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Magnetization Reversal of Strongly Exchange-Coupled Double Nanolayers for Spintronic Devices

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Abstract

Contemporary spintronics devices, notably GMR and TMR-sensors with high linear ranges, stand to benefit from large bias fields in excess of 1 T that have been observed in exchange-coupled double layers using rare earth-transition metal ferrimagnets as pinning layers. However, before exploiting the strong coupling the reversal process must be understood at the scales of the smallest devices, where it would be effective. Little is known about these processes to-date, owing to the technical difficulty of imaging magnetic structures smaller than 20 nm in fields of several Tesla. Here we image the nanoscale magnetic structures of a double layer

comprising a polycrystalline Co/Pt-multilayer (Co/Pt) which is strongly exchange-coupled to an amorphous and magnetically-hard ferrimagnetic TbFe alloy layer. High resolution magnetic force microscopy provides snapshots of the magnetic structures along various applied fields between 0 and 7 T as the magnetization reversal takes place. In contrast to a magnetization reversal process governed by nucleation followed domain wall motion observed in weakly exchange-coupled systems, a complex nucleation-dominated magnetization process with three distinct reversal stages is found. Micromagnetic modeling reveals that the grain-to-grain variation of the Co/Pt anisotropy and the strength of the interfacial exchange-coupling affect this reversal process. We show that we can control the coupling and thus the reversal process by using spacers at the ferrimagnet-ferromagnet interface, a useful option when designing spintronics devices.

Keywords

magnetization reversal, exchange spring systems, spintronics, quantitative MFM, micromagnetic modelling

Introduction

Stabilizing a specific direction of a ferromagnet's magnetization is a key capability in many spintronics devices. Presently it is provided by using the exchange-bias effect.¹ This technique enables applications such as spin valves,^{2,3} magnetic tunnel junction devices,^{4,5} and it has also been considered in heat-assisted magnetic recording for raising the bit areal density in bit-patterned media to several tens of Tb/in².^{6,7} For the latter example in particular,^{8–10} but likewise in general spintronics applications, exchange-coupled double layer (ECDL) structures using rare-earth-based ferrimagnets (FI) might provide useful design flexibility.

What sets apart FI-based ECDL structures from conventional antiferromagnet (AF)-based exchange-bias systems, is that coupling frustration can largely be avoided because the antiparallel-aligned

magnetic moment groups in the ferrimagnet are borne by different atomic species.¹¹ One of the consequences is that during the reversal of the ferromagnet (FM) at low temperatures, high-energy in-plane domain walls can form.^{12–16}

It is also known from conventional exchange-bias systems, that the inhomogeneity of the coupling between the AF and the FM strongly influences the domain wall motion in the latter.¹⁷ In FI-based strongly coupled ECDL structures, the reversal could be very different from conventional systems, and it is important to assess it for future devices. A difficulty in this regard is that the structural features of the magnetization are likely very small, account taken of the large magnetic anisotropy of TbFe,^{18–22} so imaging the ECDL during reversal requires large fields and high resolution. This is a difficulty we overcome in the present work, using high-resolution magnetic force microscopy (MFM) in fields up to 7 T to image an area of a Co/Pt-multilayer/TbFe ECDL during the magnetization reversal at 10.5 K.

Experimental Results

For our study we utilize magnetron sputtered single layer (sample SL) and exchange-coupled double layer (sample ECDL) samples. Sample SL consists of a 20 ± 1 nm thick ferrimagnetic layer of an amorphous $\text{Tb}_{26.5 \pm 1.5} \text{Fe}_{73.5 \pm 1.5}$ alloy on a Pt seed deposited on thermally oxidized Si substrates. The composition (atomic ratio) denoted by the subscript numbers was obtained from Rutherford backscattering spectrometry (RBS). For oxidation protection the ferrimagnetic layer is covered by 8.5 nm of Pt. Sample ECDL consists of the same ferrimagnetic film onto which we deposit a perpendicular-anisotropy $(\text{Co}_{0.4 \text{ nm}}/\text{Pt}_{0.7 \text{ nm}})_{\times 5}$ multilayer. The thickness of the Pt oxidation protection layer was reduced to 3 nm in sample ECDL to obtain the same 8.5 nm distance between the top surface and the top of the rare-earth layer. This is convenient when comparing the MFM data for these two samples (schematics of these are displayed in section 1 of the Supporting Information). Vibrating sample magnetometry (VSM) measurements are performed after zero-field cooling the samples from room temperature to 10.5 K. The magnetic imaging is performed with

