Strain engineered domain structure and their relaxation in perpendicularly magnetized Co/Pt deposited on flexible polyimide

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Abstract

The demand of fast and power efficient spintronic devices with flexibility requires additional energy for magnetization manipulation. Stress/strain have shown their potentials for tuning magnetic properties to the desired level. Here, we report a systematic study for the effect of both tensile and compressive stresses on the magnetic anisotropy (MA). Further the effect of stress on the domain structure and magnetization relaxation mechanism in a perpendicularly magnetized Co/Pt film has been studied. It is observed that a minimal in-plane tensile strain has increased the coercivity of the film by 33% of its initial value, while a very small change of coercivity has been found under compressive strain. The size of ferromagnetic domains decreases under tensile strain, while no change is observed under the compressive strain. Magnetization relaxation measured at sub-coercive field values yields a longer relaxation time in the strained state.

1. Introduction

Miniaturization and flexibility with high speed and low power consumption are the key aspects for developing next generation spintronic devices [1–3]. Perpendicular magnetic anisotropic (PMA) systems such as Co/Pt bilayers have shown their importance in increasing thermal stability and developing non-volatile magnetic random access memories (MRAM’s) with low current density requirement for magnetization switching at 20 nm bit size level [1, 4–9]. The major challenge in further downscaling of devices is the higher power dissipation [10]. Various approaches have been taken to reduce the power consumption during magnetization switching e.g. spin transfer torque (STT), current induced domain wall (DW) motion etc. [11, 12]. However, involvement of charge current is still a major factor to further reduce the power consumption. In this context, stress controlled switching of magnetization has been proven to be a promising one [13]. Energy dissipation in a non-volatile properly scaled memory cell to write 1 bit of information by stress is found to be within a few tens of aJ, which lies close to the thermodynamic limit [14]. Energy-efficient memory components could also be implemented in advanced flexible 2D spintronic devices for low power flexible spin current circuits [15]. Hence strain engineering became an appealing approach to meet the current device requirement. There are various possible ways to generate strain on a thin film deposited on rigid substrate e.g. by lattice mismatch [16, 17], voltage application via transducers [18] etc., whereas for a film deposited on flexible substrate bending, stretching, peeling or twisting mechanism also can generate adequate stress [19]. Such mechanical methods transfer almost uniform stress from substrate to the film, which gives rise to device flexibility. In recent years the effect of stress application on magnetic anisotropy and domain structure in various magnetic films with in-plane MA have been studied [3, 20]. However, the stress effect in flexible PMA films is less explored. Theoretical studies predicted that magnetic anisotropy energy as well as the easy axis of Fe/Pt multilayers can be modified by strain [21]. Shepley et al showed that the MA of a Co/Pt film decreased with increasing out-of-plane tensile strain while the DW velocity was increased by 30 to 100% [22]. Later the stress tuned DW velocity of Co/Pt depends on DW formation energy (balance of anisotropy and exchange energy) is also reported [23]. However, a
systematic study on the effect of different types of strain e.g. tension, compression, peeling etc. on the MA, domain nucleation, its structure along with relaxation mechanism is still lacking. Exploring the effect of such magneto-mechanical coupling on different magnetic properties will not only help to understand the basics but also holds importance from application viewpoint. Here, we report a comprehensive study of the effect of tensile and compressive strain on the MA, domain nucleation and relaxation mechanism in a perpendicularly magnetized Co/Pt film prepared on polyimide substrate.

2. Experimental details

We have prepared platinum (Pt)/cobalt (Co) thin film upon 25 μm thick polyimide (PI) substrate with a tantalum (Ta) buffer layer by DC magnetron sputtering in a high vacuum chamber. PI substrate was cleaned ultrasonically by isopropanol before deposition. The sample structure is the following: PI/Ta(7 nm)/Pt (10 nm)/Co(0.7 nm)/Pt(3 nm) as shown in figure 1(a). The base pressure of the deposition system was lower than 1 × 10⁻⁷ mbar. The rate of deposition for Co, Pt and Ta layers were 0.10 Å s⁻¹, 0.15 Å s⁻¹ and 0.13 Å s⁻¹ respectively. The thickness was measured by a quartz crystal monitor (QCM) mounted close to the substrate holder. Substrate was rotated at 10 rotation per minute (rpm) during deposition to get uniformity in the samples. A 7 nm thick Ta buffer layer was used for better adhesion and growth of Pt layer along (111) direction. A 3 nm thick Pt was used as a capping layer to prevent the oxidation of Co layer.

