Perpendicular anisotropy and out-of-plane exchange bias in nanoscale antidot arrays

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Abstract. We report on tailoring the perpendicular anisotropy, and out-of-plane exchange bias in (Co/Pd) multilayer exchange coupled to CoO, by patterning them into ordered arrays of antidots or ‘holes’. The coercivity in the multilayers is enhanced due to strong domain wall pinning in the vicinity of the antidots. We also observed a smaller degree of asymmetry in the reversal of the exchange-biased antidots as compared to the continuous film. Moreover, a strong interplay between thermal activation and CoO domain size confinement results in the exchange bias for the antidots being either smaller or larger than the continuous film.

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1. Introduction

There has been emerging scientific interest in the magnetization reversal mechanisms of patterned nanomagnets with perpendicular anisotropy due to potential applications in a wide range of technologies, such as ultra-high density data storage [1] and the implementation of spintronic-based devices [2]. From a fundamental viewpoint, nanomagnets offer a gamut of tailored magnetic phenomena by virtue of their low dimensionality, specifically when the physical dimensions become comparable to or smaller than certain relevant length scales [3]. It is evident that achieving a consistent and experimentally controllable switching mechanism in nanomagnets with perpendicular anisotropy is likely to be one of the key issues for further technological developments. Since nanomagnets with a uniform and ordered geometry are highly desirable for implementing perpendicular anisotropy in confined geometries, it would be convenient to exploit the efficient geometry of antidot arrays, which consist of ‘holes’ embedded in a continuous magnetic film [4], especially in view of recent results on percolated perpendicular media comprising exchange coupled grains with non-magnetic pinning sites [5]. These ‘holes’ alter demagnetizing fields and act as domain wall pinning sites that hinder the propagation of domain walls, thus resulting in an enhanced coercivity as compared to continuous films, and also they provide an effective technique for engineering the magnetic anisotropy.

Additionally, it has been shown that as a source of tunable unidirectional anisotropy, exchange bias, which refers to the unidirectional shift of the hysteresis loop along the magnetic field axis in exchange interacting ferromagnetic/antiferromagnetic (FM/AFM) systems, can be exploited to control the reversal mechanism in patterned nanomagnets [6]. From a fundamental point of view, investigation of exchange bias in systems with lateral dimensions comparable to AFM or FM domain sizes is crucial since it allows probing of the role of domains in exchange bias [7]–[10] and also aids in understanding the influence of lateral confinement and shape anisotropy on exchange bias fields and magnetization reversal mechanisms [11, 12]. Alternatively, from a technological aspect, exchange bias can be employed as a tunable source of unidirectional anisotropy to stabilize the magnetization in nanostructures and hence enable the reduction of length scales that determine the superparamagnetic limit [13].

Although there have been several studies on exchange bias in antidot arrays with in-plane anisotropy [9, 10], [13]–[15], the effect of antidots on perpendicular anisotropy and out-of-plane exchange bias is not fully understood. This may be attributed to the fabrication technique employed, which could result in the deterioration of perpendicular anisotropy due to possible material degradation. Hence, it is imperative to investigate such effects in ordered antidot arrays wherein effects arising from the reduction of lateral dimensions can be clearly distinguished.

In this work, the evolution of perpendicular anisotropy and out-of-plane exchange bias in antidot arrays consisting of [(Co/Pd)] multilayer exchange coupled to CoO has been studied. Using CoO as the AFM layer is appropriate due to its Néel temperature $T_N = 291$ K, which is just below room temperature, thus enabling the exchange bias to be reset conveniently. We demonstrate that the use of antidots greatly enhances the parameters available for engineering the perpendicular anisotropy and exchange bias in nanostructures.

2. Experimental details

The nanoscale antidot arrays were fabricated over a large area ($4 \times 4$ mm$^2$) on commercially available Si substrates using deep ultraviolet (DUV) lithography at a 248 nm exposing...
Figure 1. (a) Schematic figure of multilayers deposited using dc magnetron sputtering and (b) XRD patterns for Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/Au(40 Å) multilayers.

