



Magnetic properties of single-layer and multilayer structured $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ thin films



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ABSTRACT

We report systematic investigations of thickness dependent magnetic properties of amorphous $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (x nm) single-layer films and effects of number of multilayers and thickness of spacer layer on the magnetic properties of multilayer structured $[\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (y nm)/Ta (z nm)] _{$n=1-3$} / $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (y nm) films prepared directly on thermally oxidized Si substrate using magnetron sputtering technique. All the as-deposited films at ambient temperature exhibit amorphous structure. For single-layer films, coercivity (H_C) and field required for saturation (H_S) increase gradually with increasing x from 10 to 30 nm, but exhibit a rapid increase when $x \geq 67$ nm. This behaviour was attributed to the change in the magnetic domain structure from in-plane magnetization to stripe domains caused by the development of effective magnetic anisotropy instigated by stress accumulated during the deposition. High temperature thermomagnetization data show the Curie temperature of $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (20 nm) film as 512 K. On the other hand, the introduction of thin Ta spacer layers in multilayer films helps reducing H_C and H_S substantially, but magnitude of the reduction depends strongly on the values of n and z . The increase of n diminishes the development of effective magnetic anisotropy in CoFeB film. This is due to the reduction in the thickness of the ferromagnetic layers, which changes the magnetic domain structure, and the optimum value of z enhancing interlayer coupling between CoFeB layers. Hence, the magnetic properties of the multilayer thin films are improved. The observed results are elucidated on the basis of change in the magnetic domain structure with increasing film thickness in single-layer films, and with increasing number of multilayers and spacer layer thickness dependent interlayer coupling in multilayer structured thin films.

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1. Introduction

A key trend in recent science and technology is the exploitation of phenomena occurring at nanoscales. This has led not only to the development of new fields of research, but also to the practical applications of nanotechnology. During the last few decades, the tendency to miniaturize dimensions of magnetoelectronic devices has created a demand for new materials and new methods for their production. With this connection, magnetic thin films with enhanced soft magnetic properties such as high saturation magnetization (M_S), low coercivity (H_C), large relative permeability, controllable magnetic anisotropy and good thermal stability are highly required for practical applications in various modern magnetoelectronic devices [1–7] and for scaling down the integrated circuits in such devices including consumer electronics. Hence, searches for magnetic thin films with different artificial structures have been carried out widely both for practical applications and for gaining fundamental knowledge on the effect of artificial structures in low-dimensional systems to control the resulting magnetic properties [8–

11] such as surface magnetism, interlayer magnetic coupling [12–15], surface anisotropy and different kinds of magnetoresistance [16]. These studies summarily reveal that soft magnetic properties of magnetic thin films exhibit a strong dependence on film thickness and growth conditions. The thickness dependent magnetic properties of diverse alloy films do not show unique behaviour, but vary strongly on the constituent elements [17–22]. Nevertheless, it has been reported that soft magnetic properties of the magnetic thin films are degraded at higher film thickness (≥ 100 nm) due to change in magnetic domain structure caused by the development of stress induced effective magnetic anisotropy [20,22–24]. Furthermore, these types of soft magnetic thin films display spin-reorientation transition from in-plane orientation of magnetization to out-of-plane multi-domain state or vice-versa either with changing film thickness or with changing substrate temperatures during sample preparation [22,24,25].

In order to control the development of effective magnetic anisotropy at larger film thickness and to improve the soft magnetic properties, the multilayer structured thin films having ferromagnetic layers separated by thin non-magnetic layers (metallic and non-metallic nature) were reported [26–30]. Nakagawa et al. reported that $[\text{Fe-Co-Ta:N}$ (50 nm)/Ti (10 nm)]₄ multilayered film exhibits relatively high permeability and better thermal stability as compared to the single-layer film of

