Hindawi Publishing Corporation
Journal of Nanomaterials
Volume 2012, Article ID 814162, 7 pages
doi:10.1155/2012/814162

Research Article

Temperature-Driven Spin Reorientation Transition in CoPt/AlN Multilayer Films

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Received 10 May 2012; Accepted 23 May 2012

Academic Editor: Weichang Hao

Spin reorientation transition phenomena from out-of-plane to in-plane direction with increasing temperature are observed for the 500°C annealed CoPt/AlN multilayer films with different CoPt layer thicknesses. CoPt-AlN interface and volume anisotropy contributions, favoring out-of-plane and in-plane magnetization, respectively, are separately determined at various temperatures. Interface anisotropy exhibits much stronger temperature dependence than volume contribution, hence the temperature-driven spin reorientation transition occurs. Interface anisotropy in this work consists of Néel interface anisotropy and magnetoelastic effect. Magnetoelastic effect degrades rapidly and changes its sign from positive to negative above 200°C, because of the involvement of stress state in CoPt films with temperature. By contrast, Néel interface anisotropy decays slowly, estimated from a Néel mean field model. Thus, the strong temperature dependence of CoPt-AlN interface anisotropy is dominated by the change of magnetoelastic effect.

1. Introduction

Spin reorientation transition (SRT) is one of the most interesting phenomena occurring in thin ferromagnetic films. During this process, the orientation of the magnetization changes spontaneously and often reversibly from one direction to another. This process can be induced by varying related parameters such as measuring temperature and film thickness. Tremendous efforts have been made to investigate SRT in various systems such as Fe/Ag(100) [1–4], Fe/Cu(100) [1, 3, 5–7], Gd/W(110) [8–10], and Co/Au(111) [11–14]. Generally, with increasing temperature and film thickness, SRT from perpendicular to the in-plane magnetic direction has been observed. However, Ni/Cu(001) thin films exhibit the reversed SRT, namely, from in-plane to perpendicular orientation [15–17]. That is due to the different signs of the surface anisotropy between Ni and Fe, Co, Gd, which favors in-plane magnetization for Ni and out-of-plane magnetization for Fe, Co, Gd.

SRT phenomena are generally caused by the competition between several anisotropy contributions favoring different directions of magnetization. For example, different temperature dependences of these anisotropy contributions may make a temperature-driven SRT occur. For a thin ferromagnetic film, the surface or interface anisotropy (K_S) becomes comparable with the other effects like shape and magnetocrystalline anisotropies. Hence, it is important to study the temperature dependence of K_S to understand the SRT process. Farle et al. found an almost linear decrease of K_S with increasing temperature for Gd/W(110), Ni/W(110), and Ni/Cu(001) thin films [10, 18–20]. Similar linear temperature dependence of K_S has been observed for Ni/Re(0001) [21] and Fe/Cu(001) [7]. On the other hand, Pechan found a much more abrupt decrease of K_S for Ni/Mo multilayers, which is inexplicable in terms of a simple Néel mean field model [22]. The author then supposed such strong temperature dependence of K_S was due to the involvement of Ni-Mo interface strain, which generates a magnetoelastic term in K_S. But quantitative study of the magnetoelastic effect has not been conducted.

For the metal/metal films, the interlayer mixing or interfacial reaction at elevated temperatures will destroy the interface and make an irreversible change to the magnetic properties. In our previous work, to avoid such structural changes, AlN instead of metal was used to form CoPt/AlN multilayer films [23–25]. Flat and continuous CoPt-AlN interface has been observed even after high-temperature postdeposition annealing at temperatures as high as 500°C [23, 24]. CoPt layers remain FCC structure when the annealing temperature is below 600°C [25]. High crystallinity, strong (111) texture in CoPt layers and (002) texture in AlN layers were exhibited. Moreover, a large perpendicular magnetic anisotropy was obtained mainly due to the tensile stress in the CoPt layers [23]. In the present work, we have investigated the temperature dependence of CoPt-AlN interface anisotropy up to 350°C and its relationship with the temperature-driven SRT in CoPt/AlN multilayer films. The temperature dependence of the stress-induced magnetoelastic term in interface anisotropy has also been quantitatively studied.