1
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3 a home-built low temperature UHV MFM²³ at 10.5 K and in magnetic fields of up to 7 T after
4 zero-field cooling the samples in an as-grown state. We use high-aspect ratio silicon cantilevers
5 (made sensitive to magnetic stray fields of the samples by a few nanometers of Co) with a res-
6 onance frequency of about 50 kHz and a nominal stiffness of 0.7 N/m. Additional details on the
7 MFM data acquisition and processing, particularly how the magnetic, topographical and electro-
8 static contributions to the measured frequency shift contrast are disentangled, are given in section 2
9 of the Supporting Information.
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13 Sample SL exhibits a strong perpendicular magnetic anisotropy that leads to the large coercivity
14 displayed in the hysteresis loops of Figure 1A. The hysteresis loop of sample ECDL displays
15 the Co/Pt reversal starting at the exchange-bias field $\mu_0 H_{\text{ex}} \approx 1100$ mT (Figure 1B) due to the
16 antiferromagnetic exchange-coupling to the Tb-dominated ferrimagnet, which in turn switches
17 when the applied field exceeds the coercivity $\mu_0 H_{\text{c,TbFe}} \simeq 6500$ mT. The gradual increase of the
18 net magnetization in fields above 1100 mT indicates a strong antiferromagnetic coupling energy
19 compatible with our earlier results on similar systems.¹⁴
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23 MFM data of the ferrimagnetic sample SL show domains with a magnetization pointing up ($\odot$)
24 and down ($\otimes$) (Figure 1C). Note that the magnetization of the MFM tip was set in the up (positive
25 external field) direction. Consequently, a domain with up magnetization direction generates an
26 attractive force (gradient) resulting in a negative shift of the cantilever resonance frequency. As
27 reviewed in section 3 of the Supporting Information, the *thickness loss* dictates the predominance
28 of domain wall contrast in the MFM image, consistent with the spatial locations with large stray
29 field amplitude. At the imaging temperature of 10.5 K the magnetic moments of the Tb atoms
30 dominate those of Fe in the ferrimagnetic layer and the moments of the Co/Pt-multilayer.¹⁴ Hence,
31 we can refer to the Tb- or ferrimagnet moment direction indistinctly. Applying fields of 5000 mT
32 in the up direction, just short of the coercivity, has only a minor effect on the micromagnetic state
33 of sample SL; the arrows in Figure 1F highlight some of the few locations where a slight domain
34 wall motion and a re-pinning of the wall at other locations occurred when the field was raised from
35 3000 to 5000 mT. The high coercivity can thus be attributed to strong domain wall pinning. We
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Figure 1: Hysteresis loops of a single ferrimagnetic TbFe layer (sample SL) (A), and an exchange-coupled double layer structure consisting of a Co/Pt-multilayer on an amorphous ferrimagnetic TbFe layer (sample ECDL) (B), obtained from VSM measurements at 10.5 K. For a field larger than 1100 mT (highlighted by the red vertical line) applied parallel to the magnetization of the ferrimagnetic film, the magnetization of the Co/Pt-multilayer begins to revert. (C) and (D) MFM frequency shift Δf data of a $(3\mu\text{m})^2$ area of sample SL (gray background) at 0 mT and 3000 mT field, respectively. The $\odot$ and $\otimes$ symbols indicate the Tb-dominated magnetization direction. The field is applied in the up ($\odot$) direction, and the MFM tip has an up ($\odot$) magnetization. (E) and (F) Higher magnification images of MFM data acquired at 3000 and 5000 mT at the location highlighted by the dashed rectangle in panels (D). (G) to (N) MFM frequency shift Δf data of sample ECDL acquired in different fields. Again, the $\odot$ and $\otimes$ symbols indicate the Tb-dominated magnetization direction of the TbFe film. Note that the magnetization of the Co/Pt-multilayer is initially down inside the domain with a Tb up magnetization. (O) Comparison of the overall frequency shift contrast of the MFM data of sample SL (filled blue circles) with that of sample ECDL (filled red squares). Three magnetization stages can be observed for sample ECDL.

conclude that surprisingly large spatial variations in the magnetic properties of the TbFe layer must exist, confirming conclusions of earlier work performed on similar amorphous ferrimagnetic thin films.^{24,25}

The comparison of MFM data taken on sample SL in different fields reveals a monotonic increase of the MFM contrast with field (see filled blue circles Figure 1O). Because all measurements are carried out with the exact same MFM tip and with a tip-sample distance actively maintained at 7.0 ± 0.5 nm,²⁶ the frequency shift levels and thus the image contrast can be quantitatively compared. We can attribute the increase of the image contrast to an improved alignment of the magnetic moments in the MFM tip which must be taken into account when modeling high field MFM data as discussed in the next section. Other changes in the imaging properties of the MFM tip remain negligible.