wavelength. To create patterns in the resist, the substrates were coated with a 60 nm thick anti-reflective layer followed by 480 nm of positive DUV photoresist. A Nikon lithographic scanner with KrF excimer laser radiation was used for exposure, resulting in the formation of resist pillars. For pattern transfer, two series of multilayers (as shown schematically in figure 1(a)) of composition Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/Au(40 Å) and Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/CoO(50 Å)/Au(40 Å) were deposited using dc magnetron sputtering. The crystalline anisotropy of these films was examined by conventional $\theta$–2$\theta$ x-ray diffraction (XRD) scans using Cu K$_\alpha$ radiation. A typical $\theta$–2$\theta$ XRD scan for Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/Au(40 Å) multilayers is shown in figure 1(b). We observe a strong hcp $\alpha$–Co (100) + fcc Pd (111) peak at 2$\theta$ = 40.8$^\circ$, which thereby suggests strong perpendicular anisotropy in the multilayers. Moreover, the respective shifts in the $\alpha$–Co (100) peak from 2$\theta$ = 41.7$^\circ$ and the fcc Pd (111) peak from 2$\theta$ = 40.4$^\circ$ may be attributed to stress induced in the films during the deposition of ultrathin Co and Pd multilayers [16]. The magnetic film was then removed from the unexposed areas by ultrasonic-assisted lift-off in OK73 resist thinner. Lift-off completion was determined by the color contrast of the patterned film and confirmed by inspection under a scanning electron microscope (SEM). Details of
Figure 2. (a) SEM image of antidot array A with center-to-center spacing $\lambda = 415$ nm and diameter $D = 265$ nm and (b) room temperature hysteresis loops measured out-of-plane for antidot array A and the corresponding continuous film of composition Pd$^{(50 \text{ Å})}/[\text{Co}^{(5 \text{ Å})}/\text{Pd}^{(30 \text{ Å})}]_{10}/\text{Au}^{(40 \text{ Å})}$.

the fabrication process are described elsewhere [3]. The final structure consists of a uniform and perpendicularly magnetized antidot array A of diameter $D = 265$ nm and center-to-center spacing $\lambda = 415$ nm in a square lattice geometry, as shown in the SEM image in figure 2(a). For direct comparison, antidot array B with center-to-center spacing $\lambda = 415$ nm, diameter $D = 145$ nm, and identical film composition was also fabricated. The out-of-plane hysteresis loops were recorded using a focused magneto-optical Kerr effect (MOKE) setup with a spot size of 5 $\mu$m at room temperature.

For samples with CoO, exchange bias was set by field cooling the samples in the presence of an out-of-plane magnetic field $H_{\text{FC}} = 5$ kOe from $T = 300$ K (above the Néel temperature $T_N = 291$ K for bulk CoO) to the desired set point temperature $T$. After each measurement, the samples were warmed back to $T = 300$ K to reset the exchange bias. As reported earlier [17], exchange-biased systems with thicker CoO layers ($t_{\text{CoO}} \geq 5$ nm) reveal smaller training effects as compared to thinner CoO layers. Although training effects were negligible for our exchange-biased antidot arrays and films due to the choice of thicker CoO layer, we started the data acquisition only after the first five loops had been completed. The results presented in this work thus correspond to the state when no further variations in exchange bias or coercive fields occur.

3. Results and discussion

Figure 2(b) shows typical out-of-plane hysteresis loops recorded at room temperature for antidot array A of film composition Pd$^{(50 \text{ Å})}/[\text{Co}^{(5 \text{ Å})}/\text{Pd}^{(30 \text{ Å})}]_{10}/\text{Au}^{(40 \text{ Å})}$ and a continuous film.
deposited under identical conditions. We observed that both the continuous film and the antidot array demonstrate strong perpendicular anisotropy. The continuous film exhibits a square hysteresis loop with almost 100% remanence-to-saturation ratio and coercivity $H_C = 500 \text{ Oe}$. The reversal is typical of thin films with perpendicular anisotropy and is characterized by unimpeded domain nucleation and domain wall motion. For the antidot array, however, we observed that $H_C$ increases to 750 Oe while retaining a remanence-to-saturation ratio of close to 100%. Moreover, we also note that the abrupt transition in magnetization at $H_C$ is followed by a broadened transition at higher reversal fields. This broader transition may be attributed to the signal detected by the MOKE setup from sidewalls that are formed during deposition and liftoff. The enhancement in coercivity is ascribed to the domain-wall-pinning controlled magnetization reversal mechanism, wherein the non-magnetic antidots act as defect centers or pinning sites in the vicinity of which domain walls are pinned. For antidot array $A$, a large pinning field is expected due to the large diameter ($D = 265 \text{ nm}$) of the antidots. This may be attributed to domain wall energy being saved as a consequence of the non-magnetic defects [18], given that magnetic material is absent inside the antidots. Recently, Punz et al [19] have estimated the pinning energy of a domain wall with defects, and shown that in the limit $\delta_0 \ll d$, the pinning energy can be approximated by

