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Mechanically tunable magnetic properties of Fe$_{81}$Ga$_{19}$ films grown on flexible substrates

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We investigated on magnetic properties of magnetostrictive Fe$_{81}$Ga$_{19}$ films grown on flexible polyethylene terephthalate (PET) substrates under various mechanical strains. The unstained Fe$_{81}$Ga$_{19}$ films exhibit a significant uniaxial magnetic anisotropy due to a residual stress in PET substrates. It was found that the squareness of hysteresis loops can be tuned by an application of strains, inward/compressive or outward/tensile bending of the films. A modified Stoner-Wohlfarth model with considering a distribution of easy axes in polycrystalline films was developed to account for the mechanically tunable magnetic properties in flexible Fe$_{81}$Ga$_{19}$ films. These results provide an alternative way to tune mechanically magnetic properties, which is particularly important for developing flexible magnetoelectronic devices. © 2012 American Institute of Physics.

FeGa magnetostrictive alloys exhibit moderate magnetostriction (~350 ppm for Ga content of 19%) under very low magnetic field (~100 Oe) but good mechanical properties. Due to the great promise as engineering materials for applications in sensors and actuators, the magnetomechanical characteristics of FeGa alloys, including the magnetization and magnetostrictive responses to magnetic fields have been extensively investigated under applied tensile or compressive stresses. If the excellent magnetomechanical behaviors of magnetostrictive alloys can be achieved in thin films, they are possibly applied in magnetic microelectromechanical systems, and also important for developing functional materials, i.e., layered multiferroic composites. Most of previously studied magnetostrictive thin films and spintronic devices are deposited on stiff and thick substrates. Due to the significant clamping effect caused by substrates, when magnetostrictive films applied in micro-force sensors and multiferroic composited materials, their key performances, such as the strain sensitivity and the magnetoelectric coupling, are strongly depend on the magnetomechanical behaviors of magnetostrictive layers, and therefore are drastically reduced. Recently, flexible magnetic films and spintronic devices grown on plastic substrates which can be shaped into almost any arbitrary geometry have attracted much attentions. The deformability of substrates may partially eliminate the substrate clamping and enhance the response of magnetostrictive films to external mechanical stress. So far, only few works systematically studied the stress dependence of the hysteresis loop properties for magnetic films and heterostructures. The magnetomechanical behaviors of magnetostrictive FeGa films especially grown on flexible plastic substrates are not well known. Here, we fabricated magnetostrictive FeGa films on flexible polyethylene terephthalate (PET) substrates. Due to the residual stress of the flexible substrates, a uniaxial magnetic anisotropy is observed in the as-grown FeGa films. The magnetic properties can be significantly tuned with the application of strain by directly inward/compressive or outward/tensile bending of the films. Taking into account the distribution of the uniaxial anisotropy, the mechanically tunable magnetic behaviors can be qualitatively interpreted by a modified Stoner-Wohlfarth model.

Fe$_{81}$Ga$_{19}$ films with a thickness of 150 nm were deposited on both PET and Si substrates by radio frequency magnetron sputtering at room temperature. Before the substrates were transferred into the sputtering chamber, they were cleaned in ethyl alcohol using ultrasonic agitation for 15 min. The base pressure of the sputtering chamber was below 6.0 × 10$^{-5}$ Pa. During deposition, the argon flow was kept at 50 sccm and the pressure was set at 1.0 Pa. An evaporation rate of 2.0 nm/min was used for the growth of Fe$_{81}$Ga$_{19}$ alloy. Prior to be taken out of the vacuum chamber, Fe$_{81}$Ga$_{19}$ films were capped by a 5 nm Au layer to avoid oxidation. The surface morphology of the films was characterized by atomic force microscopy (AFM) using Veeco Dimension 3100 V. The angular dependence of hysteresis loops was measured using vibrating sample magnetometer (VSM, Lakeshore 7410) at room temperature.