While the domain structure and the MFM contrast of sample SL show only a minor evolution in fields up to 5000 mT (Figure 1C to F), a significant change in the micromagnetic structure and image contrast are apparent in the corresponding MFM images acquired on sample ECDL (Figure 1G to N). Phenomenologically, the magnetization reversal of the ferromagnet proceeds in three stages. **Stage 1:** between 0 and about 1000 mT (Figure 1G and H), a faint granular pattern appears inside the up domains (with Tb magnetization up and Co magnetization initially down), and the overall image contrast increases slightly from about 2.2 to about 2.9 Hz (part of the red curve in Figure 1O highlighted with yellow background color). **Stage 2:** above 1000 mT and below 2000 mT, a strong increase of the image contrast is observed, accompanied by the appearance of a pronounced granular contrast inside the up domain (part of the red curve in Figure 1O highlighted with green background color). **Stage 3:** above 2000 mT, the overall image contrast gradually approaches that of sample SL (compare the red to the blue dashed curve in Figure 1O in the area highlighted with blue background color), while the granular contrast becomes noticeably fainter (c.f. Figure 1K to N). At 7000 mT, above the coercive field of the TbFe layer, the down domains vanish and the granular contrast persists, albeit with further decreased amplitude (Figure 1N).

At first glance, the pronounced increase of the overall frequency shift contrast observed in

Stage 2 (see filled red squares in Figure 1O) is consistent with the formation of an in-plane domain wall near the Co/Pt-multilayer/TbFe interface, as established previously.^{13,15,27,28} However, our high-resolution MFM images (Figure 1G to N) do not show a homogenous increase in contrast, but reveal a highly inhomogeneous reversal process within the Co/Pt-multilayer.

To better assess the process taking place in the up domains (Tb up and Co initially down), panels A to O of Figure 2 show a series of image cuts taken inside the area highlighted by the dotted square in Figure 1G, in fields increasing from 0 to 7000 mT, with some extra fields that were not shown in Figure 1G to N. The data are displayed in a ± 1 Hz frequency shift scale, smaller than the ± 2.5 Hz scale used for Figure 1C to N. A more quantitative characterization of the pattern type can be obtained from a calculation of the cross-correlations between the different images (see Figure S7B and Figure S7C of the Supporting Information.).

Modeling of the MFM data and Discussion

The characteristics of the magnetic structures that give rise to our observations can be inferred from matching simulated MFM contrast from candidate micromagnetic structures to measured MFM data. The results for the three magnetization stages and schematics of the magnetization structures used for the simulations are displayed in Figure 3. A schematic of the zero-field case, where the magnetization of the domains in the Co/Pt-multilayer is antiparallel to that of the TbFe layer is displayed in Figure 3A. The domain pattern can be extracted from the MFM image in Figure 3C. The MFM contrast in Figure 3B is then calculated from the stray fields of the domains in the Co/Pt and TbFe using the calibrated tip-equivalent magnetic charge distribution (see section 5 of the Supporting Information). The latter is obtained from an MFM tip calibration procedure²⁹ (see sections 3 and 4 of the Supporting Information). Similarly, the MFM contrast for the more complex cases can be obtained from the different layers indicated in Figure 3D, G, J, and M and from the corresponding TbFe up/down domain pattern and Co/Pt-multilayer subdomain patterns. The detailed procedure used for the simulation of the MFM contrast is described in sections 5-8 of

Figure 2: Magnified views of the MFM frequency shift Δf data obtained on the ECDL sample, cut from the up domain (TbFe up, Co/Pt down) at the location of the dotted square in Figure 1G. (A) to (H): fields from 0 to 1000 mT, representing magnetization **Stage 1**. The contrast increases with the field, but the granular pattern does not change its appearance. (I) to (K): fields from 1200 mT to 2000 mT, representing magnetization **Stage 2**. The granular pattern changes substantially and its contrast reaches a maximum. (L) to (O): fields from 3000 mT to 7000 mT, representing magnetization **Stage 3**. The contrast decreases with increasing field, but the pattern does not change its appearance.

the Supporting Information.