$$E_{\text{pin}} \approx 4N\sqrt{AK_1t d},$$  

where $N$ is the number of defects, $K_1$ is the perpendicular anisotropy constant, $A$ is the exchange constant, $\delta_0$ is the bloch wall parameter, $t$ is the film thickness and $d$ is the defect diameter. The effective pinning becomes stronger with increasing $d$, and coercivity should thus exhibit large magnitudes for large defect diameters. Our experimental results for the antidot arrays are thus in qualitative agreement with equation (1).

The strong perpendicular anisotropy and domain wall pinning effects in the antidot arrays were further confirmed using out-of-plane MR measurements as a function of temperature. Figure 3(a) shows the representative MR curves for antidot array $A$ at $T = 200 \text{ K}$ and $T = 5 \text{ K}$, respectively. We observed that the key features of the magnetization reversal process that were noticeable in the hysteresis loops at room temperature also manifest themselves in the MR curves at low temperature. The enhanced coercivity of the antidot array can be ascertained from the two distinct maxima in the MR curves. Another prominent characteristic in the out-of-plane MR curves is the presence of a linear slope in the curves at high magnetic fields. This linear increase in resistance as the external magnetic field is reduced from saturation results from misalignment between the current density and magnetization at saturation due to geometrical confinement introduced by the antidots. The linear increase in resistance thus indicates that the local spins are more aligned with the current density during the initial reduction of external magnetic field from saturation [9]. We also observed that $H_C$ exhibits a near linear dependence on temperature, as shown in figure 3(b), and increases appreciably to 2.25 kOe at $T = 5 \text{ K}$. Such a temperature dependence of $H_C$ has been analyzed using the model developed by Kronmüller [20, 21], in which $H_C$ is related to the magnetic anisotropy $K_U$ and saturation magnetization $M_S$. A linear dependence of $H_C$ on temperature clearly demonstrates the role of domain wall pinning in the coercivity mechanism of the [Co/Pd] multilayers. Moreover, the enhancement of $H_C$ may be attributed to a reduction in thermal excitations with decreasing temperature, thus entailing higher switching fields to overcome the pinning energy barriers necessary for magnetization reversal. To corroborate the strong domain wall pinning in the antidot arrays as a function of temperature, we have also carried out identical MR measurements
Figure 3. (a) Out-of-plane MR curves for antidot array $A$ as a function of temperature and (b) temperature dependence of coercivity $H_C$ for antidot array $A$ (squares) and the continuous film (dots).

Having established conclusively that antidot arrays provide a simple yet efficient technique to tailor the switching field and magnetization reversal mechanisms in Co/Pd multilayers with perpendicular anisotropy, we have also investigated the effect of antidots on the perpendicular exchange bias in similar multilayers with CoO as the AFM layer. Shown in figure 4 are the representative out-of-plane MR curves as a function of temperature for the continuous film and antidot array $A$ of composition Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/CoO(50 Å)/Au(40 Å). At $T = 200$ K, we observed that the MR curve for the continuous film is shifted by $H_E = 285$ Oe from the zero field axis, and the magnetization reversal is highly asymmetric, as illustrated clearly by the difference in heights of the MR peaks at both the coercive fields in figure 4(a). The peak amplitude of the descending branch is $\approx 2.2$ times larger than that of the ascending branch. This distinction reflects the difference between the domain states in the two reversal branches. The observation of a higher MR peak has been correlated to higher effective domain wall density in the descending branch as compared to the ascending branch [22]. This mechanism for
Figure 4. Out-of-plane MR curves as a function of temperature for the exchange-biased continuous film and antidot array \( A \) of composition \( \text{Pd}(50 \text{ Å})/\text{[Co}(5 \text{ Å}))/\text{Pd}(30 \text{ Å})]_{10}/\text{CoO}(50 \text{ Å})/\text{Au}(40 \text{ Å}) \).