Magnetic properties of the sample have been recorded at both the flat as well as bend state. Different tensile and compressive strains have been generated on the film by fixing it on convex and concave shaped molds, respectively. Schematic of the convex and concave shaped bent sample is shown in figure 1(b) and the details of stress generation method using molds are discussed in the supplementary information (figure S1, S2 available online at stacks.iop.org/NANOX/1/010037/mmedia). Domain nucleation and propagation at both states of the sample have been recorded by polar magneto optic Kerr effect (PMOKE) based microscope. Further, relaxation measurements have been carried out in the strained as well as unstrained state of the film.

3. Results and discussion

3.1. Hysteresis loop

We have measured magnetic hysteresis loops in the polar mode by bending the sample towards inward and outward direction. Such bending induced strain on the film can be determined as,

$$\epsilon_s = \frac{t}{2r \pm t}$$

(1)

here ‘$$\epsilon_s$$’ denotes the applied tensile strain whereas ‘$$\epsilon_c$$’ is for compression, r is the radius of the mold used and t is the total thickness of the film and substrate [24–27]. Using molds almost uniform strain is possible to transfer from the substrate to the thin film (as thickness of the film is very low in comparison to the substrate). Coercivity ($$H_c$$) of the sample was found to be 26.9 mT in flat state which increases to 35.3 mT under 0.13% tensile strain (figure 2(a)). Even a minimal in-plane tensile strain (<0.1%) has increased the coercivity largely (~9 mT) whereas the effect of compression on decreasing coercivity is almost negligible. Such a different behaviour can be explained by comparing the strength of stress induced anisotropy over interfacial anisotropy. Magneto-elastic energy of a thin film can be expressed as:

$$E_{ME} = \frac{3}{2} \lambda \sigma \sin^2 \theta$$

(2)

where λ is the coefficient of magnetostriction, $\sigma$ is the applied stress, $\theta$ is the angle between the $\sigma$ and magnetization vector [28]. As reported earlier λ is negative for Co/Pt system along (111) direction [22] while the
applied stress $\sigma$ is positive for tensile strain, hence the $\lambda\sigma$ product is negative which induces an easy axis (EA) along the perpendicular plane of the sample to minimize total energy of the system. However under compressive strain the stress induced anisotropy should orient in the plane of the sample. As the unstrained sample has a PMA due to $d-p$ orbital hybridization at Co/Pt interfaces, hence application of external stress will modify the strength of this anisotropy. A symmetric variation of PMA under out-of-plane tensile and compressive strain has been reported by Shepley et al when the Co/Pt sample is prepared near to spin-reorientation transition (SRT) region [22]. In our case the thickness of Co is 0.7 nm which is far away from the SRT region which might be a possible reason for the asymmetric variation of $H_C$ at different strain state.

The variation of coercivity with increasing tensile/compressive strain has been plotted in figure 2(b). Here the 1st cycle represents the values of $H_C$ obtained from the hysteresis loops when the sample is bent for the first time after deposition at different radii. After that we kept the sample under 0.42% tension for 15 days and re-measured $H_C$ at the same strain values of 1st cycle, which is plotted as 2nd cycle. The nature of the two graphs indicates an excellent endurance of the film under long term stress application. However under compression the effect is almost negligible and $H_C$ decrease by $\sim 0.8$ mT when 0.42% strain is applied. Dai et al have reported a similar asymmetric modification of squareness of a hysteresis loop under different strain [26]. For a FeGa sample, when $H$ and stress were applied along EA and hard axis (HA), respectively, the squareness of the hysteresis loop modified slightly under compressive while largely under tensile strain. In our work we have also applied $H$ along EA and strain along HA and found a similar asymmetric variation in $H_C$. Hence it can be concluded that the effect of tensile strain on $H_C$ can be negligible if we measure hysteresis loop along HA while applying stress along EA of the sample.

### 3.2. Bubble domain

Bubble domain has been observed at the initial flat state of the sample, as the anisotropy ratio $Q = K_u/K_d \gg 1$, where $K_u$ is the uniaxial anisotropy constant and $K_d$ is the stray field energy constant [29]. The domain images are recorded at saturation, near nucleation and sub-coercive field values of each hysteresis loops as shown in figure 3. All the domain images are captured at the same place of the sample to easily compare them under different strained conditions. Red marked circles in figure 3 ($b_1$ and $c_1$) are showing the domain size for better visualization in the flat state of the sample. Circles of same diameter are drawn on the domain images recorded under tensile ($b_3$ and $c_3$) and compressive ($b_2$ and $c_2$) strained states to quantify the contraction/elongation of the domains in comparison to the flat state. It is observed that application of an in-plane tensile strain reduces the size of bubble domains.

