Field mapping and temperature dependence of magnetic domain memory induced by exchange couplings

Karine Chesnel\textsuperscript{1,5}, Brian Wilcken\textsuperscript{1}, Matthew Rytting\textsuperscript{1}, Steve D Kevan\textsuperscript{2,3}, and Eric E Fullerton\textsuperscript{3,4}

\textsuperscript{1} Physics Department, Brigham Young University, Provo, UT, USA
\textsuperscript{2} Physics Department, University of Oregon, Eugene, OR, USA
\textsuperscript{3} Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA, USA
\textsuperscript{4} Center for Magnetic Recording Research, UCSD, La Jolla, CA, USA
E-mail: kchesnel@byu.edu

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\textbf{Abstract.} Strong magnetic domain memory is achieved in [Co/Pd]IrMn exchange-biased ferromagnetic thin films when zero-field-cooled (ZFC) below their blocking temperature \(T_B\). By mapping out the amount of memory throughout the entire magnetization cycle, from nucleation to saturation, at different temperatures below and above \(T_B\), we discover how microscopic morphological changes in the magnetic domain patterns correlate with the macroscopic magnetic hysteresis, in the presence or absence of exchange couplings. Our unique inter-field correlation maps show that in the ZFC state, the film exhibits the highest amount of domain memory, exceeding 90\%, when domain patterns are compared at the same field value, in the coercive region of the magnetization loop. However, domain patterns also cross-correlate surprisingly well when measured at different field values, on a wide field range centered about the coercive region. The shape and symmetry of the correlation maps provide further insights into the microscopic morphological changes in the domain patterns and the amount of reversibility in the reversal process, at the nanoscale.

\textsuperscript{5} Author to whom any correspondence should be addressed.
1. Introduction: hysteresis and magnetic domain memory

Magnetic hysteresis, the occurrence of lagged responses in the magnetization of a material, when applying a magnetic field in reversed directions, has stirred up curiosity since ancient times [1, 2]. Commonly present in ferromagnetic (F) materials, hysteretic effects are generally observed at the macroscopic scale, but their microscopic origin is often unclear. Whereas a wide variety of hysteresis shapes and behavior has been measured in different types of magnetic materials, only a few theories of hysteresis exist [3, 4]. In the case of F thin films with perpendicular anisotropy, hysteresis effects may occur along the magnetization easy axis, perpendicular to the film. It is established that in such films, microscopic magnetic domains tend to form, often in the shape of serpentine stripes [5]. Understanding the morphological changes of these domains throughout the magnetization cycle is essential to explain the occurrence of hysteresis for these perpendicular F films. While most magnetometry tools provide macroscopic information, only a few sophisticated tools can probe the microscopic magnetic state. Among them, magnetic imaging techniques, such as magnetic force microscopy (MFM) and transmission x-ray microscopy, allow visualizing magnetic domains in limited regions of the material [6, 7]. The nucleation, growth, propagation and saturation of domains can usually be observed via these imaging techniques, but the relationship between the microscopic domain morphology and the shape of the magnetization loop is usually very complicated, as illustrated in various studies combining magnetometry and imaging measurements [8, 9].

In this paper, we show a complementary statistical way to study the microscopic origin of magnetic hysteresis, via magnetic domain memory (MDM), with a unique x-ray magnetic speckle correlation approach. MDM is the ability of magnetic domain patterns to retrieve their spatial morphology at specific points on the magnetization loop, after one or more cycles. Here, we perform a field mapping of MDM. The resulting maps, compared to the magnetization loop, unveil new insight into the microscopic origin of the hysteresis, the morphological changes in magnetic domains and the amount of reversibility at various stages of the reversal.

The observation of disorder-induced MDM in Co/Pt thin films has revealed the impact of film roughness on the behavior of domains at the microscopic scale and the occurrence of
partial memory at nucleation [10]. In this paper, we show memory effects occurring throughout the entire magnetization loop by exploiting another type of MDM, this one induced by exchange couplings between a Co/Pd F layer and an IrMn antiferromagnetic (AF) layer [11]. This exchange-bias MDM can be increased from zero to full 100% strength in different regions of the hysteresis loop by tuning the coupling parameters. We show here a field mapping of MDM when the film is brought into a zero-field-cooled (ZFC) state, where MDM is greatly enhanced [12]. The inter-field maps of MDM reveal microscopic changes in the domain morphology in the presence of exchange couplings.