Asymmetric magnetization reversal in Co/Pd exchange-biased multilayers with perpendicular anisotropy is in contrast to our previous study on Co/CoO exchange-biased systems with in-plane anisotropy \([9, 10]\), wherein the asymmetric nature of the MR curve was attributed to different reversal mechanisms, i.e. nucleation of reversed domains and propagation of domain walls in the descending branch as compared to rotation of the magnetization in the ascending branch. As the temperature was reduced, we observed that the asymmetry in the MR curves decreased gradually due to a more balanced domain wall distribution in both branches, and is also reflected in a peak amplitude ratio of \( \approx 1.6 \) for \( T = 5 \text{ K} \).
In contrast to the continuous film, we observed that the corresponding MR curve for antidot array \( A \) exhibits negligible asymmetry at \( T = 200 \) K, and is shifted by \( H_E = 210 \) Oe. As expected, \( H_C \) for the antidot array is greatly enhanced as compared to the continuous film and also its unbiased counterpart. A discussion about the comparative magnitudes of \( H_C \) and \( H_E \) as a function of temperature will be presented subsequently in figure 6. We also established that the antidot arrays exhibit noticeable asymmetric reversal only below \( T = 100 \) K. Unlike the continuous film, however, variations in the degree of asymmetry for the antidot arrays were minimal as the temperature was reduced below 100 K. Figures 4(e) and (f) clearly show that the ratio between the peak amplitudes of the descending and ascending branches remains almost constant (\( \approx 1.4 \)) in the low-temperature regime. We attribute this trend to the fact that the density of geometrical pinning centers hindering domain wall motion is identical for both the descending and the ascending branches during the magnetization reversal process of the antidot array.

The evolution of \( H_C \) and \( H_E \) has also been systematically studied as a function of temperature for both the antidot array and the continuous film. To demonstrate the efficiency of our technique in tailoring the perpendicular anisotropy and exchange bias, we have also shown results from antidot array \( B \) with identical center-to-center spacing \( \lambda = 415 \) nm, diameter \( D = 145 \) nm and identical film composition. The SEM image of antidot array \( B \) is shown in figure 5(a). Typical out-of-plane hysteresis loops recorded at room temperature for antidot array \( B \) of film composition \( \text{Pd}(50 \, \text{Å})/\text{[Co}(5 \, \text{Å})/\text{Pd}(30 \, \text{Å})]_{10}/\text{Au}(40 \, \text{Å}) \) are shown in figure 5(b). We observed that antidot array \( B \) also demonstrates strong perpendicular anisotropy, and the reversal is similar to that of antidot array \( A \) with an abrupt transition in magnetization at \( H_C = 580 \) Oe, followed by a broadened transition at higher reversal fields. It is evident that the coercivity value for antidot array \( B \) is in agreement with equation (1), since the antidot diameter, and hence effective pinning, is smaller than that of antidot array \( A \).

Figure 5. (a) SEM image of antidot array \( B \) with center-to-center spacing \( \lambda = 415 \) nm and diameter \( D = 145 \) nm, and (b) room temperature hysteresis loops measured out-of-plane for antidot array \( B \) of composition \( \text{Pd}(50 \, \text{Å})/\text{[Co}(5 \, \text{Å})/\text{Pd}(30 \, \text{Å})]_{10}/\text{Au}(40 \, \text{Å}) \).
Figure 6. (a) Temperature dependence of coercivity $H_C$ and (b) temperature dependence of exchange bias field $H_E$ for antidot arrays $A$ and $B$, and the corresponding continuous film of composition Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/CoO(50 Å)/Au(40 Å).

Figure 6 shows the corresponding temperature dependence of $H_C$ and $H_E$ extracted from the out-of-plane MR curves for film composition Pd(50 Å)/[Co(5 Å)/Pd(30 Å)]$_{10}$/CoO(50 Å)/Au(40 Å). We observed that in good agreement with equation (1), $H_C$ for antidot array $B$ is larger than the continuous film and smaller than that of antidot array $A$ at all temperatures. $H_E$ for the antidot arrays also shows strong dependence on the antidot diameter $D$. The presence of antidots imposes physical constraints and restricts the AFM domain size. In addition, the AFM domain size is reduced further with increasing $d$ [14]. In agreement with Malozomoff’s static model, which predicts an inverse proportionality between the magnitude of $H_E$ and AFM domain size [23], figure 6(b) shows that $H_E$ for antidot array $A$ is larger than that of antidot array $B$ at all temperatures. Moreover, we also note that there is a crossover temperature in the evolution of $H_E$, which determines the temperature range in which $H_E$ for both of the antidot arrays is either larger or smaller than the continuous film. This
is attributed to strong competition between thermal activation effects that favor a reduction in $H_E$ in nanostructures, and constraints imposed on the AFM domain size by the reduced lateral dimensions of the antidot arrays, which favor an enhancement of $H_E$. It is evident that once thermal activation effects are minimized below 185 K (for antidot array $A$) and 115 K (for antidot array $B$), they exhibit larger $H_E$ values as compared to the continuous film.

