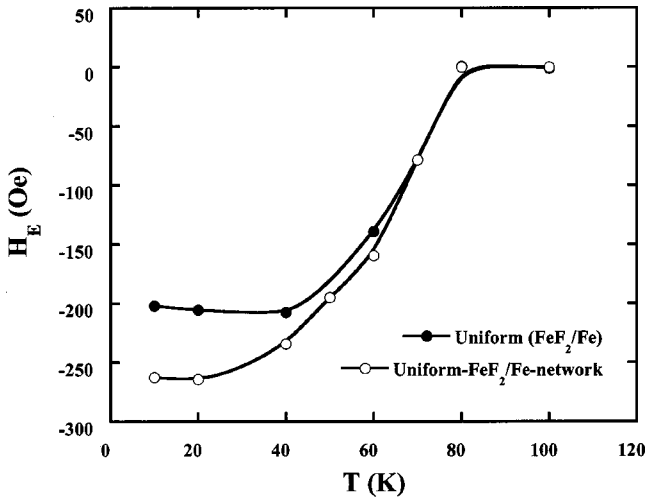

FIG. 3. Temperature dependence of exchange field H_E in uniform FeF_2/Fe film (solid symbols) and uniform-FeF_2/Fe network (open symbols), after field cooling in 5 kOe from 300 K.

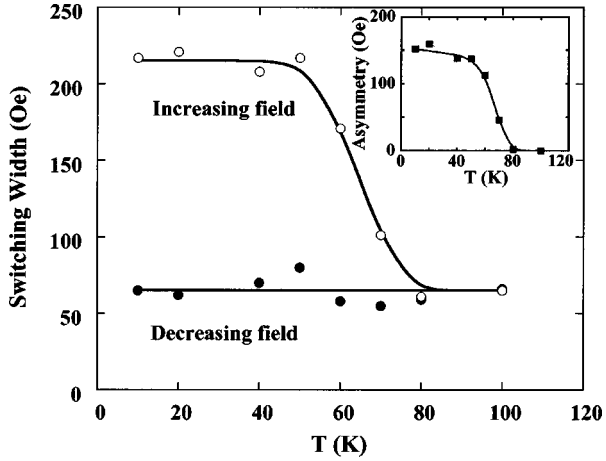

FIG. 4. Temperature dependence of magnetization switching width between ±70% M_S for uniform-FeF_2/Fe-network. The inset shows the difference between the two branches, or the loop asymmetry. Solid lines are guides to the eye.

stays around 210 Oe until 50 K, beyond which it precipitously drops and eventually overlaps with the decreasing-field branch at 80 K and above. The difference between the two switching width, or the loop asymmetry, is shown in Fig. 4 inset as a function of temperature. Interestingly, it bears remarkable resemblance to that of H_E (inverted). In comparison, in uniform FeF_2/Fe film, the switching width is about 300 Oe in both branches at 10–100 K and the asymmetry is no more than 60 Oe. This indicates that the pronounced asymmetry observed in the networks is not only due to the exchange bias between FeF_2 and Fe, but also intrinsic to the network nanostructure. Likely different magnetization reversal mechanisms are involved in decreasing- and increasing-field branches of the loop.

To determine the reversal mechanisms of the network sample at the two branches of the loop, we have performed the following reversibility tests from saturation. For the decreasing/increasing field branch, the sample was brought from positive/negative saturation to every field value along the branch, then back to the exchange field to measure the remanence. It is observed that only the initial gradual reversal process from negative saturation is reversible, while the sharp switching processes are irreversible. This is also confirmed by minor loop measurements along different locations of the major loop. These results indicate that the reversal from negative saturation (leg II) initiates by coherent rotation. Along legs I & III, the reversal likely involves both domain-wall motion and incoherent rotation processes that cannot be distinguished by the reversibility test.

We have also measured AMR of the uniform-FeF_2/Fe network, as it is sensitive to the relative orientation of magnetization and the sensing current direction. AMR has been shown to be effective in revealing magnetization reversal mechanisms in exchange biased systems.[19] Figure 5 shows the transverse AMR results measured at 10 and 100 K, using the same field cooling procedure described earlier. At 100 K, above T_N of FeF_2, the AMR shows the expected symmetric double peak pattern, similar to that of a single Fe layer [Fig. 5(b)]. Strikingly, below T_N, the pattern is not only shifted,