2. Magnetic characterization via magnetometry and magnetic force microscopy

Our exchange bias thin films are made of a stack of F [Co(4 Å)/Pd(7 Å)]12 multilayers, interleaved with 24 Å thick AF IrMn layers. The stacking includes four repeats, adding up to a total of 48 Co/Pd bilayers. This layer structure was chosen to tune the magnetic properties, in particular the domain size and structure, the exchange couplings and the intensity of the magnetic x-ray scattering signal. Co/Pd multilayers are chosen to provide a strong perpendicular magnetic anisotropy resulting from the Co–Pd interfacial magnetic anisotropy. To achieve well-defined stripe domains, we need F films whose thicknesses are above a characteristic domain size, defined by the ratio of the domain wall to shape anisotropy energies. For Co/Pd multilayers, this requires at least 20 Co/Pd repeats, but better domains are observed for about 50 repeats [13]. For thinner films, the domain size expands exponentially with decreasing film thickness and the x-ray scattering signal will quickly move to a range not accessible to our experiments. The 50 repeats in the multilayer also optimize the x-ray scattering intensity. If a 50 Co/Pd multilayer is coupled to a single IrMn layer, the biasing is quite small and is not appropriate for the current experiments. To enhance the biasing, we interleaved IrMn layers after every 12 Co/Pd repeats to enhance the exchange biasing. The stacking does not alter the properties of the (F/AF) bilayer; it just increases the amount of coupling. The dipolar fields couple the magnetization in adjacent Co/Pd stacks, yielding structure that forms well-defined stripes and strong exchange biasing [14].

A view of the magnetic domain pattern, measured by MFM on a 10 µm scale, is shown in figure 1(a). This image, collected at room temperature (∼300 K) and zero field, shows the typical domain pattern forming in our film with labyrinthine stripes, with an average width of about 200 nm. A bulk measurement of the magnetization via vibrating sample magnetometry (VSM) is shown in figure 1(b). At a temperature slightly above room temperature (320 K), the magnetization loop exhibits some hysteresis with a coercivity $H_c \sim 220$ Oe. The loop is actually narrower at its center and wider at its top and bottom, toward saturation. The gap between saturation and nucleation points is about ∼1800 Oe. This loop shape suggests different stages in the reversal process of the magnetic domains: a quick nucleation and domain propagation, followed by a slow domain expansion and saturation, as observed in various imaging studies on other materials [8, 15]. When our film is cooled down below the blocking temperature ($T_B \sim 300$ K), exchange couplings occur and the hysteresis shape is significantly changed. In our study, the film is cooled in a ZFC state after an initial demagnetization at 400 K. The magnetization loop measured at 20 K does not exhibit any macroscopic bias, and is still centered about zero. However, it differs in shape from the one at 320 K. The coercive field does not change much—it only increases to 275 Oe—but the curve’s inflection is drastically accentuated, and the gap between saturation and nucleation point increases to ∼3600 Oe. We learn, from our
Figure 1. (a) 10 µm MFM image of the magnetic domain pattern in our [Co/Pd]IrMn film, in the remanent state ($H = 0$), at room temperature ($T \sim 300$ K). (b) Magnetization loop at 320 K and in ZFC state at 20 K, measured by VSM. (c) Series of MFM images taken at various points along the descending branch of the magnetization loop at 300 K (points are indicated with dots on the loop). (d) Series of CXRMS patterns measured at 20 K in ZFC state, at selected field values along the descending branch. The dark oblong shape at the center of the images is the shadow of a blocker. The first image, taken at positive saturation, shows pure charge scattering and was used for background subtraction in the following images.

inter-field MDM measurements discussed below, that this inflection effect is accompanied by a large amount of reversibility in the domain reversal.

Local views of the domain morphology in our [Co/Pd]IrMn films throughout the nucleation phase are shown in figure 1(c). These 10 µm MFM images collected on the descending branch, from nucleation down to the remanent point, indicate that domains first nucleate as sparse bubbles, then propagate quickly and branch out, so as to fill the space and create a maze. Beyond remanence, the domains continue to propagate, connect and stay in a maze-like pattern throughout the coercive region and up to higher fields. The domains expand in overall length
and width, and eventually cover the entire film as saturation is approached. These MFM images provide good insight into the domain morphology at room temperature. In order to study the magnetic domain pattern in the ZFC state, we need to make measurements at low temperature and also be able to reach higher \textit{in situ} field values. Our instrument, like most MFM instruments, does not allow us to image at low temperature. We can only visualize the domain patterns at room temperature, prior to the cooling experiment. When cooling the sample down to 20 K, in the absence of field (remanent state), we expect that the morphology of the domains and their length scale remain essentially the same as at room temperature. In fact, our prior studies of spatial dependence in the correlation patterns [12] have indicated that the correlation length scales remain the same at all temperatures. Only the shape and size of the net magnetization loop change when cooling the sample at low temperature, but the length scale for the domain pattern is unchanged. Coherent x-ray resonant magnetic scattering (CXRMS) is a perfect complementary tool to MFM as it offers the possibility to study magnetic configurations at low temperature and with \textit{in situ} magnetic field, which can be reversed in direction, so as to complete a full hysteresis loop. Furthermore, the CXRMS technique provides statistically quantitative information about the domain morphology over larger areas of the film.

