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Positive exchange bias in epitaxial permalloy/MgO integrated with Si (100)

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ABSTRACT

In magnetic random access memory (MRAM) devices, soft magnetic thin film elements such as permalloy (Py) are used as unit cells of information. The epitaxial integration of these elements with the technologically important substrate Si (100) and a thorough understanding of their magnetic properties are critical for CMOS-based magnetic devices. We report on the epitaxial growth of $\text{Ni}_{82.5}\text{Fe}_{17.5}$ (permalloy, Py) on Si (100) using a TiN/MgO buffer layer. Initial stages of growth are characterized by the formation of discrete islands that gradually merge into a continuous film as deposition times are extended. Interestingly, we find that the magnetic features of Py films in early stages of island coalescence are distinctly different from the films formed initially (discrete islands) and after extended deposition times (narrow distribution of equiaxed granular films). Isothermal in-plane and out-of-plane magnetic measurements performed on these transitional films show highly anisotropic magnetic behavior with an easy magnetization axis lying in the plane of the film. Importantly, when this sample is zero-field cooled, a positive exchange bias and vertical loop shift are observed, unusual for a soft ferromagnet like Py. Repeated field cycling and hysteresis loops up to the fields of 7T produced reproducible hysteresis loops indicating the existence of strongly pinned spin configurations. Classical interface related exchange bias models cannot explain the observed magnetic features of the transitional Py films. We believe that the anomalous magnetic behavior of such Py films may be explained by considering the highly irregular morphology that develops at intermediate growth times that are possibly also undergoing a transition from Bloch to Neel domain wall structures as a function of Py island size. This study broadens the current understanding of magnetic properties of Py thin layers for technological applications in magneto-electronic devices, integrated with Si (100).

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Motivated by the proposed racetrack memory patented [1] by Parkin et al, a great deal of research activity has been devoted to Permalloy (Py) magnetic nanostructures for high frequency applications. Py usually refers to $\text{Ni}_{1-x}\text{Fe}_x$ alloys that have a vanishingly small magnetocrystalline anisotropy and magnetostriction but extremely large magnetic permeability. These unique properties make Py one of the most important soft magnets for a variety of applications, such as free layers in spin-valve magnetic-reading heads [2,3]. The Py/MgO combination has been widely used in magnetic tunnel junctions (MTJs) devices. More recently, Py is being extensively used [4,5] to demonstrate electric field control of ferromagnetism (FM) in multiferroic based devices. Hence,

exploring and understanding the magnetic properties of Py when interfaced with magnetic/non-magnetic oxide materials is of primary importance.

To date, epitaxial Py layers have been deposited [6] on MgO bulk substrates using molecular beam epitaxy, e-beam evaporation and magnetron sputtering deposition techniques. However, MgO substrates are unsuitable from a viewpoint of practical applications in the magnetic recording industry because of its high single-crystal cost, limited wafer size, hygroscopic nature, and inferior mechanical properties. The unique feature in the present study stems from the usage of single crystal Si (100) substrates. Silicon is inexpensive and readily available, but most importantly, by using Si (100) we can achieve integration with present day microelectronic devices.

The magnetic properties of Py have been extensively reported for the case where it is interfaced with anti-ferromagnetic (AFM)

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materials such as hematite [7], CoO [8], IrMn [9], FeMn [10] and NiO [11]. In addition, some preliminary studies have been reported [6] on the deposition of Py on MgO bulk wafers. However, little or no attention has been devoted to studying the magnetic properties of this epitaxial heterostructure, particularly as a function of Py film thickness (island size) and morphology when it is integrated with the technologically important substrate Si (100), which motivates the current work.

The performance of magnetic devices strongly depends on the thin film morphology and microstructure. In this work, we have investigated the magnetic characteristics of Py under various thermomagnetic conditions as a function of varying deposition times. The most important finding of the present investigation is that for the Py layer grown at intermediate times where the discrete islands are just beginning to merge into continuous films the magnetic behavior indicates the existence of strongly pinned spin configurations and unsaturated hysteresis loops. A profound positive exchange bias (PEB), i.e. a shift of M–H loop toward positive field axis, is also observed even when the sample is zero field cooled. This behavior is unusual for a soft ferromagnets like Py, which is expected to show symmetrical M–H loops (no H_{EB}) when it is deposited onto non-magnetic surfaces like MgO. Furthermore, when the sample is positive/negative field cooled, the M–H loops are shifted along both the horizontal and vertical axes with a clear field and temperature dependence. Classical interface related exchange bias models fail to explain the observed PEB in this sample.

