Magnetic Behavior of FePt/AlN Layered Structure

Cong Zhang¹, Ryūtarō Tajima¹, Takumi Sannomiya, Shinji Muraiishi, Yoshio Nakamura and Ji Shi²

Department of Metallurgy and Ceramics Science, Tokyo Institute of Technology, Tokyo 152-8552, Japan

FePt(½)/AlN(20 nm) layered structures (x = 1.5–9 nm) were fabricated on fused quartz substrate by magnetron sputtering method. The magnetic behaviors of as-deposited and annealed films have both been studied. It has been found that annealing of the films leads to a transition of magnetic anisotropy from in-plane to perpendicular direction for layered structures with thinner FePt layer thickness. The interface evaluation performed by X-ray reflectivity (XRR) measurement and transmission electron microscopy (TEM) observation indicates that perpendicular magnetic anisotropy of the annealed layered structure can be attributed to the improved interface anisotropy, which is due to the flattening of interfaces. However, for films with thicker FePt layer thickness (above 5 nm), the in-plane anisotropy was enhanced after annealing. The results of stress analysis reveal that the residual stress change inside FePt layers upon annealing can override the interface contribution through magneto-elastic effect in such layer thickness range. [doi:10.2320/matertrans.MBW201304]

(Received October 16, 2013; Accepted December 6, 2013; Published January 18, 2014)

Keywords: layered structure, perpendicular magnetic anisotropy, thermal annealing, residual stress measurement, interface quality evaluation

1. Introduction

Thin films with perpendicular magnetic anisotropy (PMA) have been extensively studied in the past decades, because their suitability in vertical recording and magnetic field sensors has drawn much research interest.¹,² Layered structures (LS) or multilayers, consisting of a periodic stacking of metal layer A on metal layer B, have been a hot issue for obtaining PMA owing to the capacity of multiple sources of magnetic anisotropy. García et al. firstly reported PMA for layered structure in Co/Pd system,³ and attributed the PMA to the interface anisotropy which is originated from the lowered symmetry at the interface. Chappert et al. suggested a magnetoelastic anisotropy in Co/Au (Cu) layered structures,⁴ ascribing it to the stresses stored inside Co layer due to the lattice mismatches at interfaces. Later, PMA has also been found in other metal/metal LS, such as Co/Pt.⁵ Co/Ru.⁶

For M/M films, interfacial diffusion often occurs and results to a decrease of perpendicular anisotropy.⁷ In our previous work, we tried to use nitride layers instead of the noble metal layers to avoid such diffusion and firstly reported PMA in magnetic alloy/nitride-CoPt/AlN layered structures.⁸ The films undergo a gradually transition from in-plane anisotropy to perpendicular anisotropy as the annealing temperature increasing. Good consistence between anisotropy transition and in-plane stress change inside CoPt layers strongly implies the contribution from magneto-elastic anisotropy.

In this paper, we report the magnetic behavior in another magnetic alloy/nitride layered structure-FePt/AlN LS and show the different annealing effect on magnetic anisotropy in various FePt layer thicknesses. The appearance and absence of perpendicular magnetic anisotropy is related to the changes of interface quality and residual stress inside FePt layer.

2. Experimental Procedure

The FePt/AlN layered structures are prepared by a DC magnetron sputtering method onto fused-quartz substrates. The sputtering apparatus is equipped with two pairs of facing targets. One pair is consisting of two FePt composite type targets, and the other is composed of two Al targets. The sputtering chamber pressure was reduced to below 5 × 10⁻⁶ Pa before deposition and the sputtering process was carried out at room temperature in a gas mixture of argon and nitrogen with a total pressure of 0.2 Pa. The layer formed at the Fe–Pt target side is FePt solid solution with FCC structure, and the layer formed at Al target side is aluminium nitride (AlN) with wurtzite structure. The atomic ratio of Fe to Pt is fixed at 4:6, and the layered structures were deposited as sub/AlN20 nm/[FePt(x) nm/AlN20 nm]₃ (x = 1.5, 2, 5, 7, 9 nm) in the present work. After deposition, the films were annealed at 500°C in a vacuum condition with the pressure below 5 × 10⁻⁴ Pa for 3h.

The structure of layered structures was characterized by X-ray diffraction (XRD) using XRD 2θ–ω scan method (Bruker D8 Discover, Cu Kα radiation, at 30 kV, 300 mA). The interface quality was evaluated by X-ray reflectivity (XRR) measurements (Bruker D8 Advance, Cu Kα radiation, at 30 kV, 300 mA) and transmission electron microscopy (TEM) observation (JEM-3010, at 300 kV). Magnetic properties were measured by vibrating sample magnetometer (VSM) (RIKIN BVH-50V) at room temperature.