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Fe-Co-Ta:N (200 nm) [26]. Similarly, Naoe et al. [27], Huang et al. [28], Nakamura et al. [29] and Mishra et al. [30] reported correlative study between a thick single-layer ferromagnetic film [Fe-Cu-Nb-Si-B (140 nm), HITPERM (100 nm), FeSiAl (200 nm), FeTaC (200 nm)] and multilayered film in the form of bilayer, trilayer and more multilayers by keeping total thickness of ferromagnetic layers at a constant value. Such works were carried out mainly to understand the role of spacer layers and number of multilayers on tuning the soft magnetic properties at higher thicknesses. The above studies evidently confirmed the advantage of multilayered films to improve the soft magnetic properties and its applicability in devices. However, various parameters such as growth conditions, surface morphology, interface roughness, microstructure, and number of multilayers and thickness of spacer layer are major concerns in optimizing the magnetic properties.

Recently, CoFeB based alloy thin film is found to be one of the pioneering materials for various applications [4,18,31,32] such as magnetic tunnel junctions (MTJ) [20], magnetic random access memory [33] and spin-logic based devices [34], as they exhibit extremely large tunnelling magnetoresistance (TMR) at room temperature in a stack structure of CoFeB/MgO(001)/CoFeB. Hence, extensive studies have been carried out on CoFeB films to understand magnetic domain structure [35], spin polarization [36], ferromagnetic resonance and damping properties [37] and corrosion properties [38]. However, it is revealed from the literature that most of the reported works on CoFeB films have focused mainly on tuning the magnetic properties of ultrathin films from application point of view and there are only a few reports available on the thickness dependent magnetic properties of CoFeB films from the fundamental point of view [39,40]. For instance, Kipgen et al. [39] reported the structural and magnetic properties of Co₂₀Fe₆₀B₂₀ films in the thickness range of 5–50 nm prepared by ion-beam sputtering. Chen et al. [40] presented the electric, magnetic, thermal and adhesive properties of amorphous Co₆₀Fe₂₀B₂₀ thin films over the thickness range of 100–500 nm prepared on glass substrates using DC magnetron sputtering technique. However, no systematic study has been carried out to understand the thickness dependent magnetic properties of CoFeB alloy films over a wide range of thickness prepared by a single preparation technique. Naik et al. [33] recently reported the effect of an ultra-thin Ta layer insertion in the CoFeB films on the magnetic and TMR properties of CoFeB-MgO system and showed that the effective magnetic anisotropy can be doubled and thermal stability was enhanced by a factor of 2.5 with Ta insertion. Sato et al. [41] have also reported the increase of thermal stability factor of MTJ by the insertion of Ta in CoFeB films. Nevertheless, the systematic investigation of effects of number of multilayers and thickness of spacer and ferromagnetic layers on the magnetic properties and interlayer coupling between the ferromagnetic layers of the multilayer film have not been well addressed so far. Hence, in this study, we report (i) thickness dependent magnetic properties of single-layer Co₄₀Fe₄₀B₂₀ (x nm) films in amorphous form over a wide range of thickness from 10 to 200 nm at first and then (ii) tuning the magnetic properties of Co₄₀Fe₄₀B₂₀ films at higher thicknesses by utilizing multilayered structure, as presented in literature [26–30], with a careful variation of thickness of ferromagnetic and spacer layers. We observed that the magnetic properties of amorphous Co₄₀Fe₄₀B₂₀ films are degraded with increasing film thickness beyond critical thickness due to the development of effective magnetic anisotropy caused by the stress quenched in during films' deposition. This results in a transition from in-plane orientation of magnetization to stripe domain structure and increases H_C and the field required for saturation (H_S) largely. On the other hand, the magnetic properties of multilayer films could be improved by the introduction of thin Ta spacer layer, but the improvement strongly depends on number of multilayers and thickness of spacer layer, which changes effective magnetic anisotropy, magnetic domain structure and interlayer coupling between ferromagnetic layers, respectively.