3. Magnetic domain memory probed via x-ray speckle correlation

We found that the film exhibits strong MDM in the coercive region when cooled in a ZFC state [11, 12]. The occurrence of strong MDM suggests that the magnetic pattern imprinted in the AF layer via the uncompensated spins plays the role of a template for the reversal of the magnetic domains in the F layer. MDM was first evaluated by comparing magnetic patterns at the same field value after field cycling. This type of MDM is also called return-point memory (RPM). For our [Co/Pd]IrMn films, in the ZFC state, the RPM follows a low–high–low behavior along each branch of the magnetization loop. It is typically low (under 20%) at nucleation and then sharply increases toward a maximum (above 90%) around a field $H^* = 1600$ Oe, with a plateau effect extending almost all the way to saturation, at around 3500 Oe, where the RPM decreases again down to 20%. This result helps quantify the ability of the magnetic domains in the [Co/Pd] multilayer to retrieve their topology after field cycling. However, the RPM does not provide direct information on how the magnetic patterns correlate between each other at different points of the magnetization loop. With our inter-field correlation maps discussed in this paper, we provide a complete view of MDM, beyond the case of RPM, by comparing magnetic patterns at all field values throughout the entire magnetization cycle.

3.1. Methods

We have measured the amount of domain memory using CXRMS and probing the magnetic domain configuration throughout the magnetization process. Examples of CXRMS patterns measured on our [Co/Pd]IrMn multilayer are shown in figure 1(d). Magnetic speckle patterns, collected at different points of the magnetization loop after successive cycles, are cross-correlated to statistically estimate the amount of similarity between the actual domain patterns [12]. Our CXRMS measurements were carried out at beamline 12.0.2 of the Advanced Light Source, Lawrence Berkeley National Laboratory [16]. The x-ray light was tuned to the Co L$_3$ edge, around 778 eV, and its polarization was set to be linear in-plane. The sample was mounted in transmission geometry and coherent magnetic scattering patterns were collected.
onto a CCD camera, placed downstream at about 1 m from the sample. As demonstrated by Hannon and Trammel, as well as Blume in the 1980s [17, 18] and Hill and McMorrow in 1996 [19], the scattering factor for the scattered x-rays for an electric dipolar transition (here 2p–3d transition), can be written to first order as

$$f_{\text{res}} \sim F_0 (\vec{e} \cdot \vec{e}^\prime) + i F_1 (\vec{e} \times \vec{e}^\prime) \cdot \vec{M},$$

where $\vec{e}$ and $\vec{e}^\prime$ represent the polarization vectors of incoming and scattered light, respectively, and $\vec{M}$ is the magnetization in the material. The factors $F_0$ and $F_1$ are spectroscopic terms, accounting for the electronic configuration of the given chemical magnetic element. $F_0$ and $F_1$ include absorption factors, leading to resonance effects when the x-ray energy is tuned to absorption edges. $F_0$ refers to the zero order (also known as the Thomson scattering factor). $F_1$ refers to the first order with respect to magnetization $\vec{M}$ (higher quadratic order is not included here, as it is negligible). The dependence on $\vec{M}$ in the second term leads to the magneto-optical contrast, enhanced at resonance. This effect, first observed experimentally in 1981 in magnetite and hematite compounds [20], has been later observed for transition metals and rare earth elements at different edges [21]. We apply it here to the Co element, in our F layers. As highlighted by the cross-product, the direction of the polarization in our setup is optimized for our perpendicular magnetic sample. Also, our magnetic field is applied in situ perpendicular to the sample surface.

Due to the coherence of the x-ray light, our CXRMS patterns exhibit intensity fluctuations in space, also called speckles. Unique information is contained in these speckle patterns, as they provide a fingerprint of the local morphology of the magnetic domains in the material. Our speckle cross-correlation technique exploits this information as a way to estimate the amount of MDM, or the similarity between domain patterns, throughout the magnetization process and as we cycle the field. In practice, we calculate the degree of correlation between two speckle patterns $I_1$ and $I_2$, collected at different field values $H_1$ and $H_2$, by performing the following operation:

$$\rho = \frac{I_1 \times I_2}{\sqrt{(I_1 \times I_1)(I_2 \times I_2)}}, \quad \text{where } 0 \leq \rho \leq 1,$$

where $\times$ represents the correlation operation between two images. With our cross-correlation procedure finely optimized for our scattering patterns [22], we have measured the degree of correlation $\rho$, for all possible combinations of pairs of images, collected at fields $(H_1, H_2)$, and thus generated maps, as in figure 2, where $\rho(H_1 \times H_2)$ is plotted in color (red being high and blue being low). Because of the hysteresis effect in the magnetization loop, ascending and descending branches are treated separately, and referred to by the letters $A$ and $D$, respectively. The maps $\rho(H_1 \times H_2)$ calculated between same-side branches $A \times A$ are shown on the top, and the maps calculated between opposite-side branches $A \times D$ are shown on the bottom. Computing $A \times D$ or $D \times A$ gives exactly the same result because of the symmetry of the operation. Also, we have computed $D \times D$ maps, and found that besides local deviations due to experimental noise, the $D \times D$ maps are identical to $A \times A$ maps (apart from the fact that the field is reversed). We therefore chose to only display $A \times A$ maps in this paper. A distinction nevertheless needs to be made on the number of loops separating the selected branches. For example, the left side in figure 2 shows correlation maps that were computed within the same cycle and the right side shows correlations computed between branches separated by at least one complete cycle. Other maps measuring the correlation after two and three cycles were computed but are not represented. The signal plotted in each of the maps is the average of the results on all
3.2. Inter-correlation maps throughout the hysteresis cycle in the zero-field-cooled state

