Perpendicular magnetic anisotropy and the magnetization process in CoFeB/Pd multilayer films

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Abstract
The perpendicular magnetic anisotropy (PMA) and dynamic magnetization-reversal process in [CoFeB t nm/Pd 1.0 nm]n (t = 0.4, 0.6, 0.8, 1.0 and 1.2 nm; n = 2 – 20) multilayer films have been studied by means of magnetic hysteresis and Kerr effect measurements. Strong and controllable PMA with an effective uniaxial anisotropy up to 7.7 × 10^6 Jm^-3 and a saturation magnetization as low as 200 emu cm^-3 are achieved. The surface/interfacial anisotropy of the CoFeB/Pd interfaces—the main contribution to the PMA—is separated from the effective uniaxial anisotropy of the films and appears to increase with the number of CoFeB/Pd bilayers. Observation of the magnetic domains during a magnetization-reversal process, using polar magneto-optical Kerr microscopy, reveals the detailed behavior of the nucleation and displacement of the domain walls.

Keywords: magnetic multilayer, perpendicular magnetic anisotropy, interfacial magnetism, magnetic domains and domain walls, magnetization reversal

(Some figures may appear in colour only in the online journal)
considering PMA amorphous CoFeB films as an excellent candidate for spin-transfer torque applications:

- Importantly, the CoFeB amorphous films possess high spin polarization (over 65%) [20]—even higher than that in crystalline CoFeB film [29].
- The absence of grain boundaries in an amorphous thin layer should significantly reduce the pinning site density that hinders the magnetization reversal. This can enhance the speed of the spin switching, which is desirable for high speed spin-transfer torque devices. Beside that, amorphous layers could create very smooth interfaces that would reduce the attenuation of the electron spin during transfer through the interface, which is desirable for high tunneling magnetoresistance [30, 31], when the use is in magnetic tunneling junctions.
- The multilayer structure of amorphous CoFeB and the noble metal spacing layers (e.g. Pd or Pt) removes the risk of oxidation and diffusion that would affect the magnetic properties of the films, in comparison with crystalline CoFeB film interfaced with an oxide layer (e.g. MgO).
- The presence of B atoms, with smaller radius than those of Co and Fe, resulting in a high glass-forming ability [32] and, consequently, high stability of the amorphous structure as compared to rare earth-transition metal amorphous films (e.g. TbFeCo and GdFeCo) [16–19] is expected even under heating.
- Moreover, the PMA in amorphous CoFeB films could occur easily without any further heat treatment and with any kind of common substrate. From the technical viewpoint, this makes the fabrication process simple and less expensive.

This paper presents a systematic study of perpendicular magnetic anisotropy and the detailed magnetization process on the basis of in situ observation of the magnetic domain structure in a series of amorphous CoFeB multilayer films with changing magnetic layer thickness as well as number of CoFeB/Pd bilayers, in which the noble metal Pd is used for the spacing layers. The contribution of the interfacial magnetization and anisotropy to the PMA of the films is separated from the variation of the effective uniaxial anisotropy with the CoFeB thickness, which plays an essential role in the formation of PMA in such CoFeB-based multilayers.

2. Experiments

The multilayer films with Ta 2.0/Pd 2.0/(CoFeB t/Pd 1.0 nm)2 samples were grown on Si wafers with a native oxide layer by using a DC magnetron sputtering system (ULVAC UHV). The base pressure was better than 5.0 × 10−9 Torr and the plasma Ar pressure was kept at 1.0 mTorr. The sputter power was fixed at 30 W for the CoFeB target, 25 W for the Pd target and 50 W for the Ta target to ensure a very low deposition rate (~0.01–0.02 nm s⁻¹). The nominal composition of the CoFeB target was 40:40:20, as this composition is expected to possess the highest spin polarization and have the best glass-forming ability among the amorphous ferromagnetic Co–Fe–B alloys [20, 32]. The underlayer Ta 2.0/Pd 2.0 nm was selected to work as the buffer layer, for its good adhesion and good surface morphology. Atomic force microscopy imaging (data not shown) revealed a root mean square surface roughness of the Ta/Pd buffer layer of 0.20 nm. The top Ta layer (1.0 nm thick) was used as a cap layer to protect the films from ambient oxidation. Structural analysis was conducted using high resolution transmission electron microscopy (HRTEM) on an FEI Tecnai F20. Cross-section specimens for TEM imaging were prepared by conventional mechanical polishing and low energy Ar ion milling. Magnetic properties of the films were measured using a vibrating sample magnetometer (VSM) with a maximum field of 2.0 T (Lakeshore 7400 VSM). Magnetization-reversal processes in the films were observed, in terms of magnetic domain, by polar Kerr microscopy with an out-of-plane field up to 140 mT [33].