Figure 3: Comparison of modeled (panels in the second row) and measured (panels in the third row) MFM data. (A), (D), (G), (J), and (M) Schematic of candidate magnetization structures used for modeling the MFM contrast of the different magnetization stages. (B), (E), (H), (K), and (N) Simulated MFM images obtained from the candidate magnetization structures (see sections 5-8 of the Supporting Information). (C), (F), (I), (L), and (O) Measured MFM images.

At zero field, the local magnetization of the ferromagnetic Co/Pt-multilayer is coupled antiparallel to that of the ferrimagnetic layer, so that each domain comprises an up (or down) magnetization direction of TbFe and correspondingly down (or up) magnetization direction for Co/Pt (see schematic in Figure 3A). The MFM data simulated from it (Figure 3B) match the weak domain contrast and also the apparent structure of the domains walls of the measured data (Figure 3C) well (see also Figure S3 and corresponding text in section 5 of the Supporting Information.). Because the model magnetization pattern schematic in Figure 3A does not account for small variations of the magnetic moment areal density inside the domains, the weak granular contrast visible in the measured data (Figure 3C) is not reproduced. This deviation is negligible for the simulation of the zero-field MFM data. For fields other than zero, the evolution of the local areal magnetic moment with the external field must be considered to explain the pronounced granular contrast, i.e. the

magnetization model must be refined, as we now discuss.

In **Stage 1** (0-1000 mT), the gradual increase of the contrast of the granular structures in Figure 2A-H in increasing fields is compatible with a rotation of the initially down magnetic moments of the Co/Pt towards the field (up) direction. Because the magnetic moments of Co/Pt-multilayer at the interface are pinned to the magnetic moments of the high-anisotropy TbFe film, the Co/Pt-multilayer magnetic moments at the top surface are expected to rotate more than those near the interface; this is the spin chain oriented along the z -axis that we consider in Figure S4Q of the Supporting Information. Our MFM data further suggest that the degree of rotation varies over a lateral length scale of about 30 nm corresponding to the Co/Pt magnetic grain diameter (see Figure 2A to H). To model the observed MFM contrast, we significantly reduced the complexity of the system and with it also the number of free parameters. The vertical structure of the spin chains at any given location on the image plane is modeled by a corresponding spatial distribution of ‘sub-domain blocks’ with zero magnetization, located at the top of Co/Pt-multilayer and reaching into different depths toward the interface (gray blocks in Figure 3D and Figure S4P of the Supporting Information). The lateral distribution is depicted in Figure S4L of the Supporting Information. The excellent agreement between modeled (Figure 3E) and measured (Figure 3F) MFM data demonstrates that such a model of the micromagnetic state is a reasonable description of the more complex state.

The significant increase of the contrast and the major changes of the patterns occurring in **Stage 2** ($1000 \text{ mT} < B \leq 2000 \text{ mT}$) (Figure 2I-K) are consistent with a pronounced increase of the lateral inhomogeneity of the z -component of the magnetic moments inside the Co/Pt-multilayer. The latter is compatible with isolated Co/Pt grains switching their magnetization from a canted down to a canted up state (as used in our modeling). This is reminiscent of a Stoner-Wohlfarth magnetization process with a field applied away from the easy axis where an instability of the magnetization state occurs. We expect that the rotation of the magnetization near the top of the Co/Pt-multilayer will increase with the applied field, up to a critical field, where the magnetization switches to a direction better aligned with the external field within a small additional field interval.

Then, an in-plane domain wall will appear on both sides of the interface of the ECDL structure. Thus, at specific locations the Co/Pt magnetization will have flipped towards the up direction of the applied field, resulting in a profusion of small domains. Their appearance defines **Stage 2** in the reversal process of Co/Pt-multilayer in these systems. Because these columns with a flipped magnetization are surrounded by columns with a rotated (not flipped) magnetization (similar to that of **Stage 1**), a strong stray field is generated from the increased local up/down magnetic moments of the Co/Pt-multilayer (Figure 2I to K). Figure 3G depicts a schematic model of this micromagnetic state. Using a pattern similar to that displayed in Figure S4L of the Supporting Information but reflecting the up/down pattern observed for Stage 2 by MFM, and a vertical moment distribution as displayed in Figure 3G, an agreement between the simulated (Figure 3H) and measured (Figure 3I) data is again obtained. For further details of the modeling process see Figure S5 of section 7 of the Supporting Information and the corresponding text. Note that the magnetic moments of the reversed grains align well with the external field at the top surface of the Co/Pt-multilayer and deviate more from the external field direction towards the interface with the ferrimagnetic film.