4. Conclusions

In summary, we have successfully demonstrated the possibility of inducing perpendicular anisotropy and out-of-plane exchange bias in ordered nanoscale antidot arrays of varying size. We observed that the antidot arrays show larger $H_C$ values as compared with the continuous film due to strong domain wall pinning. For the exchange-biased system, the continuous film exhibits asymmetry in the magnetization reversal process due to differences in domain wall density in the two reversal branches, while the corresponding asymmetry in the antidot arrays is relatively smaller. Moreover, strong competition between thermal activation effects and AFM domain size confinement leads to $H_E$ for the antidot arrays being either smaller or larger than the continuous film. Our results demonstrate a convenient technique for tailoring the perpendicular anisotropy and exchange bias in systems with reduced lateral dimensions.

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References

[1] Hellwig O, Moser A, Dobisz E, Bandic Z Z, Yang H, Kercher D S, Risner-Jamtgaard J D, Yaney D and Fullerton E E 2008 Appl. Phys. Lett. 93 192501
[2] Mangin S, Ravelosona D, Katine J A, Carey M J, Terris B D and Fullerton E E 2006 Nat. Mater. 5 210
[3] Adeyeye A O and Singh N 2008 J. Phys. D: Appl. Phys. 41 153001
[4] Adeyeye A O, Bland J A C and Daboo C 1997 Appl. Phys. Lett. 70 3164
[5] Rahman M T, Shams N N, Wu Y-C, Lai C-H and Suess D 2007 Appl. Phys. Lett. 91 132505
[6] Nogués J, Sort J, Langlais V, Skumryev V, Suriñach S, Muñoz J S and Baró M D 2005 Phys. Rep. 422 65
[7] Fraune M, Rudiger U, Guntherodt G, Cardoso S and Freitas P 2000 Appl. Phys. Lett. 77 3815
[8] Baltz V, Sort J, Rodmacq B, Dieny B and Landis S 2004 Appl. Phys. Lett. 84 4923
[9] Tripathy D and Adeyeye A O 2009 Phys. Rev. B 79 064413
[10] Tripathy D and Adeyeye A O 2009 J. Appl. Phys. 105 07D703
[11] Hoffmann A, Grimsditch M, Pearson J E, Nogués J, Macedo W A A and Schuller I K 2003 Phys. Rev. B 67 220406
[12] Sort J et al 2008 J. Appl. Phys. 103 07C109
[13] Liu K, Baker S M, Tuominen M, Russell T P and Schuller I K 2001 Phys. Rev. B 63 060403
[14] Tripathy D, Adeyeye A O and Singh N 2008 Appl. Phys. Lett. 93 022502
[15] Rahman M T, Shams N N, Wang D S and Lai C-H 2009 Appl. Phys. Lett. 94 082503
[16] Thiyagarajah N, Bae S, Joo H W, Han Y C and Kim J 2008 Appl. Phys. Lett. 92 062504
[17] Brems S, Buntinx D, Temst K, Van Haesendonck, Radu F and Zabel H 2005 Phys. Rev. Lett. 95 157202–4

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[18] Rahman M T, Shams N N, Lai C H, Fidler J and Suess D 2010 Phys. Rev. B \textbf{81} 014418
[19] Punz D, Lee J, Fuger M, Fidler J, Schrefl T and Suess D 2010 J. Appl. Phys. \textbf{107} 113926
[20] Kronmüller H 1987 Phys. Status Solidi \textbf{144} 385
[21] Suzuki T 1995 Scr. Metall. Mater. \textbf{33} 1609
[22] Camarero J, Miguel J, Goedkoop J B, Vogel J, Romanens F, Pizzini S, Garcia F, Sort J, Dieny B and Brookes N B 2006 Appl. Phys. Lett. \textbf{89} 232507
[23] Malozemoff A P 1987 Phys. Rev. B \textbf{35} 3679