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Effect of buffer layer and external stress on magnetic properties of flexible FeGa films

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We systematically investigated the effect of a Ta buffer layer and external stress on the magnetic properties of magnetostrictive $Fe_{81}Ga_{19}$ films deposited on flexible polyethylene terephthalate (PET) substrates. The Ta buffer layers could effectively smoothen the rough surface of PET. As a result, the FeGa films grown on Ta buffer layers exhibit a weaker uniaxial magnetic anisotropy and lower coercivity, as compared to those films directly grown on PET substrates. By inward and outward bending the FeGa/Ta/PET samples, external in-plane compressive and tensile stresses were applied to the magnetic films. Due to the inverse magnetostrictive effect of FeGa, both the coercivity and squareness of hysteresis loops for FeGa/Ta films could be well tuned under various strains. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4793602]

The magnetic and electrical properties of magnetic thin films are strongly affected by their film thickness, crystallinity, surface roughness, and mechanical stresses, properties which have been extensively investigated in recent years.[1,2] Buffer or seeding layers, such as Ta and Pt due to their chemical inertness, are often used to reduce the interfacial roughness,[3] induce an out-of-plane texture of antiferromagnetic layers,[4] and release the inherent stress of magnetic films.[5] Most of the previous works relating to the impact of buffer or seeding layers on the magnetic properties of ferromagnetic films were conducted on rigid substrates.[3,6] Over the last years, magnetic and spintronic systems grown on flexible substrates have gained a great amount of interest due to the exciting new applications provided by arbitrary surface geometries possible after fabrication.[7] When magnetostrictive materials, for example, FeGa alloys which exhibit low hysteresis, good tensile strength (~500 MPa), and large saturation magnetostriction (~350 ppm) at a low magnetic field (~100 Oe), are utilized in torque and stress sensors and actuators, the deformability of flexible substrates may enhance the response of magnetostrictive films to external mechanical stress.[8] Prior to fabricating flexible spintronic devices, an appropriate buffer layer needs to be introduced to decrease the roughness of flexible substrates, improve the crystal orientation of magnetic films, and release the residual stress. In this work, we investigated the effect of a Ta buffer layer and external stress on the magnetic properties of magnetostrictive FeGa films grown on flexible polyethylene terephthalate (PET) plastics.

FeGa/Ta bilayers were deposited on flexible PET substrates using a vacuum DC magnetron sputtering system with a base pressure of better than 6×10^{-7} Torr at room temperature. The thickness of magnetostrictive FeGa layer was selected at 20 and 50 nm, and the thickness of Ta buffer layer, t_{Ta}, was varied from 0 to 30 nm. All samples were protected from oxidation by a 5-nm-thick Ta capping layer. The surface morphologies of the samples were characterized by atomic force microscope (AFM, Veeco Dimension 3100V). The magnetic properties of FeGa films at various magnetic field orientations and under different external strains were measured by a vibrating sample magnetometer (VSM, Lakeshore 7410).

Figure 1(a) shows the surface topography for a bare PET substrate with a root-mean-square (RMS) roughness of 2.32 nm, which is obtained over a scan area of $2 \times 2\,\mu m^2$ by AFM. After directly growing the FeGa layers on PET, the film surface becomes rough and the RMS roughness for the 20-nm-thick FeGa/PET films becomes 2.72 nm, as shown in Fig. 1(b). With increasing the thickness of FeGa, because the FeGa layer itself may act as a buffer layer for the following FeGa deposition, the films become smooth and the RMS roughness of the films is slightly reduced to 2.60 nm for the 50-nm-thick FeGa/PET films, as shown in Fig. 1(c). Inserting a buffer layer of Ta between FeGa layers and PET substrates can effectively improve the growth condition and reduce the roughness of FeGa layers. For FeGa(50 nm)/Ta(10 nm)/PET films, the RMS roughness is significantly reduced to 2.04 nm, as shown in Fig. 1(d). With further increasing the thickness of Ta buffer layers, the roughness of the films is decreased slowly to a constant value, as shown in Fig. 1(e). Our AFM micrographs indicate that the Ta buffer layer is able to effectively reduce the roughness of magnetic films grown on flexible plastic substrates and may be applied in fabricating flexible magnetoelectric devices. It should be noted that the current PET has rather low processing temperatures, when applying at high temperature, the substrates