Figures 1(a) and 1(b) show the AFM patterns for a bare PET substrate and Fe$_{81}$Ga$_{19}$/PET film, respectively. The root mean square (RMS) roughness of 2.16 nm for the PET substrate is much larger than that of 0.60 nm for the thermally oxidized Si substrate. For the fabrication of flexible magnetoelectronic devices, the surface roughness of the flexible substrate is required to decrease to a low value close to that...
of Si wafers by means of spin-coating a photoresist buffer layer onto the plastic substrates. In our works, Fe81Ga19 layers are directly grown on PET substrates, the corresponding AFM image confirms the growth of polycrystalline Fe81Ga19 films with an RMS roughness of 3.34 nm. Due to the rough morphology of both PET substrates and Fe81Ga19 films, the coercive field of the as-grown Fe81Ga19/PET films is obviously larger than that of the reference samples grown on oxidized Si substrates.

In order to study the magnetic properties of Fe81Ga19/PET films at unstressed state, the hysteresis loops were measured at room temperature with a magnetic field applied parallel to the film plane. The field orientation \( \phi \) is varied from 0\(^\circ\) to 360\(^\circ\) by rotating the sample with an increment of 10\(^\circ\), as shown in the inset of Fig. 2(a). The hysteresis loop for the field applied along the easy axis, i.e., \( \phi = 0^\circ \), is relative square with a \( M_s/M_r \) ratio of 0.86, while the hysteresis loop along the hard axis, i.e., \( \phi = 90^\circ \), is sheared with a squareness of 0.37, as shown in Fig. 2(a). The squareness and coercivity as a function of \( \phi \) are illustrated in Fig. 2(b). Both of them possess a uniaxial symmetry about the easy or hard axes of Fe81Ga19/PET films, which indicates a significant uniaxial magnetic anisotropy, but not the magneto-crystalline anisotropy in Fe81Ga19/PET films due to the polycrystalline structure. In contrast, the uniaxial anisotropy for the reference samples grown on oxidized Si substrates is too weak to be observed. Therefore, we ascribe the uniaxial anisotropy of the flexible Fe81Ga19 films to the residual stress caused by the slightly inevitable deformation of PET substrates. Due to the adhesion of both mechanical interlocking and chemical bonding between Fe81Ga19 films and PET substrates, the residual stress and the applied stress could be effectively transferred from flexible substrates to magnetostrictive films. The strength of the induced uniaxial anisotropy, \( K_u \), is quantitatively evaluated by calculating the difference between works done in magnetization along different directions. Using the relation \( K_u = \frac{1}{2} \left( M_s - \frac{1}{M_s} \right) HdM \) and the initial \( M-H \) curves along both the easy and hard axes shown in Fig. 2(a), \( K_u \) is obtained to be 1.81 \( \times 10^5 \) erg/cm\(^3\), which is comparable to the magneto-crystalline anisotropy of Fe81Ga19.

