Exchange Bias and Asymmetric Reversal in Nanostructured Dot Arrays

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The size dependence of exchange bias field H_E and coercivity H_c was studied by measuring exchange biased Fe-FeF_2 dot arrays in comparison with an unstructured exchange biased Fe-FeF_2 bilayer. The domain sizes in the ferromagnet (FM) and the antiferromagnet (AFM) play an important role for exchange bias (EB), and thus interesting phenomena may be expected when the size of an EB system becomes comparable to these sizes. We observe drastic changes of H_E and H_c in nanostructured Fe-FeF_2, which are unexpected because they appear even at a structure size which is too large for matching with AFM or FM domain size to play a role. We propose that under certain conditions the hysteresis loop is affected differently in the two branches of the reversal by shape anisotropy due to patterning. This is possible because the EB induces a reversal asymmetry already in the unpatterned bilayer system.

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Exchange bias (EB), a shift in the hysteresis loop away from the normal $H = 0$ field [1], is intensely investigated in search of the controlling mechanism and because of its importance in a variety of applications [2]. In particular, EB is considered to stabilize magnetism in small magnetic structures, which otherwise would be superparamagnetic and unsuitable for, e.g., magnetic storage. In antiferromagnetic (AFM) and ferromagnetic (FM) bilayers the interfacial coupling is believed to control the magnitude of the loop shift and domains in the AFM and/or the FM play a crucial role [3]. Thus drastic changes of magnetic properties are expected when the size of an EB system becomes comparable to these sizes. A less explored possibility is that in EB systems nanostructuring produces major changes in the demagnetizing energy which may affect the magnetic properties in a fundamental way. The reversal mechanism in magnetic materials originates from a balance between different energies: Zeeman, demagnetizing, anisotropy in the FM and AFM, and interfacial exchange. In contrast to the pure FM layer, those energies are not symmetric in the two branches of the hysteresis loop, but asymmetric because of the unidirectional anisotropy induced by EB. This can produce not only shifted but also asymmetrically shaped hysteresis loops. Patterning a FM layer changes the demagnetizing energy drastically which often leads to smaller FM domains and increases in coercivity. This may affect the reversal mechanism in FM-AFM bilayers, perhaps influencing differently the two reversal branches and changing the magnitude of the EB. The possibility of such effects has not been emphasized in recent experiments on patterned EB bilayers [4–6]. There, the interpretation of size dependent exchange bias field H_E is often focused on modifications of the exchange interaction at the AFM-FM interface and/or matching of length scales. To investigate this possibility we have embarked on a systematic study of nanostructuring exchange biased Fe-FeF_2 bilayers.

Fe-FeF_2 bilayers are prepared by *e*-beam evaporation on MgO(100) substrates. From x-ray diffraction we determine that the 23.8 nm thick FeF_2 layer, grown at 0.2 nm/s and 200 °C, is quasiepitaxial with twinned in-plane structure. Because of the large anisotropy and based on the bulk spin structure, the $FeF_2(110)$ surface is expected to be compensated. The 12 nm thick Fe layer, grown at 0.2 nm/s and 150 °C, is polycrystalline [7]. The sample is capped with a 4.8 nm thick layer of Al to prevent oxidation. Different square arrays of circular Fe dots with diameter of 600 ± 10 nm and 100 ± 10 nm were prepared by Ar^+-ion milling through a resist mask produced by *e*-beam lithography. The center-to-center distance of the dots was twice their diameter. On the same sample, an unpatterned area was kept covered with resist during ion milling to provide a continuous film control sample. To minimize possible differences due to processing, the patterned and unpatterned area were subject to the same processing steps together. Because of the small *overall* area of electron beam patterned samples, the magnetic properties were measured by magneto-optical Kerr effect (MOKE) using an optical magnet cryostat. The MOKE signal was measured after ramping the field with 1 Oe/s to a specific value, waiting for 5 s and then taking the average of five consecutive values measured within 10 s. The HeNe laser beam was focused down to a 50 μm diameter to measure the unpatterned or patterned area (size: 80 μm × 80 μm) on the same film individually. The samples were field cooled from 150 to 10 K through the Néel temperature $T_N = 78.4$ K of FeF_2. The cooling field $H_{fc} = 2$ kOe, oriented parallel to the film surface along the [010] direction of the MgO substrate, is large enough to saturate the FM layer and the FM dots. Using *p*-polarized light (inci-