In the third magnetization stage ($2000 \text{ mT} < B \leq 7000 \text{ mT}$), the pattern of the granular contrast inside the up domain no longer changes, and a reduction of its contrast in increasing fields is observed (see Figure 2L to O), as the interfacial wall is compressed. As the Co/Pt-multilayer approaches saturation, its magnetic moment distribution becomes more homogeneous and its stray field thus decays (note that the stray field of a layer with a homogeneous magnetization vanishes). The measured contrast then predominantly arises from the stray field of the TbFe layer that is strongest near the walls of the micron-sized domains of the TbFe layer (see also section 3 of the Supporting Information). We model this third magnetization stage with a spatial distribution of ‘sub-domain blocks’ which now expand to both sides of the ECDL interface (see gray areas depicted in Figure 3J and Figure S6 of the Supporting Information). Again, a match between simulated (Figure 3K) and measured MFM data (Figure 3L) is obtained. When the applied field reaches the TbFe coercivity, lateral domain wall motion occurs inside the TbFe film, and the type of pattern and contrast found inside the up domain (as illustrated in Figure 3J) is established over the

complete film (Figure 3M). Further details on the modeling of the MFM contrast of magnetization stage 3 can be found in section 8 of the Supporting Information.

Micromagnetic Modeling

To understand the nucleation-governed magnetization process for the ECDL sample, modeling was performed using the finite element package *magnum.fe*.³⁰ The idea is to attribute the local contrast changes observed by MFM to variations in the anisotropy of the Co/Pt-grains, and their exchange with the TbFe ferrimagnetic layer. Typically, a strong perpendicular anisotropy with a small grain-to-grain variation is obtained if the Co/Pt-multilayer is grown on a 5-10 nm thick (111)-oriented Pt seed, which is however not the case here. Instead, the Co/Pt-multilayer is grown on the top of the amorphous TbFe ferrimagnetic layer. We argue that this reduces the anisotropy of the Co/Pt-multilayer, and leads to an increased grain-to-grain variation of the anisotropy, and possibly also affects the interlayer exchange. We further assume the existence of a strong spatial variation of the interlayer exchange as also confirmed by previous work.^{15,31} The parameters for the Co/Pt and TbFe layers used for simulations are given in Table 1.

Table 1: Material parameters of the (Co/Pt)/TbFe ECDL sample used for the micromagnetic modeling. K_{eff} is the effective anisotropy constant, J_S is the saturation magnetization, K_u is the uniaxial anisotropy constant, obtained from the effective anisotropy and the saturation magnetization, A_{ex} is the exchange coupling, and $J_{\text{TbFe/CoPt}}$ is the exchange between the TbFe layer and the Co/Pt-multilayer. The angle of the grains relative to the z-axis was tilted to avoid metastable states. α is the dimensionless Gilbert damping constant.

layer	Co/Pt	TbFe
K_{eff} [kJ/m ³]	50 to 250	2500
J_S [T]	0.4	0.25
K_u [kJ/m ³]	114 to 314	2525
A_{ex} [pJ/m]	4	4
$J_{\text{TbFe/CoPt}}$ [mJ/m ²]	-1 to -8	
$\angle(\mathbf{K}_{\text{uni}}, \mathbf{e}_z)$ [°]	10	1.0
α	1	1

In a first approach, we perform 1-dimensional spin-chain calculations, using a discretization length of 2 nm. The applied field was raised in steps of 50 mT, and the micromagnetic state was relaxed for 1 ns. Only minor loops, where no reversal of the TbFe magnetization occurs are calculated. Referring to Figure 4B, two different types of minor loops are apparent:

Loops of **type 1**, for an interlayer exchange of $J_{\text{TbFe/CoPt}} = -1, -1.5$, and -2 mJ/m^2 show a hysteretic switching of the magnetization at the critical fields $\mu_0 H_{\text{cu}}$ and $\mu_0 H_{\text{cd}}$ for increasing and decreasing fields, respectively. Although the magnetic moments of the spin chain do not rotate uniformly, the dependence of their directions on the applied field is reminiscent of a Stoner-Wohlfarth magnetization process: if the applied up-field is increased, the initially down magnetic moments of the Co/Pt-multilayer gradually rotate upward. At a nucleation field $\mu_0 H_{\text{cu}}$, the magnetic moments switch into near-alignment with the applied up-field, aligning asymptotically as it is further increased. If the applied field is decreased again, the flip of the magnetic moments towards the down direction occurs at the field $\mu_0 H_{\text{cd}} < \mu_0 H_{\text{cu}}$, such that a hysteresis occurs.