2. Experiment

CoPt/AlN multilayer films with the configuration as [AlN (10 nm)/CoPt (x nm)]$_5$/AlN (10 nm) ($x = 2, 4, 6, 8$) were prepared at ambient temperature by direct current (dc) magnetron sputtering on fused-quartz substrates. Two pairs of facing targets, one pair composed of Co and Pt targets and the other pair composed of two Al targets, were equipped at two sides in the sputtering chamber. Sample depositions were conducted in an Ar and N_2 gas mixture. CoPt/AlN multilayer films were formed by switching the substrate holder alternatively to the two targets sides. The atomic ratio in the CoPt layer is $Co_{44}Pt_{56}$. As-deposited films were then annealed in a vacuum furnace (below 1×10^{-4} Pa) at 500°C for 3 hours.

Out-of-plane (along the film normal, $\perp$) and in-plane (in the film plane, $\|$) magnetization-temperature (M-T) curves were measured by a vibrating sample magnetometer (VSM) with heating rate as 5°C/min. Before the measurement of M-T curves, the sample was magnetized in an external field of 1 T, and then the external field was removed. However, a small constant external field about 60 Oe, which is the residual magnetization of the cores inside the electromagnets, still exists. In-plane and out-of-plane hysteresis loops at various temperatures up to 350°C were measured with the same VSM.

3. Results and Discussion

Figure 1 shows the evolutions of in-plane and out-of-plane remnant magnetizations with temperature for CoPt/AlN multilayer films with different CoPt layer thicknesses. SRT from out-of-plane to in-plane direction, indicated by an abrupt decrease of out-of-plane magnetization ($M_\perp$) followed by rapid increase of in-plane magnetization ($M_\|$), can be observed for the films with CoPt layer thickness larger than 2 nm. Moreover, SRT occurs at lower temperatures when the CoPt layer thickness increases. For the film with CoPt layer thickness $d = 2$ nm, $M_\perp$ starts decreasing rapidly at a relative high temperature of 303°C, while $M_\|$ remains a low value (about one tenth of $M_\perp$) in the whole temperature range without obvious increase. When $d = 4$ nm, $M_\perp$ exhibits an abrupt degradation from a much lower temperature of 110°C. On the other hand, $M_\|$ starts a sharp increase around 250°C and reaches a considerable magnitude. For the thicker films with d as 6 and 8 nm, it seems that the rapid depression of $M_\perp$ occurs from a temperature below room temperature. The starting temperature of the rapid increase of $M_\|$ is about 100°C when $d = 6$ nm, while it should be lower than the room temperature when $d = 8$ nm. The final decay of $M_\|$ in the in-plane M-T curve is due to the ferromagnetic-paramagnetic transition.

As mentioned in Section 1, the temperature-driven SRT is generally caused by the change of magnetic anisotropy with temperature. Hence, we measured the in-plane and out-of-plane hysteresis loops for each film at various temperatures up to 350°C in order to measure the effective anisotropy energy. Typical results for the CoPt/AlN multilayer film with $d = 4$ nm are shown in Figures 2(a)–2(c), which show a smooth transition from perpendicular to in-plane magnetic anisotropy. At 250°C, the film almost exhibits a magnetic isotropic behavior (Figure 2(b)). Figure 2(d) shows the change of saturation magnetization (M_S), obtained from the hysteresis loops, with temperature. The degradation of M_S exhibits a growing rate with increasing temperature.