In our earlier work [12], we have reported that Py grows epitaxially as islands on MgO/TiN buffered Si (100) substrates via domain matching epitaxy (DME) and Volmer–Weber type growth. In the current work, we have prepared three Py/MgO/TiN/Si (100) epitaxial thin film heterostructures by pulsed laser deposition (PLD) and measured their magnetic properties as a function of increasing number (3000, 6000 and 12,000) of laser pulses. To describe briefly, TiN, MgO, and Py targets were ablated sequentially in the same experimental run. The deposition of the TiN layer was performed at 625 °C in vacuum (1×10^{-6} Torr). Following TiN deposition, the first few mono layers (for about 500 pulses) of MgO were deposited under vacuum (1×10^{-6} Torr) at 575 °C. The remaining MgO was then deposited at the same temperature under an oxygen pressure of 6×10^{-4} Torr. Finally, Py deposition was carried out under vacuum at a substrate temperature of 550 °C. The energy density and pulse frequency were 1.5–3 J/cm² and 10 Hz, respectively. Once completed, the samples were cooled slowly to ambient temperature under vacuum. The deposition conditions were kept constant for all three samples studied except for progressively increasing the number of laser pulses, which resulted in an increasing island size and improved uniformity (size and

shape) of the morphology. The current samples have not been subjected to any post annealing treatments. No capping layer has been deposited on the Py/MgO/TiN/Si (100) heterostructures. We prepared multiple samples of the 6000 pulses and found consistent results.

From the out-of-plane and in-plane XRD measurements (see, Figs. S1 and S2 in the supplementary information), we found that the Py films grew epitaxially. SEM and AFM characterization (see, supplemental information, Figs. S3(a–c) and S4) of the film morphology revealed that the films of 6000 and 12,000 pulses were surprisingly similar, which may indicate that ablation rates from the Py target were dropping significantly over time. Fig. S3(d) shows the representative SEM image of MgO thin film surface collected on MgO/TiN/Si (100) template exhibiting features of continuous film. TEM samples were prepared by focused ion beam (FIB) technique. TEM bright field cross section images (see Fig. 1) indicate that the Py layer thicknesses gradually increased from 30 nm (3000 pulses) to 45 nm (6000–12,000 pulses), and island sizes increased from discretely separated islands roughly 60–90 nm in diameter to more densely packed films composed of irregularly shaped islands (recently coalesced islands) in the range of 150–600 nm (6000 pulses) that gradually develop into fairly equiaxed grains roughly 300–700 nm in diameter (12,000 pulses). For convenience, the three samples have been labeled as Py3, Py6 and Py12.

Temperature- and magnetic-field dependent magnetization measurements were carried out using a Quantum design MPMS/PPMS SQUID VSM dc magnetometer with the sensitivity $\leq 10^{-8}$ emu at 0 Tesla (T). It should be noted that the TiN and MgO buffer layers were confirmed to be non-magnetic, and hence, are not expected to contribute to the magnetic properties of the Py3, Py6 and Py12 heterostructures presented in this work. For magnetization measurements, a 4 mm $\times$ 4 mm piece of the sample was used in each case. Measurements were performed at various temperatures with the magnetic field applied either parallel or perpendicular to the film plane. Great care was taken not to contaminate the samples. Magnetic force microscopy (MFM) images of these samples were scanned using a Digital Instruments D3100 magnetic force microscope with Co–Pt tip. All the MFM images were obtained at the same lift scan height of 20 nm, using the tapping/lift mode at room temperature. The scan speed is 1 Hz. Magnetic field (3000 Oe) is applied parallel to the sample surface. Both MFM and AFM data have been presented in supplementary information (see Fig. S4).