A selection of CXRMS patterns collected at different points along the magnetization cycle at room temperature is shown in figure 1(d). The first image was collected in the positively saturated state, where most of the scattering signal is actually not magnetic but charge scattering located around the transmitted beam. This image was subtracted from all subsequent images, to extract the pure magnetic signal. In following images, the scattering pattern evolves from a disc shape near nucleation to a ring shape whose radius is maximal in the coercive region where $M \sim 0$, and the domain period has reached its minimum $\sim 400$ nm. Our cross-correlation technique, described in section 3.1, uses the scattering patterns to quantify the amount of MDM. A selected set of the resulting inter-field maps $\rho(H_1 \times H_2)$ is shown in figure 2.

At first glance, all the inter-field $\rho(H_1 \times H_2)$ maps exhibit a strong signal (beyond 90%) at their center, with a large plateau centered about $|H_1| = |H_2| = H^* \sim 1600$ Oe. The signal
fades away down to 10–20% at the border of the maps. Map 2(a) exhibits a ridge along its diagonal. For this map, the correlation is performed within the same physical branch of the magnetization loop; therefore the signal along the diagonal \((H_1 = H_2)\) is obtained by correlating the same physical images (auto-correlation) and consequently \(\rho = 1\). On the other maps, the two correlated images are physically distinct everywhere since they come from different magnetization cycles. The signal along the diagonal on the \(A \times A\) map, after a minimum of one cycle, corresponds to RPM [23]. Similarly, the signal along the diagonal on the \(A \times D\) map corresponds to the so-called conjugate-point-memory (CPM). With our generalized inter-field correlation maps, we look beyond RPM and CPM, at morphological changes in the magnetic domain pattern throughout the reversal.

The symmetry between \(A \times A\) maps and \(D \times D\) maps, mentioned above, is consistent with VSM bulk magnetization measurements in figure 1, indicating a symmetrical magnetization loop both at room \(T\) and low \(T\) in ZFC state, due to the absence of net bias. The symmetry is maintained in the correlation maps because the film was previously brought to a demagnetized state and no field was applied during the cooling, so the global morphology of the magnetic domains with the ‘up’ and ‘down’ spins is equivalent. Although the local configurations of the up and down domains may differ at the nanometric scale, their averaged configuration on a larger scale (few microns) is equivalent. In our CXRMS experiment, we probe the sample on a large scale \((\sim 100 \mu m)\); thus we measure the statistically averaged domain configuration.

A noticeable feature, visible in all the correlation maps, is the impressively large extent of the plateau. The RPM signal, diagonally sliced from the map 2(c), and shown in figure 3(a), follows low–high–low behavior centered on \(H^*\). With the 2D maps, we see that the plateau effect occurs not only along the diagonal (where \(H_1 = H_2\)) but also off diagonal (where \(H_2 \neq H_1\)). In figure 3(a), a horizontal slice through the map 2(c), at \(H_2 = H^*\), is plotted against the diagonal RPM slice. The two slices superpose perfectly, indicating that the magnetic domain morphology outside \(H^*\) remains essentially unchanged and similar to the pattern at \(H^*\). For

![Figure 3](http://www.njp.org/)

**Figure 3.** Comparison between horizontal slices at fixed \(H_2\) and diagonal slices \((H_2 = H_1)\) from the maps in figure 2. (a) RPM slice and 1600 Oe slice and from the \(A \times A\) map, after one cycle. The magnetic moment \(M(H)\) is plotted for reference; (b) CPM slice and 1600 Oe slice from the \(A \times D\) map after one cycle, compared with the RPM slice.
example, the domain patterns at 800 and 2800 Oe both correlate well with the central pattern at $H^* \sim 1600$ Oe. Surprisingly, the net magnetization changes quite significantly in that field range ($M$ triples when the field varies from $H = 800$ to 2800 Oe), as there is an increasing imbalance between the amounts of spins up and spins down. However, the morphology of the domain remains statistically the same. This phenomenon is partly due to the natural ability of domains to breathe (expand or shrink) without changing the global morphology of the pattern at long range. This effect is particularly visible in our XRMS images, because they essentially focus on the ring, which is the first-order scattering signal from the periodic pattern, and which relates to long-range order in real space. The fact that the memory plateau extends here on such a large field range is mostly due to exchange couplings. The domain configuration in the F layer is locked into a template, which was imprinted in the AF layer during the ZFC process. Due to the nature of exchange couplings at the interface between the layers, we assume that the exact same domain pattern was imprinted from the F [Co/Pd] multilayer down to the AF IrMn layer during the cooling process. This assumption is supported by studies of the in-depth magnetic profile at the interface between the F layer and the uncompensated spins in the AF layer in other exchange-bias systems [24]. At low temperature, this domain pattern imprinted in the AF layer behaves, in our case, as a template for the F layer, as depicted in our earlier paper [11]. The imprinted template allows the F domains to breathe without losing their global morphology when applying extended field variations. This process is highly reversible, as the domain pattern can easily retrieve the imprinted template while varying the field around $H^*$.