The total magnetic anisotropy energy per volume K_{eff} (erg/cm^3) is calculated by measuring the area enclosed between in-plane and out-of-plane hysteresis loops. When the magnetic anisotropy is extremely high, the saturation of magnetization along the hard axis cannot be reached under the maximum applied field of 1 T. In such cases, the hard axis hysteresis loop is extrapolated to M_S. Figure 3 summarizes the calculated K_{eff} of CoPt/AlN multilayer films with $d = 2$, 4, 6 and 8 nm at different temperatures. At room temperature, a high perpendicular magnetic anisotropy (PMA) energy of 7.2×10^6 erg/cm^3 is obtained when $d = 2$ nm. With increasing d, PMA weakens and turns to a weak in-plane magnetic anisotropy (IMA) when $d = 8$ nm. With increasing temperature, PMA rapidly degrades. Due to the strong PMA of 2 nm thick CoPt thin film, out-of-plane easy axis is maintained up to 350°C. On the other hand, when $d = 4$ and 6 nm, the transition from PMA to IMA occurred at 250°C and 100°C, respectively. When $d = 8$ nm, only IMA is observed. Maximum K_{eff} with a value of -8.2×10^5 erg/cm^3 has been obtained at 300°C. The degradation of IMA at high temperatures may arise from the enhancing thermal fluctuations. Magnetic anisotropy could be expected to vanish at the Curie temperature.

Magnetic anisotropy decides the magnetization state at each temperature, illustrated in Figure 1(b). When K_{eff} is extremely high, for example, $d = 2$ nm below 303°C and $d = 4$ nm below 110°C, a single perpendicular domain can be maintained. Hence, $M_\perp$ only gradually decreases with increasing temperature. When K_{eff} decreases to a critical value, for example, $d = 2$ nm at 303°C and $d = 4$ nm at 110°C, the reversed magnetization starts to form.

