

## Direct observation of field and temperature induced domain replication in dipolar coupled perpendicular anisotropy films \*

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### Abstract

Dipolar interactions in a soft/Pd/hard [CoNi/Pd]<sub>30</sub>/Pd/[Co/Pd]<sub>20</sub> multilayer system, where a thick Pd layer in between two ferromagnetic units prevents direct exchange coupling, are directly revealed by combining magnetometry and state-of-the-art layer resolving soft X-ray imaging techniques with sub-100 nm spatial resolution. The domains forming in the soft layer during external magnetic field reversal, are found to match with the domains previously trapped in the hard layer. The low Curie temperature of the soft layer allows varying its intrinsic parameters via temperature and thus studying competition with dipolar fields due to the domains in the hard layer. Micromagnetic simulations elucidate the role of [CoNi/Pd] magnetization, exchange and anisotropy in the duplication process. Thermal activation kinetics need to be considered for a quantitative understanding of the experimental results. Finally, thermally activated domain replication in remanence during temperature cycling is demonstrated.

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## Text

Dipolar interactions in layered magnetic systems have recently attracted increasing attention in order to improve the reliability of magneto-electronic devices [1] and for applications in multilevel magnetic recording [2]. In this context stray field induced replication of domains or bit-patterns in perpendicular anisotropy systems has been investigated as well [3]. In previous studies, standard techniques such as magnetometry, Magnetic Force Microscopy (MFM) or Kerr microscopy have been used to characterize the evolution of the magnetic configuration during field cycle at room temperature [3,4]. However, such techniques average over the complete ferromagnetic layer stack and do not allow studying the hard or soft layer separately.

Thermally-induced magnetization reversal with or without an external static magnetic field is nowadays under heavy discussion since it is being considered for applications in Magnetic Random Access Memory (MRAM) or Magneto-Optic (MO) recording devices [5]. Domain replication has already been proposed as a readout mechanism for MO recording in Magnetic Amplifying Magneto-Optical Systems (MAMMOS), but it is also believed to be useful for thermally-induced writing [6].

In the present letter, we report on high resolution imaging of the domain replication in an all-perpendicular anisotropy system. Using a soft/Pd/hard multilayer system, namely  $[\text{CoNi/Pd}]_{30}/\text{Pd}/[\text{Co/Pd}]_{20}$ , we combine magnetometry and state-of-the-art element-specific X-ray imaging techniques [7-9] to investigate the magnetic

configuration of the hard and soft layer independently during external magnetic field cycles at temperatures in the range of 150 K up to 350 K. Field cycles lead to the formation of a soft layer magnetic domain state that is identical to the domain configuration previously trapped in the hard layer. Furthermore we exploit the strong temperature dependence of the [CoNi/Pd] soft layer to study the domain duplication as a function of temperature and demonstrate thermally activated domain replication at remanence.

Using DC magnetron sputtering, the layer sequence Pd(3nm) / [Co<sub>30</sub>Ni<sub>70</sub>(0.2nm) / Pd(1.5nm)]<sub>30</sub> / Pd(10nm) / [Co(0.3nm)/Pd(0.8nm)]<sub>20</sub> / Pd(1.2nm) is deposited onto Si wafers as well as Si<sub>3</sub>N<sub>4</sub> membranes for transmission soft X-ray imaging. The macroscopic magnetic properties are measured using low-temperature Vibrating Sample Magnetometry (VSM). Fig. 1a compares hard and soft multilayer magnetizations as a function of temperature. The hard [Co/Pd] system, deposited at 7 mTorr Ar pressure, reveals a well-defined perpendicular-to-the-sample-plane anisotropy and an almost constant magnetization within the considered temperature range from 5 K to 400 K. However, the Ni concentration in the soft [CoNi/Pd], deposited at 3 mTorr Ar pressure, is tuned to obtain a Curie temperature ( $T_C$ ) close to 350 K, i.e. its magnetization and anisotropy (Fig. 1b) decrease continuously from 5 K to 350 K, and the system turns paramagnetic for higher temperatures [10]. We measure the temperature dependence of the magnetization at remanence as well as in an external field of 1 kOe and find slight differences between 310 and 350 K, indicating that the out-of-plane anisotropy competes with an in-plane shape anisotropy when approaching  $T_C$  (Fig. 1a).