Loops of **type 2**, for an interlayer exchange of $J_{\text{TbFe/CoPt}} = -3, -4, -6$, and -8 mJ/m^2 again show a strong increase of the up magnetization within a small field interval around a critical field $\mu_0 H_c$, but the magnetization process remains rotary and thus hysteresis free, reminiscent of an exchange spring behavior.

For a low anisotropy ($K_{\text{eff}} = 50 \text{ kJ/m}^3$ displayed in Figure 4A), the magnetization remains hysteresis free (with the exception of the loop for $J_{\text{TbFe/CoPt}} = -1 \text{ mJ/m}^2$), but the critical fields are rather small and their distribution remains narrow even with a variation of the interlayer exchange by a factor of eight. Increased anisotropies lead to higher critical fields, but also to the occurrence of type 1 loops with wide hystereses (Figure 4B to E) that are not compatible with the absence of a hysteresis of the macroscopic minor loop. It thus remains challenging to simultaneously capture two relevant experimental observations, i.e. the absence of a hysteresis of the macroscopic minor loop together with the wide range of a substantial magnetization reversal of the grains (of their critical fields) observed by MFM (Figure 2I to K).

In Figure 4F magnetization curves are plotted for K_{eff} values between 50 and 250 kJ/m^3 for a

Figure 4: Minor magnetization reversal loops obtained from a spin-chain model for different values of K_{eff} and $J_{\text{TbFe/CoPt}}$. **A** $K_{\text{eff}} = 50 \text{ kJ/m}^3$, and $J_{\text{TbFe/CoPt}} = -1$ to -8 mJ/m^2 . **B** $K_{\text{eff}} = 150 \text{ kJ/m}^3$, and $J_{\text{TbFe/CoPt}} = -1$ to -8 mJ/m^2 . **C** $K_{\text{eff}} = 250 \text{ kJ/m}^3$, and $J_{\text{TbFe/CoPt}} = -1$ to -8 mJ/m^2 . **D** $J_{\text{TbFe/CoPt}} = -1.5 \text{ mJ/m}^2$, and $K_{\text{eff}} = 50$ to 250 kJ/m^3 . **E** $J_{\text{TbFe/CoPt}} = -6 \text{ mJ/m}^2$, and $K_{\text{eff}} = 50$ to 250 kJ/m^3 . **F** $J_{\text{TbFe/CoPt}}/K_{\text{eff}} = -0.027 \mu\text{m}$, and $K_{\text{eff}} = 50$ to 250 kJ/m^3 .

$J_{\text{TbFe/CoPt}}/K_{\text{eff}}$ -ratio kept constant at $-0.027 \mu\text{m}$. The loops for $50 \text{ kJ/m}^3 \leq K_{\text{eff}} \leq 100 \text{ kJ/m}^3$ are of type 2, but still exhibit a strong increase of the magnetization in a very small field interval at critical fields between about 1000 to 1500 mT. Anisotropies $K_{\text{eff}} \gg 100 \text{ kJ/m}^3$ lead to an increase of the critical fields to more than 2000 mT, but the loops are of type 1, albeit with narrow hystereses. Note that the coexistence of loops of type 1 and 2 is compatible with the absence of a macroscopic hysteresis of the minor loop, as long as the width of the hystereses of the type 2 loops remain small. This suggests that the mechanism leading to a variation of the grain-to-grain anisotropy may also affect the interlayer exchange of the Co/Pt-grain with the TbFe ferrimagnetic layer.

The results obtained with the 1-dimensional spin chain model justifies the vertical magnetic moment distributions chosen for our quantitative modeling of the MFM contrast (Figure 3). The

1-dimensional spin-chain model however does not consider a lateral exchange coupling between the Co/Pt grains. To test whether such an intergranular exchange affects the reversal process, finite element calculations were performed for a $100\text{ nm} \times 100\text{ nm}$ area of the ECDL sample containing grains generated via Voronoi tessellation, with an average grain diameter of 10 nm (Figure S8 of the Supporting Information). The uniaxial anisotropy was varied between 47 and 290 kJ/m^3 , and the $J_{\text{TbFe/CoPt}}/K_{\text{eff}}$ -ratio kept constant at $0.027\text{ }\mu\text{m}$ (see section 10 and Figure S9 of the Supporting Information). We found that the effect of the intergranular exchange on the reversal of a specific grain can be neglected even for the small grain diameter of 10 nm used in our model.