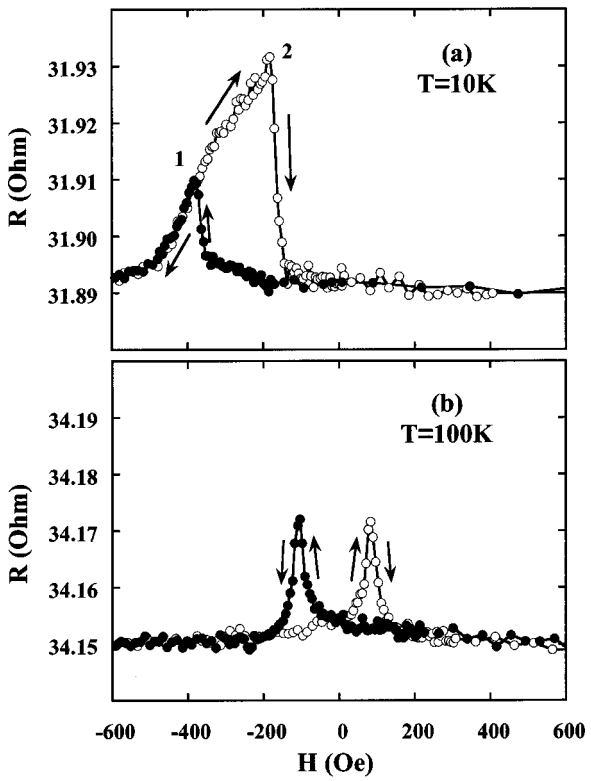

FIG. 5. Transverse anisotropic magnetoresistance of uniform-FeF_2/Fe-network at (a) 10 K, and (b) 100 K, after field cooling in 5 kOe from 300 K. Peaks in decreasing-field (solid symbols) and increasing-field branches (open symbols) are denoted as 1 and 2, respectively.

but also the double peak feature has evolved into a hysteresis loop [Fig. 5(a)]! A clear correlation can be established between the magnetic and AMR hysteresis loops. The abrupt switching in the decreasing-field branch of the *M-H* loop gives rise to the small peak 1 in AMR [Fig. 5(a)]. The gradual magnetization reversal process from negative saturation builds up AMR, before collapsing at positive saturation and forming a large peak 2 [Fig. 5(a)]. In AMR measurements, the height of the peak correlates to the amount of the magnetization that has turned orthogonal from saturation. Therefore, the big peak 2 in AMR reveals the presence of more perpendicularly oriented FM magnetization to the field direction. This can only be realized by magnetization rotation, consistent with the conclusion drawn earlier.

The interesting loop asymmetry and the larger H_E observed in the network sample are intimately related to its nanostructure. We should note that the ion milling process is not completely uniform over the sample area. With porous networks and areas of continuous but thinned film of Fe on top of the FeF_2, the magnetic hysteresis loop is the superposition of the two contributions. Recently, we have demonstrated from polarized neutron reflectivity that in uniform films of FeF_2/Fe, the reversal process in seemingly symmetric loop is predominantly by rotation and domain-wall motion in decreasing- and increasing-field branches of the loop, respectively.[20] In the present sample, in areas that are continuous but thinned Fe, those processes still exist along legs

I & III in Fig. 2(b). The sharper switching is due to the reduced anisotropy in the network sample. Additionally, in areas that are purely networks, the small dimensions impose restrains on domain-wall formation and motion. Thus magnetization reversal is achieved primarily by rotation, leading to a more gradual switching. The resultant hysteresis loop shows pronounced asymmetry. Details of the reversal processes are worth further studying, as they depend sensitively on the network morphology resulted from the ion milling process.

In previous studies of micron-sized AF/FM structures, there have been conflicting results on the magnitude of the exchange field.[21,22] Recently, Li and Zhang have proposed a random field model that yields quantitative agreements with experiments.[10] Their model emphasizes the role of FM domain size, determined by the competition between FM-FM exchange and the random field due to the interfacial FM-AF interactions. When the FM-FM interaction is strong, as favored by smaller AF grain size or thicker FM layer, the FM forms larger domains ($\gg$AF grain size) and the net random field averages out to be small, thus smaller H_E and H_C. It is clear from this model that limiting the lateral dimension of the AF/FM bilayer alone is not expected to alter exchange bias significantly until the dimensions are below the FM domain (typically on the micron scale) or AF grain size (usually much smaller) in uniform films. In the present network geometry,[23] the nanoporous Fe network effectively reduces the FM-FM interactions, thus favoring the formation of smaller FM domains, larger net random fields, and larger exchange field. A larger exchange field in nanostructured AF/FM bilayers has also been observed recently in CoO/NiFe system.[24] Ultimately, as the replication of the porous network goes deeper into the bilayer and the dimensions of the AF/FM entities become smaller and smaller, the exchange field is limited by superparamagnetism.

In summary, we have demonstrated that exchange bias can be tailored by magnetic nanostructures where the feature sizes are below typical magnetic domain sizes. We show that in uniform-FeF_2/nanoporous Fe-network bilayers, the exchange bias is larger than that in a uniform FeF_2/Fe film, but with a qualitatively similar temperature dependence. The magnetic hysteresis loops and AMR both show a pronounced asymmetry that vanishes at the FeF_2 T_N. The asymmetry is due to a magnetization rotation process that builds up magnetic moments perpendicular to the saturation directions. The unusual characteristics of the exchange bias are intrinsic to the nanostructures.

Our results not only demonstrate a feasible and convenient method to tune exchange bias and probe the role of domain size, but also show a different type of medium to study new physics in restricted dimensional systems. The fabrication technique is also relevant to such applications as in patterned recording media.

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