Fig. 2a presents hysteresis loops measured at 300 K with the field perpendicular to the sample plane. The major loop reveals two distinct steps corresponding to the independent reversals of the soft and the hard layer. Starting from positive saturation the soft layer reverses at  $H_N^0 = -350$  Oe and stays anti-parallel to the [Co/Pd] until about  $H = -4$  kOe, where the hard layer reversal occurs. A minor field cycle performed between  $\pm 2$  kOe shows no residual bias field and indicates that the Pd (10 nm) layer in between the two ferromagnetic multilayers prevents any direct exchange interaction. Sweeping the external perpendicular magnetic field from 8 kOe to -4.2 kOe, where the hard layer has reversed half of its magnetization, we create a domain state in the hard layer with about 50% up and 50% down domains. To directly image hard and soft layer domains with sub-100 nm spatial resolution, we performed magnetic transmission soft X-ray microscopy (MTXM) using the full field soft X-ray set up of beamline 6.1.2. at the Advanced Light Source (ALS) in Berkeley CA [11, 12]. X-ray magnetic circular dichroism (XMCD) provides element specific magnetic contrast, which for these studies enables to separate layer resolved magnetic domain configurations in both the hard and the soft layer by tuning the photon energy to the Co- and Ni- $L_3$  absorption edges, resp. MTXM images were recorded with magnetic fields up to 2 kOe pointing perpendicular to the sample plane. After reversing half of the hard layer magnetization, we first confirm (Fig. 2c) the presence of a labyrinth pattern with 50% up and 50% down domains in the hard layer with an average size similar to the natural size measured at 300 K with MFM (Fig. 1c and 1d). Since the soft layer magnetization is saturated for Fig. 2c, the domain configuration measured at the Co- $L_3$  edge at -2 kOe reflects the hard layer state only.

Subsequently we measure the reversal of the soft layer in a minor loop between  $\pm 2$  kOe (open circles in Fig. 2a). Corresponding MXTM images measured at the Ni  $L_3$  edge, to separate out the reversal of the soft layer only, are shown in Fig. 2d and 2e. Starting from positive soft layer saturation, we lower the external field and observe domain nucleation in the soft layer (Fig. 2d) around 0 Oe in good agreement with the VSM measurement (Fig. 2a). The nucleation field ( $H_N$ ) and the saturation field ( $H_S$ ) of the soft layer have significantly changed as compared to the previous minor loop measured for a uniform hard layer. This is more clearly visible in the derivatives of the descending hysteresis branches in Fig. 2b.  $H_N$  increases from  $H_N^0 = -250$  Oe to  $H_N^d = +130$  Oe, while  $H_S$  decreases from  $H_S^0 = -1.1$  kOe to  $H_S^d = -1.55$  kOe. Such changes in the soft layer hysteresis loop have already been reported on similar hard/soft systems and were assumed to originate from the influence of dipolar fields induced by the hard layer domains on the soft layer reversal [3]. At about  $-300$  Oe we reach a 50% up and 50% down domain state (Fig. 2e) that appears to match very well with the state previously trapped in the hard layer (Fig. 2c). At this stage of the minor loop the domain state of the hard layer has completely been copied (replicated) into the soft layer. In Fig. 2b, this replicated state reveals itself as a dip in the derivative. We find symmetric minor loops indicating close to perfect Return Point and Complementary Point Memory in the soft layer [13]. However, while the same replicated domain state was observed during several successive field loops, we found no evidence for an identical nucleation sequence.

Fig. 3a presents VSM measurements similar to Fig. 2a, but now measured at 150 K. In order to study the replication process with element-specific XMCD at different

temperature, we use the recently established lensless Fourier Transform Holography (FTH) technique [8]. The sample is illuminated with coherent X-rays through a gold mask with a circular 1.2  $\mu\text{m}$  diameter object aperture and a  $\sim 100$  nm reference hole to create a X-ray hologram that contains the relative phase between object and reference and thus, when transformed back into real space, yields a direct image of the object aperture domain pattern. Corresponding experiments are performed at beamline UE-52-SGM at BESSY in Berlin (Germany) using the ALICE setup [14], which allows holographic imaging at variable sample temperature and under external fields up to 7 kOe. Details about magnetic FTH technique can be found elsewhere [8, 9]. Figs. 3c and 3g show identical hard layer domain states measured at 150 K at the Co-L<sub>3</sub> edge at  $-4.6$  kOe before and after performing a soft layer minor loop, thus confirming the stability of the domains in the hard layer. Figs. 3d-3f show Ni-L<sub>3</sub> edge images measured along the soft layer minor loop from positive saturation to almost negative saturation. Similar as for 300 K, the hard layer domain state is replicated in the soft layer during magnetization reversal. A careful analysis of Fig. 2a and Fig. 3a, as provided by the derivatives of the descending hysteresis loop branches (Fig. 2b and 3b), reveals that the amplitudes of  $H_N$  and  $H_{\text{sat}}$  variations due to the domain state introduction in the hard layer differ with temperature. As shown in Fig. 4, the amplitudes of  $\Delta H_N = H_N^d - H_N^0$  and  $\Delta H_{\text{sat}} = H_{\text{sat}}^d - H_{\text{sat}}^0$  increase as the temperature is raised. The variation of  $\Delta H_N$  from 100 K to 310 K is two times larger than that of  $\Delta H_{\text{sat}}$  over the same temperature range.