Due to the inverse magnetostrictive effect, i.e., the Villari effect, the magnetic properties of the magnetostrictive materials is sensitive to the external mechanical stress. Thanks to the deformability of PET substrates, the compressive and tensile strains can be applied on Fe81Ga19 films by inward or outward bending the PET substrates. The hysteresis loops for Fe81Ga19/PET films under various strains are measured by slightly bending the substrates along the easy or hard axes of Fe81Ga19 films. In order to restrict the magnetic field parallel to the film plane, during the VSM measurement, the magnetic field, \( H \), is applied perpendicular to the bending direction, i.e., the bending strain, as shown in the inset of Fig. 3. The strain, \( \varepsilon \), and the stress, \( \sigma \), are evaluated by using the relations \( \varepsilon = \varepsilon_0 / 2 \) and \( \sigma = E \varepsilon_0 (1 - \nu^2) \), respectively, where \( t \) is the thickness of the substrate including the film thickness, \( \rho \) is the curvature radius of the substrate after bending, \( E \) is Young’s modulus, and \( \nu \) is the Poisson ratio. \( \varepsilon \) and \( \sigma \) are considered to be positive for the outward/tensile bending and negative for the inward/compressive bending. During the magnetic measurements, \( \varepsilon \) is applied within a maximum value of 0.78\%, which corresponds to \( \sigma = 0.51 \) Gpa by using \( E = 60 \) GPa for Fe81Ga19 and the typical value of \( \nu = 0.3 \) for metals. When the magnetic field is applied along the easy axis of the films, a tensile strain along the hard axis increasing from 0\% to 0.78\% gives rise to a drastic decrease in \( M_r/M_s \) ratio from 0.86 to 0.29, as shown in Fig. 3(a). In contrast, under a compressive strain of −0.26\%, the \( M_r/M_s \) ratio is increased to 0.89. With further increasing the compressive strain, both the squareness and coercivity become hard to be improved, as shown in Fig. 3(b). For the magnetic field oriented along the hard axis, the \( M_r/M_s \) ratio can be tuned from 0.37 at an unstressed state to 0.19 under a tensile strain of 0.78\% and to 0.79 under a compressive strain of −0.78\% applied along the easy axis, as shown in Figs. 3(c) and 3(d), respectively. The external strain dependence of squareness is summarized in Fig. 4(a). For our measurement configurations, the tensile strain leads to a decrease in \( M_r/M_s \) ratio, but the compressive strain increases this value. It should be noted that, considering the contribution on the magnetoelastic energy and the induced magnetic anisotropy, the effect of a tensile strain applied along the easy axis is equivalent to that of a compressive strain along the hard axis and vice versa. Our results suggest that the easy axis for a flexible magnetostrictive film can be tuned to the hard axis under a tensile strain applied along the hard axis or a compressive strain along the easy axis, meanwhile the hard axis of the films is changed to the easy axis. In addition, we have prepared Fe81Ga19/PET films with various thicknesses down to 10 nm. They exhibit the similar residual stress-induced uniaxial anisotropy and the similar mechanically tunable magnetic properties.
We intuitively ascribe the mechanically tunable $M_s/M_s$ ratio to an additional uniaxial anisotropy induced by the various external strains. In our experiments, the strain is either along the easy axis or along the hard axis of Fe$_{81}$Ga$_{19}$ films, that is, the external strain-induced uniaxial anisotropy, $K_{\varepsilon}$, is collinear or perpendicular to $K_u$. We consider such a system with a uniaxial anisotropy of $K_u/M_s = 120$ Oe which is close to the experimental observation for our Fe$_{81}$Ga$_{19}$ films. The tensile stress $+\sigma$ and the compressive stress $-\sigma$, which are applied perpendicular to the easy axis of $K_u$, give rise to a positive $K_u$ with the easy axis perpendicular to $K_u$ and a negative $K_u$ with the easy axis along $K_u$, respectively, as schematically shown in the inset of Fig. 4(b). Regardless of the strength of $K_u$, the hysteresis loop predicted by Stoner-Wohlfarth model$^{18}$ for $\varphi = 0^\circ$ or $\varphi = 90^\circ$ is either a perfect rectangle with a squareness of one along the easy axis or a sheared one with the remanence of zero along the hard axis. The squareness switched between 1 and 0, i.e., the easy axis changed to the hard axis or vice versa, occurs at $K_u = K_e$, as shown in Fig. 4(b). As a result, based on the magnetization reversal mechanism of coherent rotation, we cannot theoretically draw the conclusion that the external stresses applied along or perpendicular to $K_u$ could tune the squareness of hysteresis loops for magnetostrictive films.