As described in detail in our earlier work [12], c-axis oriented films composed of Py islands have been grown by pulsed laser deposition (PLD) on Si (100) using epitaxial MgO/TiN as a template

Fig. 1. Bright field cross-section TEM images collected from Py3 (a), Py6 (b) and Py12 (c). All the Py films were grown at 550 °C. All 3-layers TiN, MgO and Py in each heterostructure are seen. The top layer is of Pt sputtered during FIB sample preparation process. The scale bar is 100 nm in (a) and (c); the scale bar is 50 nm in (b). The island structures in each Py layer are clearly visible.

Fig. 2. Temperature (4–400 K) dependence of magnetization under zero field cooled (ZFC) and field cooled (FC, 1000 Oe) conditions for Py3 (a), Py6 (b) and Py12 (c).

buffer and diffusion barrier. This heterostructure is characterized by large lattice misfit strains. The lattice misfit at the NiFe ($a = 3.55 \text{ \AA}$)/MgO ($a = 4.22 \text{ \AA}$) and TiN ($a = 4.24 \text{ \AA}$)/Si ($a = 5.43 \text{ \AA}$) interfaces are 17% and 22%, respectively. Epitaxial growth of relaxed films (at the growth temperature) was achieved through a domain matching epitaxy (DME) paradigm [13] where three lattice planes of Si ($a = 5.43 \text{ \AA}$) match well with four of TiN ($a = 4.24 \text{ \AA}$) and five planes of MgO ($a = 4.22 \text{ \AA}$) match well with six of Py ($a = 3.55 \text{ \AA}$). In-plane (IP) and out-of-plane (OOP) XRD pattern and TEM studies confirm that all three layers have grown epitaxially on the Si (100) substrate with misfit dislocations present at the respective interfaces providing for fully relaxed films at the growth temperature. From the 2θ XRD data, we determined the OOP lattice parameters of Py3, Py6 and Py12 are of $3.50 \text{ \AA}$, $3.52 \text{ \AA}$ and $3.53 \text{ \AA}$, which are smaller than that ($\sim 3.55 \text{ \AA}$) of bulk Py film [14] and indicative of a film that is under in-plane tensile strain ($\sim 0.5\%$). This is consistent with the value of 0.53% one might expect to result during film cool down given the respective differences of the thermal coefficients of expansion of the different layers making up the heterostructure.

The temperature-dependent (4–400 K) magnetization measurements for Py3, Py6 and Py12 heterostructure have been performed and the data are shown in Fig. 2(a–c). The first measurement (measuring field of 300 Oe) was taken after zero-field cooling (ZFC) of the sample to the lowest temperature possible (4 K) and then recording the magnetic moment as it was warmed to above room temperature in an applied field of 300 Oe. In the second run, the measurements were taken for samples under field-cooled (FC)

(1000 Oe) conditions. An apparent blocking temperature (T_b), at which ZFC and FC curves split, is observed that appears to be $\geq 400 \text{ K}$ for all three samples. However, it should be noted that the width of splitting between the ZFC and FC curves for the Py6 sample is greater than that of the other two samples. Also, this value of T_b is much higher than the value ($\sim 78 \text{ K}$) previously reported [15] for interacting spherical Py nanoparticles prepared through magnetron sputtering on SiO_2 substrates. In the present case, the high T_b may be ascribed to the existence of large single-domain non-spherical islands that are physically isolated but magnetically coupled.

Fig. 3a compares the isothermal (4 K) M–H data taken for all three samples under ZFC conditions. As it can be seen, the three samples exhibit three distinct characteristic M–H loops. Py3 (shown in black symbols) shows saturated hysteresis with a room temperature H_c of about 200 Oe, Py6 (shown in red symbols) reveals unsaturated loops with much wider hysteresis and Py12 (shown in blue symbols) presents a narrow ($H_c \sim 100 \text{ Oe}$) and saturated hysteresis loop. This final value of H_c compares well with the values ($\sim 120 \text{ Oe}$) previously reported for Py films deposited on MgO substrates [6]. The latter were epitaxial thin films that were prepared by MBE or e-beam evaporation techniques at the substrate temperature of $500 \text{ }^\circ\text{C}$. The distinct features observed in the current study reflect the strong influence of island size and shape on the magnetic properties of Py. In addition, this plot suggests that the magnetization reversal process and domain configurations in these three samples are not the same. It appears that in the Py6 sample the domains are strongly pinned during the