We perform micromagnetic simulations to investigate the origin of the nucleation field reduction in the soft layer due to the domains in the hard layer. The hysteresis loop

of the [CoNi/Pd] is calculated from the solution of the Landau-Lifshitz-Gilbert (LLG) equations [15] with the [Co/Pd] magnetization held fixed to be in either the uniform state or the stripe domain configuration. In the simulations the domains in the hard layer are represented as 200 nm by 3000 nm parallel stripes of alternating magnetization perpendicular to the plane of the film. The CoNiPd layer is first uniformly magnetized in +7.5 kOe, then the applied field is swept between +/- 2.5 kOe and is superimposed to the magnetostatic interaction fields from the hard layer. The sweep rate of the field is sufficiently slow (6.4 ns/kOe) at a gyromagnetic damping  $\alpha=0.1$  to avoid cases where dynamic effects would dominate the switching behavior. Thermal fluctuations are not considered explicitly in the LLG equations, but are lumped into adjustments of the values for the magnetization  $M_s$ , the uniaxial anisotropy  $K_1$ , and the exchange  $A$  in the soft layer as a function of temperature.

The modeling of the experimental nucleation fields proceeds in two steps. First, we aim to match the  $H_N^0$  (without stripe domains) by adjusting the [CoNi/Pd] parameters to obtain effective values for the anisotropy  $K_{\text{eff}}$  and the exchange  $A_{\text{eff}}$  as a function of temperature. Secondly, the calculations are repeated with  $K_{\text{eff}}$  and  $A_{\text{eff}}$  to obtain the nucleation field  $H_N^d$  when stripe domains are present in the hard layer to finally estimate the reduction in nucleation field  $\Delta H_N$ . We find that it is generally possible to qualitatively obtain the loop shapes of Fig. 2 and Fig. 3 (see Fig. 4b); however,  $K_{\text{eff}}$  has to be significantly lowered compared to the experimental anisotropy measured from the hard axis loops (Fig. 1b). Typically  $K_{\text{eff}}$  was 25-30% of the experimental  $K$  with  $A_{\text{eff}}$  in the range of  $1.1-1.5 \times 10^{-11}$  J/m at  $T=150$  K, and  $0.1-0.5 \times 10^{-11}$  J/m at  $T=300$  K. Such low

values of  $K_{\text{eff}}$  in the soft layer also require a scaling of the duplication fields by a factor of  $\sim 0.35$  (e.g. by a commensurate adjustment of the hard layer magnetization). The lack of additional information at the microstructural level, e.g. pinning-site density in the [CoNi/Pd], do not allow us to go beyond this type of mean-field approach. With these caveats in mind, leading to uncertainties of the simulated nucleation fields (estimated  $\pm 100$  Oe), the calculated values qualitatively reproduce the trend of a reduction in nucleation field due to the stripe domains as well as the changes in the hysteresis loop shapes (Fig. 4b). Nevertheless, thermal variations of  $\Delta H_N$  cannot be quantitatively reproduced considering only soft layer parameters changes (Fig. 4a). We believe that thermal effects in the soft layer do not only lead to an effective reduction of the magnetization, exchange and anisotropy as a function of temperature (Fig. 1) but also initiate kinetics of magnetization hopping over local energy barriers. However, testing this hypothesis is beyond the scope of the current model and needs to be verified in future studies.

Finally, we take advantage of the temperature dependence in the competition between intrinsic soft layer parameters and dipolar interaction by demonstrating dipolar induced domain duplication during temperature cycles. First, at a sample temperature of 150 K, we reverse about 50 % of the hard layer magnetization, introducing the domain pattern seen in Fig. 5b, and subsequently measure the [CoNi/Pd]/Pd/[Co/Pd] magnetization at remanence as a function of temperature (Fig. 5a). After saturating the soft layer at 3 kOe, the external field is released to remanence again. Subsequently starting at 150 K, the temperature is cycled at remanence between 150 K and four

temperatures  $T_{\max}$  (250 K, 300 K, 310 K and 350 K). After increasing the temperature beyond 290 K, we observe an irreversible reduction in magnetization when cooling back down to 150 K. Using FTH imaging during the temperature cycles, we first verify that the hard multilayer domain state remains unaltered within the temperature range of 150 K to 350 K (Fig. 5b and 5f). Then we image the soft layer domain state at the Ni  $L_3$  edge at 250 K, 290 K and 300 K (Fig. 5c-5e). While at 250 K the soft layer remains uniformly saturated, at 290 K and 300 K, domains are progressively replicated (Fig. 5d-5e). The FTH field of view is only micrometric. To characterize the domain replication rate as a function of  $T_{\max}$  on the macroscopic scale, we calculate, from the VSM measurements, the percentage of non-reversed domains that has to be considered to fit the temperature cycle descending branches (Fig. 5a). We use the soft layer remanent magnetization curve measured for a uniform hard layer state as a reference (100 % of up domains). We reproduce the magnetization variations as a function of temperature after  $T_{\max} = 300$  K, 310 K and 350 K, considering 80 %, 61 % and 47 % of [CoNi/Pd] up domains respectively (Fig. 5a). The good agreement between the fits and the experimental data confirms, on a macroscopic scale, the formation of a stable soft layer domain state via dipolar duplication when the system is cooled back down to 150 K.