3. Results and discussion

An HRTEM cross-section image of the (CoFeB 1.2 nm/Pd 1.0 nm)2 sample is illustrated in figure 1(a), as a typical example. It is clearly seen that the buffer layer, [Ta 2.0 nm/Pd 2.0 nm], appears partially crystalline, with fcc structure in the Pd layer, denoted by the diffraction spots in the Fourier transform of some selected areas in the layer (figure 1(b)). Natural formation of fcc structure in a Pd buffer layer is not unusual and it is commonly seen in multilayer film with Pd as the buffer layer [8]. Despite being grown on a partially polycrystalline buffer layer, the main magnetic CoFeB and nonmagnetic Pd layers are obviously amorphous with well-defined layers, represented by a halo diffractogram in the Fourier transform image (figure 1(c)). Due to the amorphous structure, the interfaces between the CoFeB and Pd layers probably do not look very sharp, but they can be seen, showing layer structure in the films studied.
the film when the thickness reaches 1.2 nm. It is interesting to note that the PMA of the film plane. The magnetization appears to align in the plane of the film. Figure 2(a) shows the magnetic hysteresis loops of the [CoFeB 0.4 nm/Pd 1.0 nm]_{10} films (n = 2–20) measured with a perpendicular magnetic field.

Figure 2 shows the magnetic hysteresis (M–H) loops of CoFeB/Pd multilayers with variation of the CoFeB thickness (a) and variation of the number of CoFeB/Pd bilayers (b). The inset of figure 2(a) shows the M–H loops for a typical sample (CoFeB 0.4 nm/Pd 1.0 nm)_{10} measured in the plane and out of the plane of the film. Figure 2(a) indicates that the PMA has been established, by the rectangular hysteresis loop, in the thin films with low thickness of the CoFeB layer (lower than 1.2 nm), as measured with the applied field parallel to the film plane. The magnetization appears to align in the plane of the film when the thickness reaches 1.2 nm. It is interesting to note from figure 2(b) that the PMA still exists in the multilayer with 20 bilayers of CoFeB/Pd with the 0.4 nm CoFeB layers.

From the hysteresis loops, two of the most important magnetic parameters, namely the saturation magnetization, \(M_s\), and the effective uniaxial anisotropy, \(K_{\text{eff}}\), are deduced. By overlapping the hysteresis loops measured in two directions (in the plane and out of the plane), the saturation field, \(H_s\), is determined as the intercept of the two loops (see the inset in figure 2(a)). As a result, the anisotropy field, \(H_k\), is obtained, as [8, 14]

\[
H_k = H_s + 4\pi M_s, \tag{1}
\]

and the effective uniaxial anisotropy can be calculated from [8, 14]

\[
K_{\text{eff}} = \frac{1}{2} H_k \times M_s. \tag{2}
\]

Figure 3(a) illustrates the saturation magnetization, \(M_s\), as a function of the CoFeB layer thickness, \(t\), for the films with 10 CoFeB/Pd bilayers. Obviously, \(M_s\) increases with the CoFeB thickness, from 400 emu cm\(^{-3}\) to 820 emu cm\(^{-3}\), as a linear function, like in a previous study on a PMA CoFeB film with MgO interfaces [14]. The increase in \(M_s\) here is ascribed to the increase of the volume magnetic moment that occurs when increasing the thickness of the CoFeB layer. Increasing the number of CoFeB/Pd bilayers (detailed data are not shown here, but an example can be seen in figure 2(b)) also yields a similar tendency in the saturation magnetization, which is similar to the previous results for amorphous CoFeB multilayers published elsewhere [21]. The lowest \(M_s\) value of 200 ± 28 emu cm\(^{-3}\) is found in the sample with two bilayers of CoFeB/Pd at a CoFeB thickness of 0.4 nm. Please note that a strong PMA film with such a low \(M_s\) is desirable for spin-transfer torque devices, because a high \(K_{\text{eff}}\) and low \(H_k\) of the spin polarizer layer are the key factors for reducing the spin-switching current density [7–10, 16–18]. Qualitatively, the magnetization in the films could be considered as two terms: the volume magnetization and the surface/interface magnetization. As the number of CoFeB/Pd bilayers is fixed and the CoFeB thickness is changed, the volume term will vary, whereas the surface/interface term can be assumed to be unchanged. These assumptions allow the surface/interface magnetization to be estimated from the linear dependence of the total magnetization of the CoFeB. By extrapolating the linear plot to zero thickness (the volume term reduces to zero), this zero-thickness magnetization can be found and this will represent the surface/interface term. In the case presented in figure 3(a), the value of the surface/interface magnetization of the CoFeB/Pd interfaces is deduced to be 180 ± 10 emu cm\(^{-3}\). This value is mostly invariant when the number of CoFeB/Pd bilayers increases from 2 to 20.