Correlations on opposite sides of the magnetization loop provide further information. For instance, figure 3(b) compares the horizontal cut taken at fixed field ($H_2 = H^*$) with the CPM diagonal cut on the $A \times D$ map 2(d). We note the similarity between the horizontal cut at $H^*$ and the RPM diagonal cut from map 2(c): they overlap almost perfectly. The signal along the CPM diagonal is, however, slightly narrower than the horizontal slice. This discrepancy reveals local differences between the ‘up’ and ‘down’ domain configurations. Because each $A \times D$ map represents the correlation between an ascending and a descending branch, it compares the configurations of spins in opposite directions (spins ‘up’ and spins ‘down’). The $A \times A$ maps, on the other hand, compare the configurations of spins aligned in the same direction. By comparing slices through $A \times D$ maps and slices through $A \times A$, we can therefore get some statistical information about the relative discrepancy between the up and down domain configurations in the template. The fact that the $A \times D$ slices are slightly narrower (in field range) than the $A \times A$ slices suggests that the domain morphology of the template is not completely symmetrical in the up and down directions. The difference between the $A \times D$ and $A \times A$ slices is actually fairly small, so the symmetry between the up and down domains configurations is statistically fairly good. Only when a field is applied, deviating significantly from the central field region, does a slight difference appear. This suggests some minor local differences between the up and down domain morphologies, only visible when domains in one direction expand at the expense of domains in the opposite direction.

The $\rho(H_1 \times H_2)$ maps unveil what happens far away from the central field $H^*$, closer to nucleation and saturation points. Figure 4 shows a selection of horizontal cuts throughout the $A \times A$ and $A \times D$ maps from figure 2. These slices are measured at various field values $H_2$, from $H_2 = 0$ (shortly after nucleation) to $H_2 = 3200$ Oe (shortly before saturation). To help understand what these slices represent, a sketch has been added next to the $A \times A$ slices and next to the $A \times D$ slices, respectively, showing what points along the magnetization loop are being correlated in each case. For all the cuts, the memory stays steady on a remarkably large
range of fields, but it eventually decreases toward the extremities (nucleation and saturation), rather sharply, reaching down to $\sim 10\%$. Interestingly, the magnetization loop exhibits a similar sharpness on the nucleation side but not on the saturation side. The sharp loss of memory on both sides is a strong indication of irreversibility in the morphological change of the domains, both at nucleation and at saturation. The domain patterns at nucleation and at saturation are quite different from the imprinted pattern, and quite different from each other. At these extremities, domain nucleation and reversal are essentially driven by irreversible random processes, and not by exchange couplings.

Interesting asymmetries are revealed when comparing the $A \times A$ slices and $D \times D$ slices. We find that the $A \times A$ slices are somewhat asymmetric, whereas the $A \times D$ slices are symmetrical (with respect to field $H_1$ variation away from the central region $H^*\)$. For instance, the slices for $A \times A$ same-cycle in figure 4(a) show highly asymmetric behavior when $H_2 \neq H^*$. Because the correlation is performed on the same branch, $\rho$ gets significantly higher at one end, reaching up to 100% whenever $H_1 = H_2$. This asymmetry observed on same-branch correlations does somewhat persist after one cycle. For example, cuts at $H_2 = 0$ and 3200 Oe in figure 4(b) are asymmetric, and the correlation increases whenever $H_1 \sim H_2$. In comparison, the cuts through $A \times D$ maps (figures 4(d) and (e)) present perfectly symmetrical behavior, with a steady value at $\sim 50\%$ in the central field region, for $H_2 = 0$ and 3200 Oe. These differences
in symmetry between \( A \times A \) and \( A \times D \) confirm the presence of local discrepancies between the ‘up’ and ‘down’ domain configurations in the imprinted template, as suggested in the previous discussion related to figure 3(b). In figure 4, we look at more slices throughout the \( A \times A \) and \( A \times D \) maps to get a more general view of the magnetic behavior. Overall, the correlation between opposite branches is fairly good (\( A \times D \) cuts). This is due to the exchange couplings between the F domain pattern and the AF imprinted pattern. The domain reversal is driven by the same template for both branches, and the template is globally symmetrical in terms of up and down domain configurations. However, we can observe an extra correlation for same-side branches, whenever the two correlated points are about the same field value (thus giving an asymmetric shape to the \( A \times A \) slices). This phenomenon is due to local discrepancies between the up and down configurations, resulting in slightly different domain morphologies when domains in one direction start to expand, compared to when domains in the opposite direction start to expand. On a given branch, domains always expand and propagate the same way, but they do not expand exactly the same way on the opposite branch.