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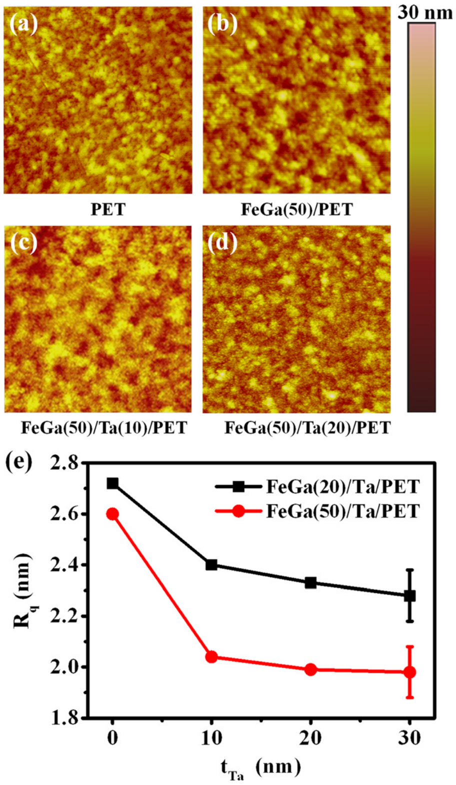

FIG. 1. AFM images ($2 \times 2\,\mu m^2$) for (a) a bare PET substrate, (b) FeGa(20 nm)/PET, (c) FeGa(50 nm)/PET, and (d) FeGa(50 nm)/Ta(10 nm)/PET films. The color contrast (dark to bright) for the height scale corresponds to 30 nm. (e) RMS roughness versus Ta buffer layer thickness for FeGa(20 nm)/Ta(t_{Ta})/PET and FeGa(50 nm)/Ta(t_{Ta})/PET films.

which can endure high temperature, such as polyimide or ultrathin metallic foils, need to be used.

The magnetic properties for the samples of FeGa(50 nm)/Ta(t_{Ta})/PET in an unstressed state are measured at different magnetic field orientations ψ with an increment of 5°. The hysteresis loop for FeGa(50 nm)/PET without Ta buffer layer ($t_{Ta} = 0$) with the applied magnetic field H parallel to the easy axis, i.e., $\psi = 0°$, indicates a relative squareness M_r/M_s of 0.99 and a coercivity H_c of 121 Oe, while the hysteresis loop measured along the hard axis, i.e., $\psi = 90°$, is sheared with a squareness of 0.25 and a coercivity of 30 Oe, as shown in Figs. 2(a) and 2(b). Both the angular dependence of M_r/M_s and H_c, as shown in Figs. 2(c) and 2(d), respectively, possess a uniaxial symmetry about the easy or hard axes of the films, which indicates a significant uniaxial magnetic anisotropy due to the residual stress caused by the slightly deformation of PET substrates.[9] With increasing the thickness of Ta buffer layer to 20 nm, M_r/M_s of hysteresis loop is slightly decreased to 0.89, and H_c for H along the easy axis is significantly reduced to 30 Oe, as revealed in Fig. 2(a). For H applied along the hard axis, the M_r/M_s ratio is increased to 0.52, and H_c is reduced to 14 Oe, as displayed in Fig. 2(b).

The strength of the uniaxial magnetic anisotropy, K_u, of the magnetic films can be obtained using the relation of $K_u = H_k M_s/2$,[10] where M_s is the saturation magnetization, and H_k is the saturation field determined by fitting the hysteresis curves measured along the hard axis. In our experiment, by increasing the thickness of Ta buffer layer from 0 to

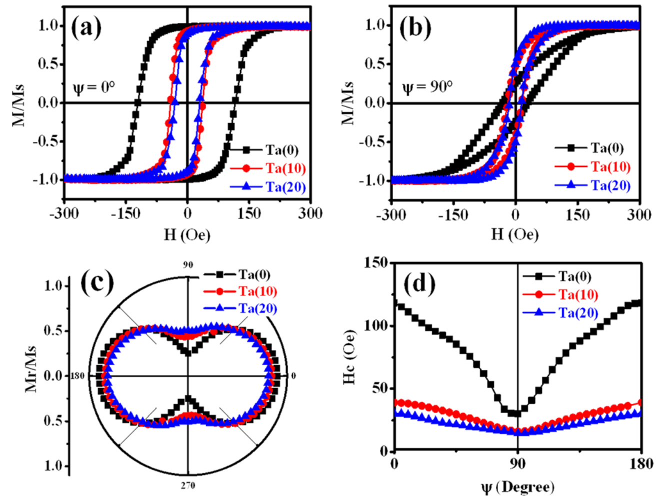

FIG. 2. Hysteresis loops for flexible FeGa(50 nm)/Ta(t_{Ta})/PET films with a magnetic field applied along (a) the easy ($\psi = 0°$) and (b) hard ($\psi = 90°$) axes, and the corresponding angular dependence of (c) squareness and (d) coercive field.