On fixing the number of CoFeB/Pd bilayers, the effective uniaxial anisotropy, \(K_{\text{eff}}\), decreases with increasing CoFeB thickness. Namely, for the multilayers with \(n = 10\), \(K_{\text{eff}}\) decreases from 7.7 × 10^6 J m\(^{-3}\) for \(t = 0.4\) nm, to
The effective anisotropy $K_v$ can be understood as two terms: the volume anisotropy, $K_v$ and the surface/interface anisotropy, $K_s$ [34]. The $K_v$, contributed by the volume magnetic moment, is negative and tends to align the magnetization in the plane of the film. In contrast, the $K_s$, arising from the surface and interfacial magnetic moments, is positive and directs the magnetization perpendicular to the film plane. Competition between these terms determines the sign of the effective anisotropy: positive for out-of-plane anisotropy, and negative for in-plane anisotropy. The relation between these terms is expressed by equation [34]:

$$K_{\text{eff}} = K_v + 2K_s/t, \quad (3)$$

where $t$ is the thickness of the CoFeB layer. This relation can be rewritten as

$$K_{\text{eff}} \cdot t = K_v \cdot t + 2K_s. \quad (4)$$

From (4), the anisotropy terms, $K_v$ and $K_s$, can be separated from the $K_{\text{eff}}$ in terms of the linear dependence of $K_{\text{eff}}/t$ on the thickness ($t$). That is, figure 3(b) plots the linear dependence of $K_{\text{eff}}/t$ on the thickness ($t$) for the multilayer system with $n = 10$ and CoFeB thickness varying from 0.4 nm to 1.2 nm. The $2K_s$ value is determined as the intercept of the linear dependence with the vertical axis, whereas the $K_v$ is the slope of the linear dependence. In this case, a $K_v$ value of $2.44 \times 10^{5}$ Jm$^{-3}$ and a $K_s$ value of $-4.16 \times 10^{3}$ Jm$^{-3}$ are deduced from the plot. This also allows determining the critical thickness of the CoFeB layer, for which the magnetic anisotropy switches from a perpendicular one (positive sign) to an in-plane one (negative sign), as $t_c = -K_v/K_s = 1.17 \pm 0.01$ nm.

Applying this approach to all the other multilayers with different numbers of CoFeB/Pd bilayers, the surface/interface anisotropy is found to change with the CoFeB/Pd bilayer numbers. Figure 4 shows the variation of the surface/interface anisotropy as a linear function of the CoFeB/Pd bilayer number ($n$). It is obvious that the increase in the number of CoFeB/Pd bilayers results in increase of the CoFeB/Pd interfaces and this logically gives rise to an enhancement of the interfacial anisotropy, as observed. All these quantifications of the surface/interface anisotropy and interfacial component of the $M_r$ confirm that the PMA in the CoFeB/Pd multilayers studied predominantly originates from the surface/interfacial magnetic moments of the CoFeB/Pd interfaces due to the broken symmetry of the ultrathin layer structure, as predicted previously [35]. Additionally, previous studies suggested that these magnetic moments might arise from the magnetic polarization of Pd 4d caused by hybridization with the Co and Fe 3d ions [8, 36]. Besides this, the use of the noble metal Pd as the spacing layers could improve the film further, if it is being used in spin-electronic devices, because of the long spin-diffusion length of Pd [37, 38] and the lower level of structural diffusion to magnetic layers required to achieve stable PMA owning to the high melting temperature of Pd. Generally, the requirement of fcc structure with (1 1 1) texture perpendicular to the film plane in the layers of Co-based (or CoFe-based) multilayers has been proved to be the first condition for the formation of PMA [2–8]. However, the results in this article (and some recent experimental works [26–28]) have demonstrated that the structural condition is not a necessary criterion for the CoFeB/Pd interfaces. This appears to be a physically interesting aspect merit a deep analysis of the interfacial magnetic moment and electronic structure of the CoFeB/Pd interfaces by a number of state-of-the-art techniques, e.g. x-ray magnetic circular dichroism [39, 40] and neutron diffraction [41, 42] investigations.