3.3. Comparison below and above the blocking point

The occurrence of high memory effects, discussed above, was observed in a ZFC state at 20 K, well below the blocking temperature \( T_B \sim 300 \) K. We have carried out measurements at higher temperatures, in particular above \( T_B \), for comparison. A set of correlation maps measured at 335 K, slightly above \( T_B \), is displayed in figure 5. The contrast with previous ZFC maps is striking. Excluding the first \( A \times A \) map (figure 5(a)), all the other maps clearly indicate an almost complete loss of memory, with a correlation signal down to 20% and below. Also, the shape of the correlation signal is not conserved through field cycling (map 5(d) is very different from map 5(b)), preventing us from relating it to the existence of true memory. Given experimental uncertainties inherent to the measurement, we consider this amount of correlation to be very low, practically insignificant. In contrast, a very high correlation signal appears in the first \( A \times A \) map. This effect is actually expected for any same-cycle \( A \times A \) correlation map, because speckle patterns, and therefore domain patterns, are here compared within the same physical branch of the magnetization loop. The signal along the diagonal is again equal to 100% because images are correlated with themselves, but the signal stays also pretty high in the vicinity of the diagonal. While domains nucleate, form, propagate, expand and then gradually reverse toward saturation, and given their ability to breathe, it is expected that there will be a natural similarity between patterns at subsequent field values. The high level of correlation within one physical branch of the magnetization loop naturally occurs as the reversal progresses, but this does not account for any type of memory. The true measure of memory only starts after the material has been saturated at least once—so the domain pattern has supposedly been completely erased—as is the case for all the other maps. These following maps show that, as soon as saturation has been reached at least once, correlation drastically drops, memory is essentially lost.

Differences between correlation maps below \( T_B \) (at 20 K) and above \( T_B \) (at 335 K) can be quantified by comparing slices through the respective maps. Figure 6 shows horizontal \( H \)-slices at the central field values (\( H = 1600 \) Oe for the 20 K data and \( H = 960 \) Oe for 335 K data)—the central field values are shifted because the magnetization loop changes in shape and size, as seen in figure 1). The comparison for the \( A \times A \) map is particularly interesting. For both temperatures, the correlation reaches 100% at the central field value. However, the shape of the slice is very
Figure 5. Correlation maps measured above $T_B$, at $T = 335$ K. (a) $A \times A$ map within the same branch, (b) $A \times D$ map within the same cycle, (c) $A \times A$ map after one full cycle, (d) $A \times D$ map after one full cycle.

different. The shape at 335 K is quite sharp, almost triangular, while the shape at 20 K in ZFC state is more rounded, and more extended with the plateau effect. These differences, which somewhat mimic the change in the shape of the magnetization loop, result from the occurrence of exchange couplings at low temperature. In the ZFC state, domains nucleate, propagate, and once they start approaching the imprinted domain pattern, they tend to stick to that pattern, for an extended range of field values. In other words, there is an extra correlation added to the natural correlation visible in the $A \times A$ same-branch map, due to exchange couplings. The comparison slices for the other maps indicate that the degree of correlation is drastically decreased at high temperature. The residual signal (below 20%) could reflect very weak memory effects induced by some defects and slight roughness of the film. However, the fact that the correlation is not particularly higher at nucleation (which is characteristic of disorder-induced MDM) and the fact that it does not persist through field cycling indicate that memory effects are almost absent at high $T$. MDM is essentially lost once the material is heated above $T_B$.

This comparison between the 335 K and the ZFC 20 K results clearly confirms that the FCo/Pd multilayer does not carry any significant MDM on its own. Interestingly, this behavior differs significantly from defect-induced memory observed in some Co/Pt multilayers, due to roughness [10]. In our case, the absence of memory at high temperature indicates that the film is very smooth, and the effects from roughness are negligible. Only when the material
is cooled down below blocking temperature and exchange couplings are occurring, a strong MDM appears in the central region of the magnetization loop. This behavior is strong evidence that memory is, here, solely induced by exchange couplings and nothing else. Furthermore, the amount of measured MDM is very high on a large extent of field. This unprecedented result exceeds the amount of memory observed so far in any other type of material. This property is unique to exchange-bias films.