20 nm, H_k is reduced from 318 to 140 Oe. By using experimentally measured $M_s = 880\,\text{emu/cm}^3$, K_u is obtained to be $1.4 \times 10^5\,\text{erg/cm}^3$ for FeGa(50 nm)/PET and $6.16 \times 10^4\,\text{erg/cm}^3$ for FeGa(50 nm)/Ta(20 nm)/PET. With further increasing Ta thickness, K_u is decreased slowly to a constant value. The uniaxial anisotropy found in our flexible magnetic films is due to the residual stress caused by the slightly inevitable deformation of PET substrates.[9] Our experimental observations indicate that the Ta buffer layer could effectively release the residual stress in PET substrates, and therefore reduce the strength of the uniaxial anisotropy of FeGa layers. The decrease of the coercivity of FeGa films may result from both the decrease of uniaxial anisotropy and the flatness of the films.

In order to investigate the magnetic properties of the magnetostrictive FeGa films under the external mechanical stresses, the samples are inward or outward bended along the easy and hard axes to generate compressive and tensile stresses. During the VSM measurement, the magnetic field,

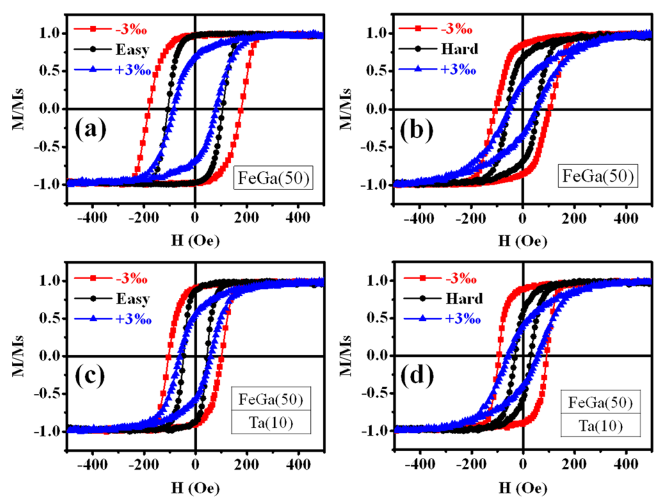

FIG. 3. Hysteresis loops for FeGa(50 nm)/PET taken with magnetic field along the (a) easy and (b) hard axes and FeGa(50 nm)/Ta(10 nm)/PET with magnetic field along the (c) easy and (d) hard axes under compressive and tensile strains.

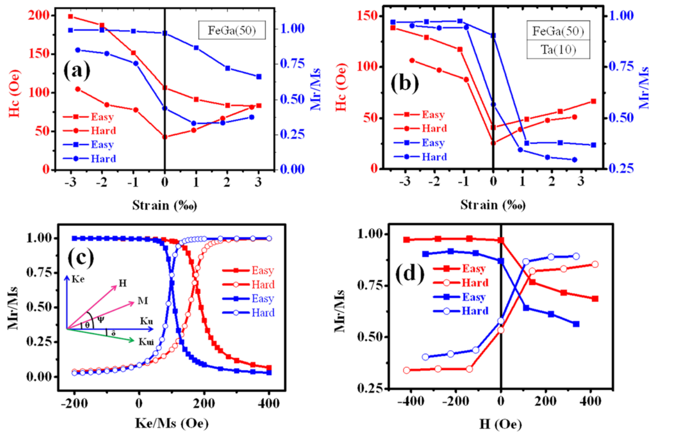

FIG. 4. Summary for the strain dependence of H_c and M_r/M_s with H applied along the easy and hard axes in (a) FeGa(50 nm)/PET and (b) FeGa(50 nm)/Ta(10 nm)/PET films. (c) Using the modified Stoner-Wohlfarth model and the anisotropy geometry in the inset of (a), the M_r/M_s of hysteresis loops for FeGa(50 nm)/PET (red lines) and FeGa(50 nm)/Ta(10 nm)/PET (blue lines) as a function of the strain-induced uniaxial anisotropy K_e/M_s can be calculated. (d) Using this simulation definition, the experimental results can be accordingly revised to the squareness as a function of K_e/M_s.