2. Experimental details

Amorphous Co₄₀Fe₄₀B₂₀ (CoFeB) single-layer films with different thicknesses ($x = 10$ to 200 nm) and the multilayer [CoFeB (y nm)/Ta (z)] _{n} /CoFeB (y nm) films with different thicknesses of Ta spacer layer and CoFeB ferromagnetic layer were deposited directly on thermally oxidized Si substrate using DC magnetron sputtering technique. All the films were deposited at ambient temperature. The base pressure of the chamber was better than 1×10^{-4} Pa. The sputtering Ar gas pressure for CoFeB and Ta layers was optimized at 1.33 Pa. The optimization of Ar gas pressure was done mainly by analysing the structural and variation in the magnetic properties of the single-layer and multilayer films. The distance between the target and the substrate was optimized at 0.1 m and the substrate was rotated uniformly for obtaining homogeneous film. The deposition rate was pre-calibrated using ex-situ surface profilometer (Veeco, Dektak 150 model) and found to be 1.2 Å/s and 0.5 Å/s for the deposition of CoFeB and Ta layers, respectively. The thickness of the individual CoFeB layer in multilayer films was controlled using the relation (as demonstrated in literature [26–30]): $y = 200 / (n + 1)$, where n is the number of multilayers varied between 0 and 3, and z is the spacer layer thickness controlled between 0 and 2 nm. Amorphous nature of the films was confirmed by X-ray diffraction (XRD) recorded using a high-power X-ray diffractometer (Rigaku TTRAX III, 18 kW) with Cu-K α radiation ($\lambda = 1.54056$ Å) and transmission electron microscopy (TEM, Jeol 2100 and Technai G2 F20) techniques. Magnetic properties of the films were analysed by using vibrating sample magnetometer (VSM, LakeShore Model 7410) by performing (i) magnetic hysteresis loops ($M-H$) at room temperature and (ii) thermomagnetization ($M-T$) measurements at a constant field over a temperature range of 300–650 K. Magnetic domain images and Kerr loops were obtained using magneto-optic Kerr effect (MOKE) microscopy (Evico Magnetics Ltd., Germany) technique. Imaging was performed using linearly polarized light with Xenon lamp as source. Magnetic domain images were observed in both branches of hysteresis cycle in longitudinal MOKE mode. Hysteresis accompanied by simultaneous imaging has been performed for magnetic fields applied along in-plane directions (easy and hard axes).

3. Results and discussion

Fig. 1 displays typical XRD pattern, bright-field TEM image and selected area electron diffraction (SAED) pattern of the as-deposited single-layer CoFeB (200 nm) film. It is clearly seen that as-deposited film exhibits only a broad peak at around $2\theta = 44^\circ$ without any other sharp peaks peculiar to any other crystalline phases. It may be noted that the XRD peak observed at $2\theta = 33.05^\circ$ represents Si(200) peak resulting from thermally oxidized Si substrate [42]. In addition, the bright-field TEM image and the SAED pattern reveal the plane and even contrast microstructure, devoid of any local lattice fringes and halo diffraction rings, respectively. These results confirm that the as-deposited CoFeB films exhibit amorphous structure.

Fig. 2 depicts the room temperature normalized in-plane $M-H$ loops of the CoFeB ($x = 10$ –200 nm) films. It is observed that (i) CoFeB films with $x < 30$ nm exhibit either rectangular shaped loop with remanence ratio of >75% or flat type loop. In addition, the loops saturate at lower applied magnetic fields. (ii) Upon increasing $x \geq 67$ nm, the loop shape changes into different nature, i.e., the $M-H$ loops are constituted by two distinct magnetization phases: (a) in-plane magnetic component, which reverses quickly at fields close to H_C and (b) perpendicular component, which rotates progressively under the application of magnetic field and results in almost a linear approach to saturation. Therefore, the value of H_S increases largely (>16 kA/m) [23,43,44]. This particular hysteresis loop shape has been referred to as a transcritical loop and correlated to the development of effective magnetic anisotropy caused by the stress induced during the deposition of the films at a higher deposition rate to form amorphous nature [22,23,25,30,45,46]. This leads to

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