As $H_C$ of the sample has increased by 33% under tension, it hindered the DW propagation which reduces the domain size. Magnetization reversal alongwith domain nucleation also takes place at higher field values under tensile strain due to the increase in overall energy barrier. For compression the magnetization reversal occurs at slightly lower value from the unstrained state. The difference between nucleation field ($H_N$) and $H_C$ was found to be $2.5$ mT at the flat state whereas it increases to 4 and decreases to 1.2 mT under tensile and compressive strain, respectively. This difference in $H_N - H_C$ might be a result of modification in MA energy due to the application of $\pm 0.13$% strain. Under in-plane tensile strain the DW motion of our film is expected to be reduced due to the increase in potential barrier for the magnetization reversal.
Thin film damage under compressive strain may occur in two different ways: (i) wrinkling and (ii) buckling. Wrinkle formation occur for elastic substrate having good adhesion with the deposited film, so that they can deform simultaneously. Critical stress required for wrinkle formation \[ \sigma_w = \frac{E_f}{4} \left( \frac{3E_f}{E_s} \right)^{2/3} \] can be written as:

where \( E_f \) and \( E_s \) are plane-strain modulus for film and substrate, respectively. The critical stress estimated for wrinkle formation in our film is \( \sim 8 \) GPa. However, we have applied maximum compressive strain \( \epsilon = -0.42\% \) and the corresponding stress is \( \sigma \sim 0.9 \) GPa, which is far away from the critical stress. The detailed calculation for wrinkle formation is shown in the supplementary information. Thus wrinkle formation might be excluded within the applied range of compressive strain in our case.

It is known that buckling of a stiff film deposited upon elastic substrate can occur at lower strain values depending on morphology of substrate, presence of localized artefacts as well as the adhesion strength between the film and substrate [31–33]. The PI substrate used in our case is elastic in nature. Ta buffer layer having high chemical reactivity has been employed to increase the adhesion of the film with PI [34, 35]. This high adhesion will help to increase the critical strain required for buckle delamination in our case. Generally, after damage formation (cracking/buckling etc.) the electrical/magnetic properties of a system becomes irreversible [36]. In our case, after applying 0.42% compressive/tensile stress all the results found to be reversible which indicates that buckle delamination of deposited film does not appear in our case. Merabtine et al have shown that the magnetic properties of a CoFeB film deposited on Kapton modified due to the high stress applied during the damage formation (cracking/buckling). But there is almost no effect of cracking or buckling on the magnetic properties of zero magnetostrictive alloy NiFe [37]. Hence we can conclude that the negligible effect of compression is not a result of structural modification of our film.

Further, as the compressive strain applied in our case is temporary in nature hence we have tried to find out the effect of permanent compression. We have prepared the same sample on a bend PI substrate by fixing it on a convex mold of 5 mm radius. After deposition the sample is made flat and a permanent compression has been generated on the film. PMOKE measurement on the as deposited and flat sample has been recorded and a clear decrease in \( H_C \) has been observed. The details of deposition geometry and hysteresis loops are shown in the supplementary information (figure S3).

### 3.3. Relaxation dynamics

As the temporary compression does not show much effect in modifying magnetic properties of our film hence we performed the relaxation measurement under tensile stress only and compared with the flat one. Under the application of a constant Zeeman energy to the system, the magnetization relaxation can be recorded with respect to time. This measurement basically reveals the efficiency of thermal activation energy to complete magnetization reversal via domain nucleation and DW motion. We have performed similar measurement on
our film by PMOKE based microscopy. The sample was first saturated at a negative saturation field, then the field was increased manually near to positive sub-coercive field values e.g. 0.95 $H_C$, 0.97 $H_C$ and kept it constant there. In this scenario, further reversal takes place with the help of thermal energy. Domain images were captured during the whole thermal relaxation process. The amount of dark grey contrast represents a measure of the magnetization for the sample. The intensity of each image was extracted by ImageJ software and the average intensity of all images captured at each second were calculated. The normalized intensity is then plotted against the time taken to complete the reversal. Here the normalized intensity as a function of time basically reflects the net magnetization relaxation of the sample. Amongst various raised models to explain magnetization relaxation phenomena Fatuzzo-Labrune model is extensively used for FM thin films [38–43]. However, the approximation of single energy barrier made in this model does not hold for a real thin film which consists of a distribution of energy barrier with defects and inhomogeneity throughout the surface. Hence to fit our experimental data we have used the compressed exponential function [39, 44] written as:

$$I(t) = I_1 + I_2 \left(1 - \exp\left(-\left(\frac{t}{\tau}\right)^\beta\right)\right)$$

where $I(t)$ is the Kerr intensity measured at time $t$, $I_1 + I_2$ is the normalized Kerr intensity, $\beta$ is an exponent varying within 1 (nucleation dominated) to 3 (DW motion dominated) and $\tau$ is the relaxation time constant. At the flat state the relaxation happens via both domain nucleation as well as DW motion ($\beta = 1.46 \pm 0.01$ and 1.19 $\pm$ 0.01) where the relaxation time constants are found to be 10.90 $\pm$ 0.04 and 6.86 $\pm$ 0.07 seconds at 0.95 and 0.97 $H_C$, respectively. However, a slower relaxation phenomenon occurs in presence of 0.13% tensile strain which started via domain nucleation and dominated further by DW motion ($\beta = 2.23 \pm 0.04$ and 1.95 $\pm$ 0.04). Relaxation time constant at the bend state was 23.63 $\pm$ 0.14 and 16.24 $\pm$ 0.13 seconds at 0.95 and 0.97 $H_C$, respectively. Nature of the relaxation curve also reflects a slower relaxation phenomenon as shown in figures 4(a)–(b). This is supported by a larger relaxation time constant ($\tau$) extracted from the fitting. To explain the origin behind this slow relaxation we have analyzed the domain images correspond to 0.95 $H_C$ of both states at different time intervals viz. 0, 5, 10 and 20 s (figure 4(c)). Here 0 s signifies the initial time when the first image was taken after giving a constant (0.95 $H_C$) magnetic field. As 0.95 $H_C$ field is greater than the nucleation field for

![Figure 4](image-url). Relaxation measurement performed at two sub-coercive field values (a) 0.95 $H_C$ and (b) 0.97 $H_C$, at 0.13% strained (blue curve) and unstrained (black curve) state. The solid lines represents fitting of the experimental data. (c) Domain expansion during thermal relaxation at 0.95 $H_C$ magnetic field at unstrained (upper panel) and +0.13% strained (lower panel) state. Scale bar is 500 µm for all domain images.
both the states, hence bubble domains are visible at both the images taken at $t = 0$ s. All the domain images were taken at the same place of the sample, hence at $0$ s the size of the bubble domain in the bend state found to be smaller. Further with increasing time (5, 10 and 20 s) it has been found that the domain propagates slower in the bend state in comparison to the flat one due to its reduced wall motion. This reduced wall motion also reflects in the difference between $H_{\text{N}}$ and $H_{\text{C}}$ field values of both the states, as discussed earlier.

In about 20 s of the relaxation process, flat sample almost completes the reversal while for the bend one, it is still far away from the saturation. From the above result we expect that a Co/Pt thin film prepared near to SRT region will exhibit a significant reduction of relaxation time under compressive strain, which holds its potential in faster device application.

For flexible substrate, the deposited film always experience some dynamic bending during its handling [45, 46]. In our case also during magnetic measurements of the film, dynamic bending might have occurred. But while we repeated the measurements for each strain values we found almost same result each time. Hence it can be concluded that the dynamic bending of flexible film does not affect its functional properties largely.

It should be noted that in Ta/Pt/Co/Pt, a proximity induced magnetism may occur at the interface of Co and Pt [47–50]. However, our studies of strain is a global one and it is difficult to differentiate the effect of strain on the Co film from the Co-Pt interface.

4. Conclusion

We have presented the effect of both tensile and compressive strain on the magnetization reversal, domain nucleation, its propagation and relaxation dynamics of a Co/Pt flexible film. It has been observed that tensile strain has largely modified the coercivity, domain nucleation and relaxation mechanism of our film, whereas the effect of compression was found to be less prominent. A substantial increase in MA, nucleation field and decrease in domain size attributed mainly to the magneto mechanical coupling effect. A longer field span required to complete the magnetization reversal in the strained state reflects an increase in potential barrier for the DW movement, which further modified the relaxation time of the film. Relaxation measurements indicates $\sim 13$ s of relaxation time difference between the unstrained and curved state which has been endorsed by comparing domain images taken at different time interval. As a future work, numerical modeling can be done to better understand the effect of magneto-elastic coupling on modifying magnetic properties of a film.

Engineering domain size or structure by strain can affect the density of data recording in storage media. In addition to this, our flexible film also exhibits excellent endurance for long term stress application which holds its potential where small but repetitive stress needs to be sustained by the film.

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