Decoupling the Co/Pt-multilayer from the TbFe layer

Given that the coupling strength between the Co/Pt-multilayer and the ferrimagnetic film is one of the parameters determining the reversal of the Co/Pt-multilayer, an interesting possibility is modifying it to shift the onset of that reversal, as for example previously explored by Tang et al.³² In their work, the ferromagnetic Co/Ni-multilayer was grown on a Ta(5 nm)/Cu(3 nm)-seed layer providing a (111)-oriented growth of the Co/Ni and therefore a strong PMA with a presumably narrow distribution. The high-coercivity amorphous TbCo alloy layer was then deposited onto the ferromagnetic multilayer leading to a coupling strength of -4.3 mJ/m^2 (which is in the middle of the range of coupling strengths used to model our system; see table 1). Cu or Ta interlayers were used to reduce the ferro-ferrimagnet coupling.

In our work, the coupling strength was progressively reduced with a Pt spacer layer of increasing thickness. The exchange bias field $\mu_0 H_{\text{ex}}$ is measured as the onset of the Co/Pt magnetization reversal in the VSM magnetization loops, with the error bars of $\pm 50\text{ mT}$ representing the uncertainty (red line in Figure 1B, blue line in Figure 5A, and green line in Figure 5B for 0, 0.7, and 2.5 nm Pt interlayer thickness, respectively). The decrease of exchange-bias field with increasing Pt spacer layer thickness is plotted in Figure 5C. It decreases from about 1100 mT for the ECDL sample without Pt spacer to about 300 mT for the one with 2 nm Pt spacer, and is almost 0 mT for

the ECDL sample with 2.5 nm Pt spacer layer.

Figure 5: (A), (B) magnetization reversal loops measured by VSM at 10.5 K for ECDL samples with 0.7 and 2.5 nm Pt interlayer thickness. (C) Dependence of the exchange-bias field on the Pt spacer layer thickness. The exchange-bias fields are determined from the VSM magnetization loops of ECDL samples with Pt spacer thicknesses ranging between 0 and 2.5 nm.

Figure 6A-J and Figure 6K-O show the dependence of the domain structures on the applied field of the samples with Pt spacer layer thicknesses of 0.7 and 2.5 nm (samples ECDL-Pt0.7nm and ECDL-Pt2.5nm), respectively. For all samples with Pt spacer thicknesses smaller than 2.0 nm, a three-stage magnetization process similar to sample ECDL is observed. As an example, for the sample ECDL-Pt0.7nm, the MFM frequency shift data of a $3\mu\text{m} \times 3\mu\text{m}$ area are shown in Figure 6A-J, where the background colors highlight the three magnetization stages. Analogous to sample ECDL, sub-images (not shown) cut from the domains with an initial down Co/Pt magnetization are used to assess the field ranges of stages 1, 2, and 3, which in this case are 0-700 mT, 800-1200 mT, and 1200-7000 mT, respectively. Notice that the field ranges for stages 1 and 2 shift to smaller values compared to sample ECDL, i.e. they decrease with increasing Pt spacer thickness. This trend is preserved for ECDL-tnmPt samples with Pt spacers of a thickness $t = 0.4, 1.2,$ and 2.0 nm, in agreement with the lower exchange-bias field and smaller interfacial exchange-coupling energy observed in the VSM loops (not shown).

In contrast, the MFM data for the sample ECDL-Pt2.5nm reveal a magnetization process different from that of samples with thinner Pt spacers (see Figure 6K-O). At zero field, micron-size TbFe domains with a strong domain wall contrast, comparable to those observed on the single layer sample SL, are visible (Figure 6K). Such a strong contrast is compatible with a fully mag-

netized Co/Pt layer, either up or down (Figure 6P). We note that a negative field was applied to the sample prior to the measurements in Figure 6K-O, which saturated the Co/Pt layer in the down direction. At zero field, the interfacial exchange is too weak to nucleate a Co/Pt domain with an up magnetization over the TbFe domain with a down magnetization, thus the Co/Pt film remains saturated in the down direction.