3. Results and Discussions

Hysteresis loops for as-deposited and annealed FePt/AlN layered structures with FePt layer thicknesses 1.5, 2, 5, 7, 9 nm is shown in Fig. 1. It is clear that all the as-deposited films display the easy-magnetization direction lies in the film plane. Such in-plane anisotropy increases obviously with increasing FePt thickness as shown in Figs. 1(a)–1(e), which implies the important role of interface/volume ratio in determining the anisotropy of layered structures. However,
for annealed films, the anisotropy can be altered in different ways. As shown in Figs. 1(f) and 1(g), the films with FePt layer thicknesses 1.5 and 2 nm, after annealing, are found to have an easy axis of magnetization perpendicular to the film plane and a high ratio of remanent magnetization to saturation magnetization for perpendicular direction, $M_r/M_s \approx 1$. For FePt thicknesses of 5 nm (Fig. 1(h)), the as-deposited and annealed films show almost the same behavior, and when continue increasing to 7 and 9 nm (Figs. 1(i) and 1(j)), the thermal annealing turns to favour the in-plane anisotropy. These results indicate that thermal annealing results in different changes in magnetic anisotropy for the films with different FePt layer thickness.

As mentioned above, magnetoelastic effect is an important source of PMA for layered structures. The stress stored in magnetic layers can significantly modify the anisotropy of layered structures via inverse magnetostriction effect. In order to understand the origin of PMA in annealed films, the residual stress conditions of FePt layers were studied. Under an assumption of equi-biaxial plane stress state, the strain at an angle $\psi$ with respect to the surface normal can be expressed as $\varepsilon_\psi = (a_\psi - a_0)/a_0 = \sigma_1((2S_{11} + 4S_{12} - S_{44})/3 + (S_{44}/2)\sin^2\psi)$ for (111) textured cubic polycrystalline films, where $\sigma_1$ is the stress in any direction in the film plane, $a_\psi$ is the lattice parameter along the direction of an angle $\psi$ with respect to the surface normal, $a_0$ is the unstrained lattice parameter, and $S_{ij}$ is a component of the elastic compliance array. For films with FCC structure and highly (111) textured, the in-plane stresses can be derived by measuring the interplanar distance of (111) planes at $\psi = 0$ and 70.53°.

Figure 2 shows the $2\theta$–$\omega$ XRD profiles of layered structures with different tilting angle of scattering vector, $\omega = 0$ and 70.53°, with respect to the film normal. The

![Fig. 2](image)

**Fig. 2** $2\theta$–$\omega$ XRD profiles of layered structures with different tilting angle of scattering vector: (a) as-deposited, $\omega = 0°$, (b) as-deposited, $\omega = 70.53°$, (c) annealed, $\omega = 0°$, (d) annealed, $\omega = 70.53°$. 
out-of-plane XRD profiles as shown in Figs. 2(a) and 2(c) confirm the preferred orientations of FePt [111] and AlN [001] parallel to the growth direction for both as-deposited and annealed films. After annealing, the peak position of FePt 111 shifts to higher angles, which implies the residual stress inside the FePt layers changes after annealing. In addition, the FePt (111) peak position also shows some dependence on the FePt layer thickness. The XRD profiles with θ = 70.53° are shown in Figs. 2(b) and 2(d). The FePt (111) peaks are clearly seen. All the peaks were fitted and the interplanar distances were specified by software HIGHEXPERT. Then the in-plane stresses can be derived with

\[ S_{11} = 8.24 \times 10^{-3} \text{GPa}^{-1}, \quad S_{12} = -3.41 \times 10^{-3} \text{GPa}^{-1} \quad \text{and} \quad S_{44} = 10.5 \times 10^{-3} \text{GPa}^{-1}, \]

which are calculated from the stiffness tensor of bulk FCC FePt crystals.\(^{11})\)

The in-plane stresses are plotted against FePt layer thickness and shown in Fig. 3. Large compressive stresses, derived with software HIGHEXPERT. Then the in-plane stresses can be calculated from the stiffness tensor of bulk FCC FePt crystals.\(^{11})\)

The XRD profile of AlN 20 nm, which shows smaller compressive stresses comparing to the as-deposited ones below 7 nm, which can be explained by the stress relief due to annealing. Another important feature is that tensile stresses are developed in annealed films at FePt thickness of 7 and 9 nm. We ascribe the appearance of the tensile stresses to the large difference in thermal expansion coefficient between FePt and AlN. It is known that FePt has a thermal expansion coefficient in bulk value more than twice as large as AlN.\(^{12})\)

The magnetoelastic effect or magnetoelastic anisotropy can be described as

\[ k_{\text{me}} = -\frac{3}{2} \lambda \sigma, \]

where \( \lambda \) is the magnetostriction constant along the magnetization direction, \( \sigma \) is the in-plane stress.\(^{13})\)

Since the FePt alloy has a positive magnetostriction constant,\(^{14,15})\) in-plane compressive stresses \((\sigma < 0)\) can result in a positive magnetoelastic anisotropy which favours the magnetization in perpendicular direction according to the equation above. However, the PMA of the annealed layered structures cannot be explained solely by the magnetoelastic effect, because the as-deposited films undergo larger compressive stress, nevertheless, they show in-plane magnetic anisotropy.