In conclusion, we present a direct visualization of stray-field induced domain duplication in an all-perpendicular anisotropy system consisting of a hard/Pd/soft multilayer system, where the two magnetic layers are exchange decoupled via a thick Pd interlayer. While varying the external magnetic field, as well as the sample temperature, and using layer resolving soft X-ray imaging techniques with high spatial resolution, we

directly show the influence of the hard layer magnetic domain configuration on the soft layer magnetization reversal. The competition between the intrinsic soft layer properties and the dipolar interaction is revealed during temperature dependent studies by tuning the soft layer Curie point well below the hard layer Curie point. Micromagnetic simulations reproduce qualitatively the temperature dependent changes of the soft layer hysteresis due to dipolar interactions. Finally, we demonstrate experimentally the concept of thermally assisted domain duplication at remanence.

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## Captions

**Figure 1.** a) Normalized magnetization versus temperature profiles of a  $[\text{Co}_{30}\text{Ni}_{70}(0.2\text{nm})/\text{Pd}(1.5\text{nm})]_{30}$  soft multilayer under 0 Oe (open circles) and 1 kOe (solid squares) and of a  $[\text{Co}(0.3\text{nm})/\text{Pd}(0.8\text{nm})]_{20}$  hard multilayer at remanence (triangles). b) Temperature dependence of the anisotropy for the  $[\text{CoNi}/\text{Pd}]$  soft layer. c) and d)  $2.5 \times 2.5 \mu\text{m}^2$  MFM images of demagnetized  $[\text{Co}/\text{Pd}]$  and  $[\text{CoNi}/\text{Pd}]$  multilayers respectively.

**Figure 2.** a) Normalized hysteresis loops measured at 300 K. Square solid symbols show the major loop, while dashed line and open circles correspond to minor loops with the hard layer in a uniform and a domain state, respectively. b) Derivative of the descending major and minor (solid and open symbols) hysteresis loop branch. c-e) MTXM images, each  $6 \times 6 \mu\text{m}^2$  in size, showing the magnetic domain configuration as observed for the three states marked in a) respectively. Image c) is collected at the Co- $L_3$  edge (778 eV) while images d) and e) are obtained at the Ni- $L_3$  edge (854 eV). Red lines are a guide to the eye.

**Figure 3.** a) Normalized hysteresis loops measured at 150 K. Full symbols represent a +/- 8 kOe major loop. Open symbols show a minor loop after trapping domains in the hard layer. b) Derivative of the descending major and minor (solid and open symbol) branch. c) and g): 1.2  $\mu\text{m}$  diameter FTH images of the magnetic domain configuration at the Co-L<sub>3</sub> edge at -4.6 kOe before and after the minor loop, respectively. d)-f): Magnetic domain configuration at the Ni-L<sub>3</sub> edge measured successively at different field as indicated on the hysteresis loop (a).

**Figure 4.** a) Experimental nucleation and saturation field difference,  $\Delta H_N$  (solid square) and  $\Delta H_{\text{sat}}$  (triangle), for the soft layer minor loops with and without domains in the hard layer, as a function of temperature. Open symbols correspond to the simulated  $\Delta H_N$  with the dashed line being a guide to the eye. b) Simulation of 300 K soft layer hysteresis loops considering a uniform (square) or stripe domain (circle) hard layer state, and their derivative in inset.

**Figure 5.** a) Remanent magnetization during temperature cycles from 150 K to  $T_{\text{max}}$  after trapping a domain state in the hard layer at 150 K, where  $T_{\text{max}}=250$  K (open triangles), 300 K (full triangles), 310 K (circles) and 350 K (squares). The crosses represent the remanent magnetization with no domains in the [Co/Pd] hard layer. The solid lines correspond to simulations as described in the text. b) and f) 1.2  $\mu\text{m}$  diameter FTH image of the magnetic domain configuration at the Co-L<sub>3</sub> edge for remanence at 150 K and 350K respectively. c) to e) Remanent magnetic domain configuration at the Ni-L<sub>3</sub> edge at 250 K, 290 K, 300 K.