A small up field of 60 mT is sufficient to nucleate Co/Pt up domains (Figure 6L and Q). This is consistent with the exchange-bias field for sample ECDL-Pt2.5nm, which almost vanishes (Figure 5B and C). The MFM data obtained at 60 mT, is compatible with the nucleation of the Co/Pt up domains within the area of the TbFe down domains followed by a rapid expansion of the Co/Pt up domains over the boundaries of the TbFe down domains. This preferred nucleation within the area of the TbFe down domains indicates that an antiferromagnetic interfacial exchange interaction, although extremely weak, must still exist and dominate the ferromagnetic dipolar coupling (as a reminder, at zero field, the interfacial exchange-coupling was too weak to allow the nucleation of a Co/Pt up domain). Furthermore, the observed crossing over of the Co/Pt domain wall into the area of the TbFe up domain (facilitated by the up field emanating from the TbFe up domain which is strongest near the domain wall) is only possible because this interfacial exchange interaction is extremely weak. In addition, the faint granular contrast inside the Co/Pt down domains in the zero-field state (Figure 6K) becomes more pronounced at 60 mT (Figure 6L). We attribute this to an incipient rotation of the down magnetic moments near the top surface of the Co/Pt film towards the applied up field, reminiscent of that occurring in stage 1 of the ECDL samples with thinner Pt spacers.

Figure 6M shows the MFM data acquired in a field of 80 mT. It can be seen that the Co/Pt up domains have expanded further into the area of the TbFe up domains (see schematic in Figure 6R), indicating that the Zeeman energy of the Co/Pt magnetic moments in a field of only 80 mT is sufficient to dominate the interfacial exchange energy. Clearly, the magnetization process of the Co/Pt layer occurs by nucleation inside the areas of the TbFe down domains, but then proceeds by conventional lateral motion of the domain walls.

For fields of 100 mT (Figure 6N), the coupling is too weak to suppress an expansion of the Co/Pt up domain over the whole area of the TbFe up domain (see schematic Figure 6S). MFM images in fields above 100 mT (see for example, Figure 6T and schematic displayed in Figure 6T) reveal no changes in the magnetic state of the sample (i.e. Co/Pt is saturated) until the coercive field of the TbFe layer is surpassed (where the TbFe down domains vanish).

An increasing thickness of the Pt interlayer is also expected to improve the anisotropy of the Co/Pt grains, while their exchange with the TbFe is decreased. Our spin-chain modeling then predicts a transition from type-2 magnetization loops that are hysteresis free to type-1 loops with a wide hysteresis (see for example the blue loop in Figure 4C). For such conditions, we also expect a hysteretic minor magnetization loop. A more detailed analysis of the hysteretic behavior of isolated grains and an analysis of the hysteresis width on the interlayer thickness is however beyond to scope of this work.

Summary

We have found that strongly exchange-coupled systems consisting of Co/Pt-multilayer/TbFe double layers exhibit a complex, nucleation-dominated three-stage magnetization process in the soft Co/Pt-multilayer. We attribute this to grain-to-grain variations in anisotropy of the Co/Pt film arising from the growth on the amorphous TbFe layer, and an inhomogeneous exchange coupling with the TbFe film. The latter has been discussed by other recent studies.^{31,33} This would strongly affect the macroscopic properties of, for example, spin-valve structures with footprints smaller than a square micrometer. Clearly, the distribution of the anisotropies of the grains in the ferromagnetic film must be kept small to avoid a granular magnetization switching process.

We further showed that the interfacial coupling energy can be tuned to smaller values with an interfacial Pt layer of increasing thickness. While the sample with the smaller interlayer thickness again shows a three-stage magnetization process (with the expected reduction of the field range for stages 1 and 2), the magnetization process of the sample with the 2.5 nm spacer layer is reminiscent

Figure 6: (A-J) MFM frequency shift Δf data of a $3\mu\text{m} \times 3\mu\text{m}$ area of the ECDL sample with 0.7 nm Pt spacer taken in various fields. The $\odot$ and $\otimes$ symbols indicate the Tb-dominated magnetization direction of the TbFe film. Note that the Co/Pt magnetization is down inside the domain with a Tb up magnetization. Three magnetization stages are visible, similar to the ECDL sample without a spacer (Figure 1). Measured MFM data of the ECDL sample with 2.5 nm Pt spacer and their candidate magnetization structures. (K), (L), (M), (N), and (O) MFM frequency shift Δf data of a $3\mu\text{m} \times 3\mu\text{m}$ area taken in various fields. The $\odot$ and $\otimes$ symbols indicate the Tb-dominated magnetization direction of the TbFe film. (P), (Q), (R), (S), and (T), Schematic of candidate magnetization structures for different magnetization stages. The yellow areas denote the 2.5 nm Pt spacer between Co/Pt and TbFe layers.

of that of ferromagnets that are weakly coupled to an antiferromagnet in conventional exchange-bias systems.^{17,34} In these systems the reversal process is governed by a lateral motion of the domain wall in the soft FM layer. The expansion of the domains with a magnetization parallel to the external field then breaks the interfacial exchange interaction.

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