Fig. 3. Comparison of isothermal (4 K) M–H loops of various Py films (a) variation of exchange bias field (H_{EB}) and coercive field (H_c) (b) as a function of pulse number. Lines guide the eye.

magnetization reversal process. Another noteworthy feature of the Py6 films is that they all have a positive exchange bias (PEB) i.e. the M–H loop is shifted toward positive magnetic field axis under ZFC conditions, unusual for soft FM like Py interfaced with non-magnetic material like MgO. One would normally expect to observe a symmetric M–H loop. The values of exchange bias (H_{EB}) and H_c of Py6 are significantly larger than those of other two samples as illustrated in Fig. 3b. As described above, the magnetic characteristics of Py6 differ considerably from Py3 and Py12, which display well behaved saturation magnetization and more symmetrical hysteresis loops. Hence, we devote the remaining part of the work to explore the anomalous magnetic properties of Py6.

Field cooled cooling (FCC) and field cooled warming (FCW) measurements have been performed on Py6 to establish the absence/presence of frustrated magnetic phases. Under FCC protocol, Py6 was field (1000 Oe) cooled from 400 K down to 4 K while M–T data were collected. In the warming cycle (FCW), the M–T data were collected from 4 K up to 400 K under the magnetic field of 1000 Oe. The data are displayed in Fig. S5 (see supplementary information). As it can be seen, FCC and FCW curves overlap and show no thermal hysteresis suggesting [16] the existence of no frustrated magnetic phases. In addition, time dependent magnetic measurements have been performed on Py6 under zero field cooled (ZFC), +7T and –7T for about 1 h. As shown in Fig. S6 (see supplementary information), we see practically no change in the magnetic moment as a function of time, thus suggesting the presence of stable pinned domain configurations.

Fig. 4a displays isothermal in-plane M–H data collected for Py6 at temperatures ranging from 4 to 400 K with the magnetic field varied from +500 to –500 Oe. Fig. 4b plots the out-of-plane M–H data collected on the same sample across the same temperature range with the magnetic field scanned from +1000 to –1000 Oe. As it can be seen, in the direction parallel to the film plane, the sample shows unsaturated M–H loops, with a H_c as high as 200 Oe at 4 K. A small moment and no magnetic hysteresis behavior is seen in the direction perpendicular to the film. This behavior is typical of that expected [17] for a film with its easy direction lying in the plane of the film.

The magnetic characteristics of Py6 can be quantitatively analyzed in terms of the H_{EB} and H_c , given by $H_{EB} = (|H_{CL}| - |H_{CR}|)/2$ and $H_c = (|H_{CL}| + |H_{CR}|)/2$ where H_{CL} and H_{CR} are the fields corresponding to $M = 0$ in the left and right branches of the loops, respectively. In each case the sample was field cooled (FC) from 400 K down to the measuring temperature. Fig. 5(a and b) shows the strong temperature dependences of H_c and H_{EB} as a function of temperature. The H_c increases linearly with the decreasing temperature suggesting that a single domain wall pinning mechanism [18] is active across the entire temperature range studied. This is consistent with thermally activated coherent domain wall motion originating within single domain particles. The H_{EB} decreases rapidly with increasing temperature and can be described by double exponential decay (solid line fit) consistent with a combination of pinned and rotatable spins. The H_{EB} in Py/MgO is accompanied by a large vertical shift in the magnetic moment, indicating the presence of a substantial number of pinned spins (discussed latter). Both H_c and H_{EB} are non-zero even at 400 K, further supporting that T_b is above 400 K. In contrast to the present case, Dimitrov and co-authors [19] have reported that both H_c and H_{EB} are independent of temperature when Py is interfaced with non-magnetic MgO as compared with the strong temperature dependences measured when Py is interfaced with AFM layers [7–11].