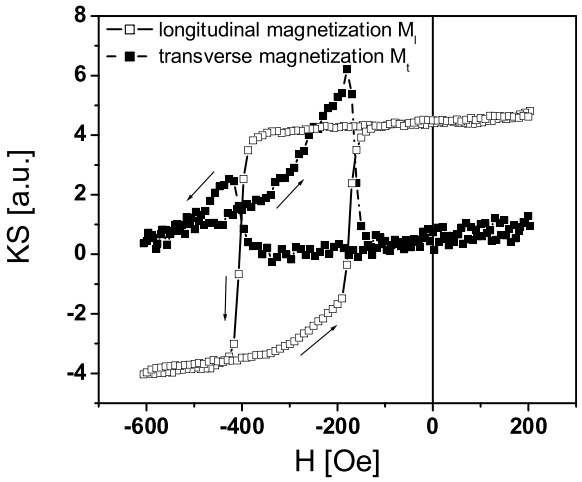

FIG. 1. MOKE signal of M_l (open symbols) and M_t (closed symbols) of the unpatterned area of the film at 10 K after field cooling in 2 kOe.

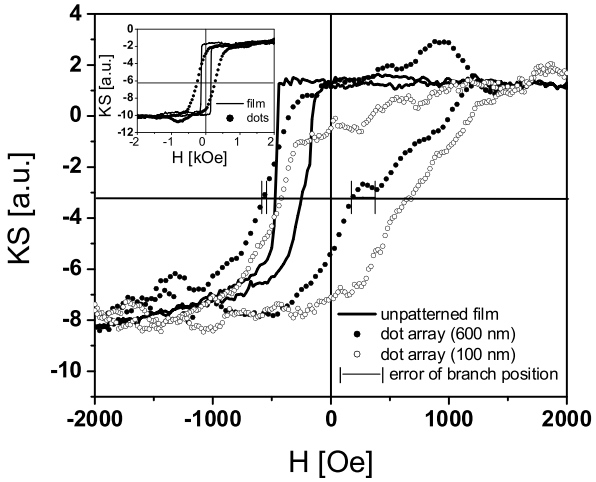

FIG. 2. MOKE signal (KS) of M_l vs H of unpatterned Fe-FeF_2 film and different Fe dot arrays on FeF_2 at 10 K (below T_N). The inset shows the signal of unpatterned Fe-FeF_2 film and the 600 nm Fe dots on FeF_2 at 90 K (above T_N).

dent at 58° with respect to the normal), the longitudinal magnetization component M_l (parallel to the field) was measured by detecting the Kerr rotation of the reflected light. The transverse magnetization component M_t was measured by rotating the polarization of the incident beam by 45° with respect to the commonly used p-polarization [8].

Figure 1 shows the MOKE signal of M_l and M_t at 10 K of an unpatterned region on the Fe-FeF_2 bilayer. Typically for many EB bilayer systems [4,7,9–12], the hysteresis loop of M_l (open symbols) is asymmetric, i.e., one branch (the increasing branch in Fig. 1) is more rounded than the other. This asymmetry is also reflected in the corresponding M_t which shows a larger signal in the increasing branch (IB) than the decreasing branch (DB). For unpatterned FeF_2-Fe bilayers, this asymmetry was attributed to the competition between the different energies giving rise to coherent rotation and/or domain wall motion [10]. By Kerr microscopy on NiO-Co bilayers, smaller domains were observed at the reversal with the more rounded M_l branch and larger M_t [12]. Accordingly, the larger M_t during the reversal was attributed to reversal by rotation and/or a much larger number of domain walls, in which the spins are oriented perpendicular to the external field. This implies that the spin structures in the two branches of the unpatterned film in Fig. 1 are different.

The same sample patterned to dot arrays has hysteresis loops with clearly larger coercive field H_c and smaller EB field H_E (Fig. 2), even if the larger errors of the exact branch positions are considered. The curves of the dots are not as smooth as for the unpatterned area, probably because of a larger linear background, which was subtracted. As expected, with increasing temperature, H_E for both the unpatterned and patterned areas becomes smaller and vanishes at T_N (inset of Fig. 2). To investigate the influence of patterning on the reversal in detail, we compared the positions of the DB and IB of unpatterned and patterned film area with 600 nm dots at different temperatures and with the external field applied either parallel or at 40° (''tilted'') with respect to the sample surface (Fig. 3). As branch position, the field value was taken at which the magnetization is zero. The error bars reflect the scatter in the data (cf., Fig. 2). At low temperature ($\leq$ 30 K) a strong shift of the IB of the hysteresis loop is caused by the patterning, while the change on the DB is much less pronounced. To compare the in-plane component of the magnetization of the two series of experiments, the mea-