For the as-grown Fe$_{81}$Ga$_{19}$ films, we notice that the squareness of the hysteresis loop is less than one along the easy axis but higher than zero along the hard axis [see Fig. 2(a)]. The experimental observation indicates that our
samples are essentially not a perfect uniaxial system. Therefore, it is reasonable to speculate that the uniaxial anisotropy for the grains in polycrystalline Fe$_{81}$Ga$_{19}$ films does not strictly orient in an identical direction, but has a distribution along its average direction, that is, $K_u$. Considering an arbitrary grain with a uniaxial anisotropy $K_u$, orienting at an angle of $\delta$ with respect to the orientation of $K_u$, an external stress $\sigma$ is applied perpendicular to $K_u$. Thus, the total energy for a grain in Fe$_{81}$Ga$_{19}$ films can be written as: $E = -K_u \cos^2(\theta - \delta) + K_e \cos\theta - M_H \cos(\theta - \phi)$, where $\theta$ is the angle between $K_u$ and the magnetization vector M. We assume that the anisotropy constant $K_u$ for each grain is identical to $K_u$, and the angle of $\delta$ for the distribution of $K_u$ is ranged from $-10^\circ$ to $10^\circ$ with respect to $K_u$. Due to the distribution of the easy axis, the average value $K_{av}$ for the whole film is only less than $K_u$, about one percent, which can be neglected in our numerical calculations. Using an intermediate value of $\delta = 5^\circ$, we obtain the hysteresis loops predicted under various $K_e$ by means of Stoner-Wohlfarth model. As shown in Fig. 4(c), the simulated hysteresis loops for $H$ along the easy axis of $K_u$, i.e., $\phi = 0^\circ$, indicate that when $K_u/M_s$ caused by a tensile stress is varied from 100 to 150 Oe, the squareness can be obviously reduced from 0.91 to 0.29 and the coercivity is slightly decreased from 30 to 21 Oe. For $H$ along the hard axis of $K_u$, i.e., $\phi = 90^\circ$, the squareness of the hysteresis loops is enhanced from 0.41 to 0.96, and the coercivity is increased from 21 to 46 Oe, as shown in Fig. 4(d). We calculated the $K_u/M_s$ dependence of squareness for $\delta$ distributed from $-10^\circ$ to $10^\circ$, the average values are displayed in Fig. 4(b). With increasing $K_u/M_s$, i.e., $\sigma$, the squareness is drastically decreased for $\phi = 0^\circ$ and is increased for $\phi = 90^\circ$, especially around the critical value of $K_e = K_u$, where $K_u$ is compensated by $K_e$. Our numerical calculations are in good agreement with the simulations done in Ni films using a micromagnetic model including the magnetoelastic energy term into the Landau-Lifshitz-Gilbert equation.\(^\text{19}\) It should be noted that to facilitate the simulations and discussions, we utilize the uniform anisotropy geometry for our calculations, which is different from the experimental configurations. In experiment, the strain of substrates bending changes the magnetoelastic energy of the films, producing an equivalent uniaxial anisotropy $K_e$. Employing the definition for simulation shown in the inset of Fig. 4(b), the strength of $K_e$ can be written as $K_e = -3/2 \lambda \sigma$, where $\lambda$ is the magnetostriction constant for polycrystalline Fe$_{81}$Ga$_{19}$ films. Using $\lambda = 100$ ppm,\(^\text{12}\) the experimental strain dependence of squareness for Fe$_{81}$Ga$_{19}$/PET films can be accordingly revised to the squareness as a function of $K_u/M_s$, as plotted in the inset of Fig. 4(a). Obviously, our simulations based on the modified Stoner-Wohlfarth model are able to nicely interpret the mechanically tunable squareness of hysteresis loops for Fe$_{81}$Ga$_{19}$ films. The discrepancy between our experimental results and simulations may arise from the uncertain coefficients used for calculations and also from the irreversible behaviors involving domain wall motion and incoherent rotation especially, while the magnetic field was applied around the easy axis.\(^\text{20}\) The distribution of easy axes, which likely come from the inhomogeneous residual stress in Fe$_{81}$Ga$_{19}$ films due to the flexible PET substrates, is the key factor for the occurrence of the experimental observations.

In summary, we fabricated magnetostrictive Fe$_{81}$Ga$_{19}$ films on flexible PET substrates, in which a significant uniaxial magnetic anisotropy is observed for the unstressed state. The hysteresis loops were measured under various compressive or tensile strains by inward or outward bending of the films. The $M_s/M_r$ ratio is increased with the application of compressive strains and is decreased with the tensile strains applied along the easy or hard axis of Fe$_{81}$Ga$_{19}$ films. Consequently, the easy axis can be tuned to the hard axis or vice versa. Based on a modified Stoner-Wohlfarth model, in which the distribution of easy axes in polycrystalline films was considered, the mechanically tunable magnetic properties in flexible Fe$_{81}$Ga$_{19}$ films can be well understood.

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