Fig. 6 compares the isothermal (4 K) M–H data collected under zero field cooling (ZFC), positive field (1000 Oe) cooling (PFC), and negative field (–1000 Oe) cooling (NFC) conditions on Py6. Under PFC, the vertical loop shifts upward; and to the left (toward negative field axis) and for the NFC, the vertical loop shifts downward

Fig. 4. Isothermal magnetization measured under in-plane (a) and out-of-plane (b) configurations collected on Py6 at various temperatures spanning from 4–400 K.

and toward the right along the positive field axis. The vertical shift is due to pinned moments that are not rotated by the applied field and therefore define the bias direction. From the asymmetry between vertical loop shift and loop height, the percentage [20] of pinned spins can be determined to be about 50% i.e. roughly half of the spins (50%) are strongly pinned. This percentage is much higher than the numbers (2–4%) usually observed for a typical FM/AFM interface [7–11]. Although the exact location of the spins has not been specified in this study, we would assume that they are the pinned spins are somehow linked to the very irregular shape of the grains in this particular sample.

To gain additional insights, extensive isothermal (4 K) M–H measurements have been undertaken for Py6 cooled under a variety of magnetic fields (500, 2000, 4000, 6000, 10,000, 70,000 Oe). As depicted in Fig. 7(a and b), we found that, when the sample was field cooled under the field of –7T, the H_{EB} (~60 Oe) is reduced by 43% from its ZFC value (~105 Oe), and M–H loop becomes more symmetrical (shifting both vertically and laterally) (see Fig. S7 in supplementary information). Interestingly, when the sample was cooled under +7T, the M–H loop remains the same as ZFC (cf. Figs. 3a and 6), that is, the H_{EB} and H_c did not show significant change. This indicates that the H_{EB} is sensitive to the direction of field cooling. These results suggest that a large fraction of the spins are strongly pinned and that fields in excess of 7T are required to rotate them away from their originally fixed direction.

Fig. 5. Temperature dependences of H_c (a) and H_{EB} (b) for Py6. The solid lines guide the eye for linear and double exponential decay of H_c and H_{EB} , respectively.

Fig. 6. Comparison of the isothermal (4 K) magnetization data measured under ZFC, positive field (+1000 Oe) cooled (PFC) and negative field (−1000 Oe) cooled (NFC) conditions on Py6. Horizontal and vertical loop shifts are observed under field cooled conditions.

H_{EB} and training effect (TE) are fundamental magnetic coupling phenomenon, and they are usually observed [7–11] in mixed magnetic metallic systems, in which AFM and FM phases coexist. The

TE is typically manifested as the reduction of the EB field (H_{EB}) upon progressive field cycling. From the variation of H_{EB} vs. the number of field cycles (n), the transition from the non-equilibrium to equilibrium nature of the spin structure can be inferred. Fig. 8 presents the results pertaining to TE as a function of n . As can be seen, the PEB is unchanged after 11 consecutive magnetic field cycles; there is no evidence of TE in this system even after 11 field cycles. In addition, we find that TE is unaffected by field sweep rate. In contrast to the present case, a strong TE is observed [7] in Py when interfaced with an AFM hematite layer. This experimental data further confirms the absence of frustrated magnetic phases and leads us to believe that the formation of AFM oxides is not the dominant mechanism for PEB and spin pinning in our case.

There have been many reports of PEB in the literature [21–28] which have been attributed to interface coupling [24], disorder [25] and training induced [28] mechanisms. But these are usually associated with interface between FM/AFM or FM/ferrimagnetic materials, which is not the case here. However, one recent work [29] is worth mentioning here. An H_{EB} of ~ 19 Oe is reported for a Fe/MgO sample. The authors attributed this to the formation (unwanted) of an Fe–O reaction product at the interface introducing an AFM spin coupling. In our sample, the possible formation of Fe-related AFM oxides such as Fe–O [30], γ -Fe₂O₃ [31] and Fe₃O₄ [32] at the Py/MgO interface cannot be entirely ruled out. But, the T_b 's associated with these oxides have been found to be ~ 30 K, 100 K and 120 K, respectively, well below the observed $T_b > 400$ K in our work. Similarly, NiO formation at the Py/MgO interface is reported [33] to have a $T_b < 300$ K. Based on this argument, we may conclude that AFM oxide formation is not likely to be major source of the PEB in Py6. From the above experimental data and discussion, it can be inferred that the PEB and associated magnetic properties in Py6 do not originate from interface effects, frustrated magnetic phases or lattice mismatch strain.