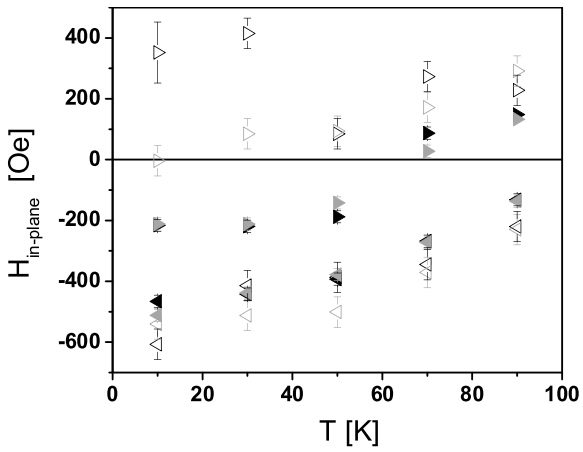

FIG. 3. In-plane magnetic field component $H_{\text{in-plane}}$ for the positions of the DB (triangles pointing to the left) and IB (triangles pointing to the right), defined by $H(M_l = 0)$, for unpatterned (solid symbols) and patterned (open symbols) sample. For the black symbols the field was parallel to the sample surface. For the gray symbols the field was tilted by 40° in respect to the surface after field cooling.

sured value for the tilted sample was multiplied by $\cos(40°)$ as shown in Fig. 3.

Since domains may play a central role, the following considerations are important. If the AFM domain sizes are much smaller than the size of the FM dots, any explanation [6] where the AFM domain size is required to be comparable to the FM dots should not play a major role in the effects observed here. The AFM domain size in such FeF_2/Fe thin films is difficult to measure because of their very small size. An indication of the domain size can be inferred from the thickness dependence of the EB. FeF_2-Fe bilayers prepared under similar conditions show an increase of H_E with increasing AFM film thickness up to about 20 nm and then a leveling off [13]. The constant H_E at larger thicknesses is an indication that the EB and the size of AFM domains are mostly determined by the size of twins in the AFM, whose size is comparable to the AFM film thickness (24 nm) [14]. Alternatively, the lateral size of the AFM domain can be estimated to about twice the AFM film thickness [15]. The width L of the AFM domain wall can be estimated with $L = \pi(A_2/K)^{1/2} = 0.6$ nm, where $K = 5.49 \times 10^6$ J/m^3 is the uniaxial magnetic anisotropy along the c axis of FeF_2 and $A_2 = 2.19 \times 10^{-13}$ J/m is the next nearest neighbor exchange stiffness constant [16]. The difference between the AFM domain size and a dot diameter of 100 nm is still small enough that in this case the reduced H_E might be explained by effects appearing if AFM domains sizes are comparable with the structure size [6]. The situation is different for the larger dots with diameter of 600 nm. There, both the AFM domain size and the wall width are considerably smaller than the dot size. In the following, we will concentrate on possible origins of the reduced H_E for these comparably large dots, which is unexpected considering interpretations discussed so far for patterned EB systems.

Since a training effect is not observed in the FeF_2-Fe system, it is generally assumed that for FeF_2 the AFM spin distribution at the AFM-FM interface is determined mostly by the conditions during field cooling and freezes below T_N to a stable configuration. In contrast, FM domains are not frozen, but change their average size from a large size at saturation to a smaller size at magnetization reversal. The latter requires the FM structure to be large enough to sustain multidomain states. We can assume this for our polycrystalline Fe dots (diameter = 600 nm), because even for epitaxial Fe dots with smaller diameters and the same thickness, multidomain states were observed [17]. Since H_c in our sample is mainly caused by pinning of FM domain walls at defects [2], it increases with higher domain wall density. Losses in the AFM layer contribute to H_c only close to T_N, leading to a maximum in the otherwise constant H_c in Fig. 3 of the unpatterned film.

The domain wall density depends on the domain size. Since the dots are large enough to sustain multidomain states, additional shape anisotropy, induced by patterning, can be reduced by forming smaller domains in the FM and H_c increases by $\Delta H_c = 1/2(\Delta H_{cD} + \Delta H_{cI})$, where ΔH_{cD} and ΔH_{cI} are the shifts of the DB and IB, respectively, towards larger coercivity. Since the domain structures at the two magnetization reversals are different in the unpatterned film, patterning might change the domain structure in the two branches differently. As a consequence, the shifts of the branches will be different, i.e., $\Delta H_{cD} \neq \Delta H_{cI}$. This changes H_E by $\Delta H_E = 1/2(\Delta H_{cD} - \Delta H_{cI})$ without need of a modified interfacial exchange interaction.