3.4. Dependence on temperature

To further study how MDM varies with temperature, we have measured the correlation maps at different temperatures while heating the sample from the ZFC state at 15 K back up to high temperature above the blocking point. Figure 7 shows a selection of $A \times A$ and $A \times D$ maps measured at 30, 60, 120, 175, 220 and 335 K, successively. A display of the complete set of measured maps (including different cycle separations, for each temperature) can be found online. These maps indicate that the amount of MDM stays relatively high for all temperatures below the blocking point and then collapses above the blocking point. Also the shape of the plateau is maintained for all temperatures, extending almost as widely at 220 K as at 20 K.

The dependence on temperature can be measured in various ways, using the correlation maps, in connection with their associated magnetization loops. Figure 8(a) shows a set of magnetization loops measured at different temperatures, heating up from the ZFC state. The shape of the magnetization loop is globally maintained while increasing the temperature. It stays
Figure 7. Correlation maps at selected temperatures, heating from the ZFC state up to 335 K.

symmetrical as no net bias was applied during ZFC. Interestingly, the first part (from nucleation to coercive point) of either the ascending or the descending magnetization branch does not significantly change while temperature increases. Only the second part of the branch (from the coercive point to saturation) is modified. The opening of the loop gradually decreases while temperature increases. This effect can be quantified by measuring the gap between saturation field $H_s$ and nucleation field $H_n$ for each loop. This difference ($H_s - H_n$) is plotted as a function of temperature in figure 8(b). The gap gradually decreases with temperature, first very slowly at low temperature and then more rapidly while traversing the blocking transition. Figure 8(c)
Figure 8. Temperature dependence. (a) Set of magnetization loops collected at different temperatures in ZFC state, heating up from 20 K back to 400 K. (b) Gap between saturation and nucleation points, versus temperature. (c), (d) Comparison slices throughout the maps at different temperatures (shown in figure 7): (c) from the $A \times A$ map; (d) from the $A \times D$ map.

shows, in comparison, horizontal slices through the $A \times A$ and $A \times D$ correlation maps at different temperatures. The slices extracted from the $A \times A$ maps suggest that the MDM stays rather high at all temperatures below $T_B$. The maximum correlation varies from about 90% at 20 K down to 77% at 220 K. Given that experimental uncertainties can lead to variations as high as ±5% or more, from one series of measurements to another at a given temperature, the result suggests that the amount of memory on same-side branches is globally maintained throughout heating from ZFC state, with a slight loss of the order of 10–15% between 20 and 220 K. On the other hand, the $H$-slices through $A \times D$ maps, shown in figure 8(d), indicate a greater decrease with temperature. The maximum correlation values vary from 89% at 20 K down to 65% at 220 K. Given uncertainties, this corresponds to a decrease of the order of 20–30%. This quicker decrease in opposite-branches correlation might arise from thermal fluctuation effects. It was already observed that in the ZFC state at 20 K, the $A \times D$ signal was slightly lower and less extended than the $A \times A$ signal. At higher temperature, the difference is more drastic, suggesting that thermal fluctuations start to weigh in, so as to diminish the effect induced by exchange couplings. The ‘up’ and ‘down’ domain patterns still tend to match in essence the imprinted pattern, but at higher temperature, they are unequally losing their amount of ‘matching’. In other words, local discrepancies between the ‘up’ and ‘down’ patterns occur at various
locations throughout the film, and the number of these locations increases when temperature increases.

The measurement at 335 K shows a drastic loss of memory, as the sample has been heated above the blocking temperature $T_B$. The loss of memory appears to be significantly more drastic than the change in the shape of the magnetization loop, which is more subtle and smooth (figure 8(b)). The smooth change in the magnetization loop actually induces a shift in the MDM plateau toward lower field values, as the temperature increases. In addition to the shift, the correlation maps reveal an abrupt loss of domain memory, which could not been seen from the bulk magnetization measurements. Above $T_B$, domain reversal is largely random throughout the whole hysteresis loop. Below $T_B$, strong memory is induced by exchange coupling, and a large part of the reversal is driven deterministically by the imprinted template. Thermal fluctuations play a minor role compared to exchange couplings, as they only slightly alter the strength of domain memory. In summary, we learn that both thermal fluctuations and structural disorder have here minor or negligible impact on memory. MDM is essentially induced by exchange couplings.