Because the islands in our samples are physically separated, the direct exchange mechanism via conduction electrons or via electron tunneling can be excluded. The demagnetization field (< 100 Oe) resulting from particle shape anisotropy cannot be responsible for observed PEB field of 50–100 Oe. According to the model proposed by Meiklejohn and Bean [34], the effective unidirectional magnetic energy is given by $\sigma = H_{EB} M_{FM} t_{FM}$, where H_{EB} is exchange bias field (~ 105 Oe at 4 K), M_{FM} is the magnetization (at 4 K) in emu/cm³ (for 4 mm $\times$ 4 mm sample) of ferromagnetic layer of thickness (t_{FM}) of 45 nm. This leads to a value of $\sigma = 8.4$ erg/cm² in our system, which is much greater than the values of 0.057 erg/cm² and 0.024 erg/cm² reported for Py interfaced with AFM layers such as FeMn [35] and BiFeO₃ [36], respectively. This is too large to explain with interfacial uncompensated spins. In addition, if the exchange bias is interface-related, we would expect to observe an inverse pulse number (or film thickness) dependence of H_{EB} , which is not the case as one can see from Fig. 3b.

To get additional insights, MFM measurements (see in supplementary information, Fig. S4) have been performed on all three samples at 300 K under zero (remanence) and 3000 Oe (maximum field available). The MFM data indicate gradual transition from single- to multi-particle domain structures. Results indicate that there is an intermediate regime where one moves from single isolated islands to fully coalesced film behavior, where the magnetization reversal is greatly hindered in as much as 50% of the film. This observation is consistent with the in-plane isothermal M–H data (see Fig. 3a), where Py3 and Py12 saturate at lower fields (< 200 Oe), but the Py6 film fails to saturate even in fields up to 70,000 Oe. We believe this anomaly can largely be attributed to the nature of the microstructure as the films transition from discrete to coalesced grain structures and a possible accompanying transition in domain dynamics. Trunk and co-authors [37] have

Fig. 7. Cooling field (H_{FC}) dependence of H_{EB} (a) and H_c (b) when Py6 is positive and negative field cooled collected at 4 K. Lines guide the eye.

Fig. 8. Isothermal (4 K) magnetization as a function of number ($n = 11$) of field cycles demonstrating no training effect. As is seen, M–H loops resulted from all 11 field cycles display robust PEB.

shown that a transition from Bloch to Neel domain wall structures takes place with decreasing Py film thickness. Neel wall dynamics were found to govern under conditions where demagnetization effects dominated, and Bloch wall switching begins to emerge as the exchange interaction becomes more prevalent. This transition has been confirmed by Chen et al. [38] in ferromagnetic resonance measurements performed on Py as a function of thickness of Py film. We believe a similar scenario is likely to be occurring in the present system as a function of island consolidation. Accordingly, the magnetic properties of Py6 are believed to be dominated by a competition between demagnetization and exchange interactions of nearly equal magnitude that lead to the observed PEB and appearance of unsaturated and broad M–H loops.

To conclude, we find that the magnetic features of Py films on the verge of coalescence are distinctly different from those films composed of either discrete island or compact equiaxed grains. In particular, the Py films in this transitional state display wider unsaturated magnetic hysteresis loops and a positive exchange bias, unusual for a soft ferromagnet like Py when the sample is zero field cooled. The exchange bias is accompanied by a vertical shift when the sample is zero/positive/negative field cooled, indicating

the presence of a large fraction of strongly pinned spins. The isothermal in-plane and out-of-plane magnetic measurements indicate that the films are highly anisotropic with an easy magnetization axis lying in the plane of the film. Repeated cycling (11 field cycles) and hysteresis loops measured up to fields of 7T produced reproducible hysteresis loops indicating the coexistence of strongly pinned spin configurations and robust ferromagnetism. From the experimental findings, it is clear that interface affects, disorder magnetic phases and lattice-misfit strain do not explain the observed magnetic properties of Py6. Classical exchange bias models alone also fail to explain the origin of the observed positive exchange bias in our systems. We believe that such anomalous magnetic features of Py6 may be understood in terms of a highly irregular and disconnected microstructure that may also be complicated by a transition from Bloch to Neel domain wall dynamics that occurs as a function of Py island size.