To investigate this issue, we compare the data of Fig. 3 in more detail. The first set of measurements was performed with the external field parallel to the sample surface, and the second at 40° with respect to the sample surface. However, in both cases the sample was field cooled in a field parallel to the sample surface. Since the domain structure of FeF_2 freezes during field cooling into a stable configuration, the AFM domain structure and the interfacial exchange interaction should be comparable for the untilted and tilted case. This is supported by the fact that the positions of the branches of the *untilted*, unpatterned film agree with rescaled positions of the *tilted*, unpatterned film over the whole temperature range (Fig. 3). The situation is significantly different for the patterned sample at $T = 10$ and 30 K, where patterning changes the in-plane component of H_E differently for the tilted and untilted case. The interfacial exchange interaction of the dot array should not be altered by tilting either, because of the much larger size of the Fe dots compared to the AFM domain size, and therefore properties of the interfacial exchange interaction comparable to the one of the unpatterned sample. For unchanged interfacial exchange interaction the different in-plane component of H_E can only be due to the FM spin structure, which is not frozen and may be influenced by tilting. Obviously H_c, mainly determined by the pinning of FM domains, changes in such a way that $\Delta H_{cD} - \Delta H_{cI} = 2\Delta H_E$ is different for the untilted and tilted sample. Because of the small AFM domain size, a change of interfacial exchange interaction by patterning is not expected in our size regime. Moreover, it is unlikely that the changes of H_c and H_E after patterning for $T \leq 30$ K stem mainly from averaging over different properties of the individual dots, because then we would expect similar large differences at all temperatures, in particular, also above T_N. Therefore, such asymmetric shifts of the branches caused by changed FM domain structure should be the main contribution to the decreased H_E in our patterned sample.

We notice that especially at $T \leq 30$ K the differences between patterned and unpatterned film in H_c and H_E are caused by a strong shift of the IB of the hysteresis loop, while the change on the DB is much less pronounced. This may be related to the different field values at the two reversals. The relative contribution of the Zeeman energy

to the total energy of the FM layer increases with the external field H_{ext} and will be different in the two branches. Therefore minimization of the Zeeman energy determines the spin structure at the reversal of the DB more strongly than the one of the IB. Since the Zeeman energy favors parallel spin orientation, the smaller M_t in the DB (compared to the IB) is an indication that this situation is present in the unpatterned area of our sample. The demagnetization field H_d due to patterning, changes the total energy in the FM layer, which is equivalent to a change of the Zeeman energy with an external field reduced by $1/2H_d$. This will effect the spin configuration only slightly if Zeeman and demagnetization energies together still dominate the total energy of the system. In order to estimate the relevance of this effect we calculate an approximate upper limit for the demagnetization field H_d. First, we consider only a single dot without interdot coupling with its neighbors. This approach seems reasonable according to measurements on polycrystalline iron dot arrays on Si with comparable geometries [18] from which interdot coupling can be neglected. The upper limit of the demagnetization field can be estimated to $H_d = -NM_s = -245 \pm 14$ Oe, with an approximated demagnetization factor $N = 0.01571 \pm 0.0002$ for a single iron dot and a saturation magnetization $M_s = 1.24 \times 10^6$ A/m, as obtained by SQUID magnetometry. The maximum field which can be produced by the next nearest neighbors is 19 Oe and much smaller than H_d, which supports neglecting interdot interaction. The relatively weak shift of the DB of the patterned area implies that the FM spin structure is dominated by the sum of Zeeman and demagnetization energy, since $1/2H_d = -122$ Oe is definitely smaller than the field value at the decreasing reversal of the unpatterned area. At the IB of the unpatterned area the estimated $1/2H_d$ is already comparable to the external field. This increases the influence of anisotropy and exchange energy in minimizing the total energy. The formation of a large number of domain walls becomes more favorable, causing a stronger average pinning with a distinct shift of the IB towards larger H_c. Consequently, the decreased H_E in the patterned sample is significantly determined by the influence of demagnetization on the FM spin structure.

Micromagnetic simulations of patterned NiFe-CoO bilayers point towards a similar direction [19]. However, in contrast to our experiments the simulated H_E of small elements is larger than those of the unpatterned films. This discrepancy is not solved yet, but might be due to different material parameters and pattern sizes in the simulation and our experiment. Also an unconsidered depth dependence in the FM spin structure might have an influence, such as a spirallike, incomplete domain wall parallel in the FM layer [20] or, at even smaller FM dots, a vortex state with perpendicular magnetization components.

In summary, Fe nanodots in contact with an FeF_2 antiferromagnetic substrate exhibit decreased H_E and increased H_c compared with continuous films, although the diameter of Fe dots is much larger compared to the AFM domain size, and therefore a change of AFM-FM exchange interaction is not expected. This effect originates in patterned EB systems when the changes in the demagnetizing energy compared to other important energies in the problem (Zeeman, exchange, and anisotropy) become a dominant factor.

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