4. Microscopic magnetic behavior and reversal throughout the hysteresis cycle

Overall, this magnetic correlation study of exchange biased films in ZFC state reveals how magnetic domains adopt different behavior in the presence or absence of exchange couplings. The correlation maps measured above $T_B$ suggest a largely random, uncorrelated process in the absence of exchange couplings. The correlation maps in the ZFC state below $T_B$ reveal the occurrence of well-correlated domain reversal processes, via the imprinting of a magnetic template. Because the correlation maps are related to the local domain morphology, they help to further understand the reversal processes and morphological changes of magnetic domain throughout the hysteresis loop. In particular, correlation maps suggest that the magnetic pattern goes through successive reversal phases, with different degrees of reversibility. It appears that at all temperatures, above $T_B$ as well as below $T_B$, the domain reversal passes through three main phases: (i) a sharp irreversible nucleation, (ii) a central reversible phase and (iii) a sharp irreversible saturation. The essential difference between the ZFC state and the high $T$ state is that the second reversible phase is drastically expanded for the ZFC state, because of exchange couplings. An illustration of these phases (for the ZFC state) is pictured in figure 9. In the first phase, at nucleation, domains nucleate rather randomly and irreversibly but quickly converge toward a ‘nucleated pattern’ where exchange couplings with the imprinted AF pattern start to weigh in and memory reaches 50%. The second phase consists of a more reversible domain growth, to match the underlying AF template; memory quickly exceeds 80%, peaking above 90%. In the third phase, domains have grown and breathed to the extent of breaking the reversible limit, and memory is eventually lost. The discussed asymmetry in the $A \times A$ cuts suggests that the ‘nucleated pattern’ (N) and the ‘pre-saturation pattern’ (S) are not equivalent in morphology. This observation is qualitatively supported by MFM images at room temperature, where domain morphologies after nucleation and before saturation are visibly different. It appears to be also the case for the ZFC state, where the reversal is under the influence of exchange couplings. However, contrary to the behavior at room temperature, where the reversal is more random and irreversible, the pattern in ZFC state quickly converges toward the reference pattern, and once the irreversible nucleation stage is passed, the reversal becomes mostly reversible, because of the exchange couplings. The correlation results also reveal that domains

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Figure 9. Illustrative phase diagram showing the regions of strongest MDM throughout the hysteresis loop, for the ZFC state at 20 K. \( \rho \) refers to the amount of correlation. (N+), (N−), (S+) and (S−) refer to the ‘up-nucleation’ pattern, ‘down-nucleation’ pattern, ‘up-pre-saturation’ pattern and ‘down-pre-saturation’ pattern, respectively.

tend to partly retrieve the same nucleated pattern and the same pre-saturation pattern when cycling the field and looking at the same-side branches (spins in the same direction), with correlation reaching up to 65%. When comparing opposite branches (spins in the opposite direction), the domains have less ability to retrieve the same nucleated pattern or the same pre-saturation pattern, because of local asymmetries between the ‘up’ and ‘down’ domains in the imprinted template. This leads to morphological differences between the ‘up-nucleation’ and ‘down-nucleation’ patterns, and between the ‘up-pre-saturation’ and ‘down-pre-saturation’ patterns, represented by (N+), (N−), (S+) and (S−), respectively, in figure 9. Despite the discrepancies between those four patterns, the memory is found to be equally high, around 50%, when cross-correlating these four regions, suggesting that exchange couplings still impact the morphology of the magnetic domains when approaching these extremities, and the impact is statistically equal in the up and down directions. The limitation at 50% reflects partial randomness and irreversibility in the nucleating and saturating processes.

5. Conclusion

In conclusion, our inter-field CXRMS correlation study of [Co/Pd]/IrMn films in the ZFC state brings more insight into the occurrence of memory effects throughout the hysteresis loop, and helps connect macroscopic hysteretic behavior to domain reversal at the microscopic scale. The amount of memory has here been quantified for all combinations of field values, on same-side and on opposite-side branches of the magnetization loop beyond the standard RPM
and CPM measurements. We have also studied the dependence of these correlation maps on temperature, while heating the material from the ZFC state back up to high temperatures, above the blocking point $T_B$. We have observed a slight decrease in the amount of memory when temperature increases below $T_B$, more pronounced for opposite-side branches than for same-side branches. But overall, a relatively strong exchange coupling-induced memory occurs at all temperatures below $T_B$, until it completely collapses above $T_B$. In the ZFC state, the generated maps $\rho(H_1 \times H_2)$ provide information about the domain reversal in the F layer when under the influence of exchange couplings. We learn that while nucleation and saturation phases remain highly irreversible and random, the domain reversal becomes reversible on a very wide range of fields centered about $H^*$ due to the presence of the AF template. This behavior greatly differs from the behavior at room temperature, in the absence of exchange couplings, where the domain reversal is mostly random. We also learn from the $\rho(H_1 \times H_2)$ maps that the ‘up’ and ‘down’ domain morphologies in the ZFC state are statistically equivalent on a large scale (100 $\mu$m) but local discrepancies at the nanometric scale slightly lower the amount of domain memory between opposite branches, compared to same-side branches. Finally, making a connection between the macroscopic and microscopic observations, we found that the change in the shape of the magnetization loop, i.e. the ‘opening’ of the curve around $H^*$, results from the enhancement of reversibility in the reversal process and not from irreversibility. This inter-field correlation, performed here in a ZFC state, is an excellent tool to study the effects of biasing in field-cooled states. It can also be applied to a variety of hysteretic systems exhibiting memory and help connect macroscopic behavior to the microscopic origins.

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