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Appendix A. Supplementary materials

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.cossms.2014.02.001>.

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Positive exchange bias in epitaxial permalloy/MgO integrated with Si (100)

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S1. Typical θ -2 θ (out of plane) XRD pattern of a Py/MgO/TiN/Si (100) film showing high crystalline quality, single phase and (00 l) orientation. The (002) reflections of TiN ($a = 4.24$ Å) and MgO ($a = 4.22$ Å) overlap as their lattice parameters are nearly identical. The (002) reflection of Py is clearly visible. Y-axis is shown in log-scale.

S2. XRD ϕ -scan pattern (in-plane) of Py (111) reflection collected from Py/MgO/TiN/Si (100) at $2\theta = 43.82^\circ$, $\omega = 21.91^\circ$ and $\chi = 55^\circ$, showing 4 peaks separated by $\sim 90^\circ$ indicating its cubic symmetry and epitaxial relationship with the substrate.

Fig.S3: SEM images of Py3 (a), Py6 (b) and Py12 (c). The scale bar is one micron.

Fig.S3(d): SEM image of MgO/TiN/Si (100) taken on MgO surface, revealing uniform continuous film.

MFM images of Py films were scanned using a Digital Instruments D3100 magnetic force microscope. The magnetization of the Co-Pt tip with high coercivity and high lateral resolution is perpendicular to the sample surface and points downward. The tip field interacts with the stray field coming from the sample surface; the magnetic transitions appear as either dark or bright in MFM images, corresponding to attractive or repulsive force gradient, respectively. All the MFM images were obtained at the same lift scan height of 20 nm, using the tapping/lift mode at room temperature. The scan speed is 1Hz.

MFM images were taken in the remanent state ($H=0$); the domain structures at remanence state seem quite complicated. This is expected, because the islands on the sample surface are interacting with each other to minimize the overall energy. Upon the application of magnetic field of 3000 Oe, the MFM images show the magnetic alignment of domains, which means the spins in all the islands are almost pulled in plane and aligned parallel each other and thus show clearer alignment of domains (the magnetization direction of islands points from dark to bright). This result is consistent with the observed in-plane magnetic hysteresis loops (saturated under this field). It should be noted that the domains are trapped due to structural imperfections of Py films, consequently, we are able to see the better contrast of domain features under the higher magnetic field.

In the Figures S4 (a), (b), (c), we present the AFM (left) and MFM (right) images collected at room temperature (300 K) under zero magnetic field (remanent state) and higher magnetic field of 3000 Oe, from Py3, Py6, and Py12, respectively. As it can be distinctly seen for Py12 (right image in Fig. S4(c)), the domains are well aligned up on the application of 3000 Oe, consistent with the in-plane M-H data (see Fig 3(a) in the manuscript).

H = 0 **H = 3k Oe**

Fig.S4 (a): AFM (left) and MFM (right) images of Py3

H = 0

H = 3k Oe

Fig.S4 (b): AFM (left) and MFM (right) images of Py6

Fig.S4 (c): AFM (left) and MFM (right) images of Py12

Fig. S5: Temperature (4-400 K) dependence of magnetization under field cooled cooling (FCC) and field cooled warming (FCW) conditions collected on Py6.

Fig. S6: Time dependence of magnetization collected under zero field cooled (ZFC) and field cooled (+70000 Oe and -70000 Oe) conditions on Py6.

Fig. S7: Isothermal M-H loops collected on Py6 when the sample was +ve and -ve field cooled under various magnetic fields of 500, 2000, 4000, 6000, 10000 and 70000 Oe).