H, is applied perpendicular to the bending direction to ensure the magnetic field is parallel to the surface of films. The magnetostrictive strain, ε, can be calculated using $\varepsilon = T/2\rho$ with T and ρ being the thickness of the sample and the curvature radius of the bended substrate, respectively. The stress, σ, is given by $\sigma = \varepsilon E_f/(1-\nu^2)$, where E_f and ν denote Young's module and Poisson ratio of the FeGa films, respectively. The stress σ is calculated using $E_f = 60$ GPa for FeGa films with a Poisson ratio ν of 0.3. Here, ε and σ are considered to be positive for the outward/tensile bending and negative for the inward/compressive bending. For FeGa(50 nm) films, when H is applied along the easy axis, a tensile strain increasing from 0% to 0.3% gives rise to a drastic decrease of M_r/M_s from 0.97 to 0.66, and a significant decrease of H_c from 107 to 83 Oe. In contrast, under a compressive strain of -0.3%, M_r/M_s is increased to 0.99, and H_c is increased to 199 Oe, as shown in Fig. 3(a). For H oriented along the hard axis, M_r/M_s can be tuned from 0.44 at the unstressed state to 0.37 under a tensile strain of 0.3%, and to 0.85 under a compressive strain of -0.3%. Meanwhile, H_c is increased from 43 to 105 Oe under a tensile stain of 0.3%, and to 81.5 Oe under a compressive strain of -0.3%, as shown in Fig. 3(b). The similar mechanical strain dependence of the M_r/M_s ratio and H_c can be also found in FeGa(50 nm)/Ta(10 nm) films, as shown in Figs. 3(c) and 3(d). Figures 4(a) and 4(b) show the summary for the external strain dependence of H_c and M_r/M_s for the samples of FeGa(50 nm) and FeGa(50 nm)/Ta(10 nm) respectively. The FeGa films grown with and without Ta buffer layers exhibit similar mechanically tunable magnetic properties. For H applied parallel to the easy or hard axes of the magnetostrictive films, tensile strain perpendicular to H leads to the decrease of M_r/M_s ratio of hysteresis loops, while compressive strain gives rise to an enhancement of M_r/M_s. Our experimental observations demonstrate that the Ta layers are a good buffer to adhere both the PET plastic and the FeGa films so that the mechanical stresses caused by the deformation of substrates could be effectively transferred to the magnetostrictive films.

In order to interpret our experimental results, we apply a modified Stoner-Wohlfarth model developed in our previous work to account for the mechanically tunable magnetic properties.[9] Considering our samples not a perfect system with a uniaxial magnetic anisotropy, we speculate that the uniaxial anisotropy has a distribution along the easy axis. As a result, we can apply a theoretical model to estimate the total energy for a grain in FeGa film: $E = -K_{ui}\cos^2(\theta-\delta) + K_e\cos^2\theta - MH\cos(\theta-\psi)$, where K_{ui} denotes an arbitrary grain with a uniaxial anisotropy orienting at an angle of δ with respect to the orientation of K_u, and θ is the angle between K_u and the magnetization M, as shown in the inset of Fig. 4(c). The changes of magnetoelastic energy of FeGa/Ta films caused by the various strains of substrates bending lead to an equivalent uniaxial anisotropy K_e, which can be calculated by $K_e = -3/2\lambda_s\sigma$. Based on the modified Stoner-Wohlfarth model, we obtain the hysteresis loops predicted under various K_e by using $\delta = 5°$. As shown in Fig. 4(c), M_r/M_s is decreased for $\psi = 0°$ and increased for $\psi = 90°$ with increasing K_e/M_s. Considering the different definition of the geometries of the stress and uniaxial anisotropy, the strain dependence of M_r/M_s for FeGa(50 nm) and FeGa(50 nm)/Ta(10 nm) films can be accordingly revised to M_r/M_s as a function of K_e/M_s using $\lambda_s = 53$ ppm for FeGa(50 nm) and 43 ppm for FeGa(50 nm)/Ta(10 nm), respectively, as plotted in Fig. 4(d). It is obvious that the simulations could nicely interpret the mechanically tunable squareness of hysteresis loops for flexible magnetostrictive films with and without Ta buffer layer.

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