To understand the magnetization-reversal mechanism of the films, in situ observation of the magnetic domain structure in the films during a magnetization-reversal process was performed using polar Kerr effect microscopy supplemented with hysteresis loops measurements [33]. Figure 5 presents a reversal process in the multilayer sample [CoFeB/Pd], with a CoFeB layer thickness of 0.4 nm. Firstly, the film is magnetically saturated in a maximum positive field of 140 mT applied perpendicular to the film plane. Then, the reversal starts, with reversing the out-of-plane field from the maximum positive field to a negative field of $-3.5$ mT. At this point, reversed domains with opposite directions of magnetization denoted by dark spots on the bright background (figure 5(a)) are nucleated. Upon slightly increasing the negative field (in magnitude) to $-3.9$ mT, these domains start growing in the opposite direction, as seen in (figure 5(b)), where the dark spots increase their size quickly. Continuing increasing the negative field (in magnitude) to the coercive field ($-4.1$ mT) makes the reversed domains, with dark contrast, increase their size (domain walls are propagating). At this field, net zero magnetization could be visible via the same areas of the bright domains (positive direction of the magnetization) and the dark domains (negative direction of the magnetization). When the negative field increases (in magnitude) slightly further (to about $-4.3$ mT), the dark region expands, becoming dominant in the film and the reversal process mostly completes (figures 5(d) and (e)). A square-shaped hysteresis loop (figure 5(f)) is also deduced from

![Figure 4. Surface/interface anisotropy, $K_s$, extracted from the effective uniaxial anisotropy as a function of the number of CoFeB/Pd bilayers.](image-url)
the MOKE images and shown to be similar to that obtained by the VSM. The same domain nucleation and movement revolution is also observed in the other samples but with a slightly different pinning effect. Upon increasing the CoFeB/Pd bilayer number, the pinning effect becomes stronger, as denoted by a slow change in the magnetization in the ‘pinning regime’ during the reversal (see figure 6).

The variation of the domain structure here shows a typical reversal process on the easy axis, in which the reversal is governed by the nucleation of magnetic domains and domain wall movement. It is noteworthy that the growth of the domains and the movement of the domain walls during the reversal seem to be steady, indicating a weak pinning effect on the propagation of the walls, as expected for the amorphous film. Similar domain structure and reversal behavior were observed previously for other PMA CoFeB films [43]. The growth of nearly circular reversed domains during the reversal presumably indicates random distribution of pinning sites as reported and discussed in a previous work [43]. As the layers are amorphous, the domain walls are essentially pinned by the pinning sites located in the interfaces rather than in the layers. Turning back to the HRTEM cross-section image (figure 1), the films appear with very smooth interfaces and this results in very weak pinning on the wall movement. This aspect looks to be very promising as regards application in domain-wall-controlled spin-transfer torque devices.

Furthermore, the nucleation of the reversed domains during the reversal seems to be different for different numbers of CoFeB/Pd bilayers. Figure 7 depicts the domain patterns of some representative samples (the thickness of the CoFeB layers here is 0.4 nm) for the state where the reversed domains are nucleated. Apparently, the number of reversed domains increases with the number of CoFeB/Pd bilayers, which indicates significant importance of the interfaces in the films. This suggests that the reversed domains are essentially nucleated at the interfaces where defects would locate, supporting the nucleation [43].

4. Conclusions and outlook

We presented a systematic investigation of perpendicular magnetic anisotropy in amorphous CoFeB/Pd multilayer films. Strong PMA can be established for a large range of CoFeB
thicknes up to 1.0 nm with controllably low saturation magnetization (down to 200 emu cm\(^{-3}\)) and high effective uniaxial anisotropy (up to \(\sim 7.7 \times 10^6\) Jm\(^{-3}\)). The PMA vanishes upon increasing the CoFeB layer thickness to 1.2 nm. By varying the CoFeB layer thickness and the number of (CoFeB/Pd) bilayers, the contributions of the volume and surface/interface magnetization and anisotropy to the magnetism of the films are separated and quantified. The magnetization of the interfacial magnetic moment is estimated to be 180 \(\pm\) 10 emu cm\(^{-3}\). Large magnetic moment is estimated to be 180 \(\pm\) 10 emu cm\(^{-3}\).

The multilayer films studied using amorphous CoFeB layers are excellent candidates for applications ranging from spin-transfer torque devices to conventional perpendicular magnetic recording. With high out-of-plane anisotropy, very thin Bloch-type walls are expected to exist in the films (the wall width can be estimated to be below 5 nm using the very thin Bloch-type walls and low damping coefficient are desirable for creating spin-transfer torque devices working with low current density for the spin switching, to compete with other PMA thin films [13, 45]. As regards the magnetization reversal, the multilayer films studied would seem to have very good potential for application in domain wall spin-transfer torque devices, particularly because the interface-supported domain nucleation and steady movement of the domain walls would be fruitful for simple control of domain wall motion. The moderately high \(M_s\) in some samples would be an interesting aspect for perpendicular magnetic recording technology [46].

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