On the other hand, interface anisotropy, as theoretically predicted by Néel for thin film materials,\(^{16})\) can be enhanced by improving the interface quality.\(^{17,18})\) To confirm the annealing effect on the interface quality in this work, XRR measurement and TEM observation were performed. Figure 4 shows the XRR profiles of the layered structures for FePt thickness 2 nm before and after annealing. A number of superlattice peaks can be clearly identified for both, indicating good periodicity of the layered structures. What is important is that, the decay rate of amplitude of the oscillation at higher angles decreases markedly for annealed one. This strongly suggests that the interfacial diffusion does not occur in the layered structures, and the sharpness and flatness of interfaces are improved by annealing. The inset TEM image taken from the annealed film reveals flat and distinct interfaces between FePt and AlN layers, which confirm such improvement. As we know the interface anisotropy arises from the asymmetric environment of atoms at the interface. With a rough interface, some in-plane neighbors are lacking, leading to a reduction of this asymmetry. So the interface anisotropy is very sensitive to interface quality, i.e., precise distribution of magnetic and nonmagnetic atoms at the interface. So by summarizing the magnetic behavior, stress analysis and interface evaluation, we conclude that the interface anisotropy which is enhanced by interface sharpening, should be responsible for the PMA in annealed FePt 1.5 and 2 nm layered structures.

![Fig. 3](image-url) In-plane stresses in FePt layers as a function of FePt layer thickness.

![Fig. 4](image-url) XRR profiles of AlN20 nm/(FePt2 nm/AlN20 nm)\(_3\) layered structures, (a) as-deposited, (b) annealed. The inset TEM image is taken from an annealed film.
The effective magnetic anisotropy energy $K_{eff}$ (J/m$^2$) was calculated from the area between the in-plane and perpendicular magnetization curves. $K_{eff}$ could be phenomenologically separated in a volume contribution $K_V$ (J/m$^3$) and an extra interface contribution $K_S$ (J/m$^3$) and approximately expressed as $K_{eff} = K_V + 2K_S/t$, where $t$ is the thickness of a magnetic layer. Figure 5 shows the value of $K_{eff}$ with FePt (mJ/m$^2$) plotted against $t_{FePt}$. It is found that all the $K_{eff}$ of as-deposited layered structures are negative and thermal annealing obviously shows opposite effects on anisotropy energy in different FePt thickness range---$K_{eff}$ is increased below 5 nm and decreased above 5 nm. It is also should be noted that the $K_{eff}$ with annealed layered structures gradually deviate from linear behavior as $t_{FePt}$ increases. By estimating the $K_{eff}$ at $t=0$, we determine that $K_S$ is about 0.2 and 0.38 mJ/m$^2$, respectively, for the as-deposited and annealed layered structures. In general, shape anisotropy (or demagnetization energy), magneto-crystalline anisotropy, magnetoelastic anisotropy $K_{me,V}$ compose the volume contribution $K_V$, and interface contribution $K_{eff}$ incorporates Néel-type interface anisotropy, magnetoelastic interface anisotropy $K_{me,S}$ originated from coherent stresses. However, since the lattice misfit between FePt and AlN layer is about 13% at interface, it is reasonable to consider that there is no coherent-incoherent strain transition occurred in FePt layers. The interface is supposed to be incoherent, consisting large quantity of dislocations. Under such assumption $K_{me,S}$ can be neglected in this work, and $K_S$ is almost solely Néel-type. So for layered structure of $t_{FePt}$ below 5 nm, although the relief of compressive stress caused by annealing (shown in Fig. 3) will decrease the perpendicular (positive) magnetoelastic anisotropy as discussed above, the enhanced interface anisotropy becomes dominant then $K_{eff}$ of annealed films increases. When $t_{FePt}$ becoming thick, the contribution of $K_V$ is relatively small, and tensile stresses developed during annealing will result in a negative magnetoelastic anisotropy which overwhelms the interface anisotropy, thus decreases $K_{eff}$ as shown in Fig. 5. The nonlinear relationship between $K_{eff}$ with $t_{FePt}$ and $t_{FePt}$, appeared in annealed layered structures at larger thickness range, can also be explained by the continuous stress change. The stress change from compressive to tensile as thickness increases could modify $K_V$ through magnetoelastic anisotropy.

4. Conclusion

The different magnetic behaviors of as-deposited and annealed FePt/AlN layered structures have been investigated. The as-deposited layered structures show in-plane anisotropy, while the thermal annealing causes the change of anisotropy in different ways as FePt layer thickness increases. PMA has been found in annealed films with FePt thickness of 1.5 and 2 nm, and we attribute it to the enhanced interface anisotropy caused by interface flattening during annealing. The stress analysis shows stresses in layered structures of FePt 7 and 9 nm undergo a transition from compressive to tensile, which may favour in-plane anisotropy via magnetoelastic effect.

REFERENCES