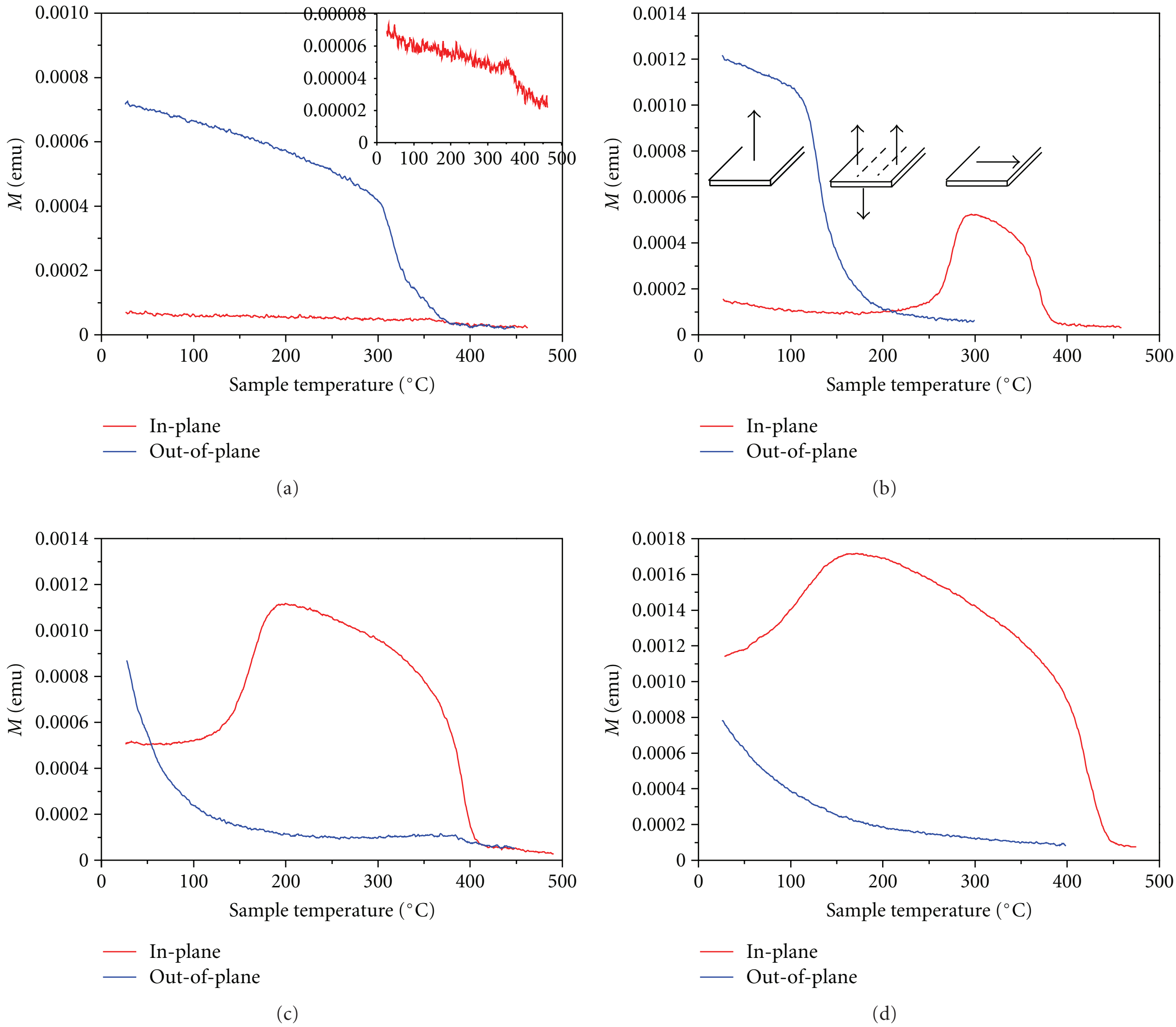

Figure 1: In-plane and out-of-plane Magnetization-Temperature (M-T) curves of CoPt/AlN multilayer films with the CoPt layer thickness of 2, 4, 6 and 8 nm for (a), (b), (c), and (d), respectively. The inset image in (a) is the in-plane M-T curve in a small magnetization range. In (b) the corresponding magnetization states are illustrated.

Consequently, the single domain state is destroyed and $M_{\perp}$ rapidly decreases. When thin films become magnetic isotropy ($K_{\text{eff}} = 0$), for example, $d = 4$ nm at 250°C and $d = 6$ nm at 100°C, perpendicular domains become unstable and in-plane domains starts to form. Thus, $M_{\parallel}$ starts a sharp increase. The experimental observation of the evolution of magnetic domain with temperature can be seen in [6]. But a detailed experimental study on the correlation between the change of magnetic anisotropy and the change of remanent magnetization was rarely reported previously. Our work reveals that at the starting temperature of the rapid decrease of $M_{\perp}$, thin films still possess considerable PMA; at the starting temperature of the rapid increase of $M_{\parallel}$, thin films exhibit magnetic isotropy.

For a ferromagnetic thin film, K_{eff} (erg/cm^3) can be phenomenologically written as $K_{\text{eff}} = (2K_S/d)+K_V$, where K_S (erg/cm^2) is the interface anisotropy energy density, d is the film thickness, and K_V (erg/cm^3) is the volume contribution, which is the sum of the magnetocrystalline, magnetoelastic, and shape anisotropies. The factor of 2 in $2K_S/d$ takes into account the two interfaces. $K_{\text{eff}} > 0$ corresponds to perpendicular anisotropy and $K_{\text{eff}} < 0$ in-plane anisotropy. To understand the observed SRT phenomena, temperature dependences of K_S and K_V need to be revealed. The individual values of K_S and K_V can be determined by the linear relationship between $K_{\text{eff}}d$ and d as $K_{\text{eff}}d = 2K_S + K_V d$, where $2K_S$ is the intercept at $d = 0$ and K_V is the slope. Good linear d dependence of $K_{\text{eff}}d$ is experimentally obtained at each measuring temperature, shown in Figure 4. Hence, the interface and volume anisotropy contributions are separately determined. Note K_S is positive and K_V is negative, indicating the interface and volume anisotropies favor out-of-plane and in-plane magnetization, respectively. The temperature dependences of K_S and K_V are then shown in Figure 5. It can be seen clearly that K_S decreases more rapidly than K_V. Consequently, K_V will become predominant and the

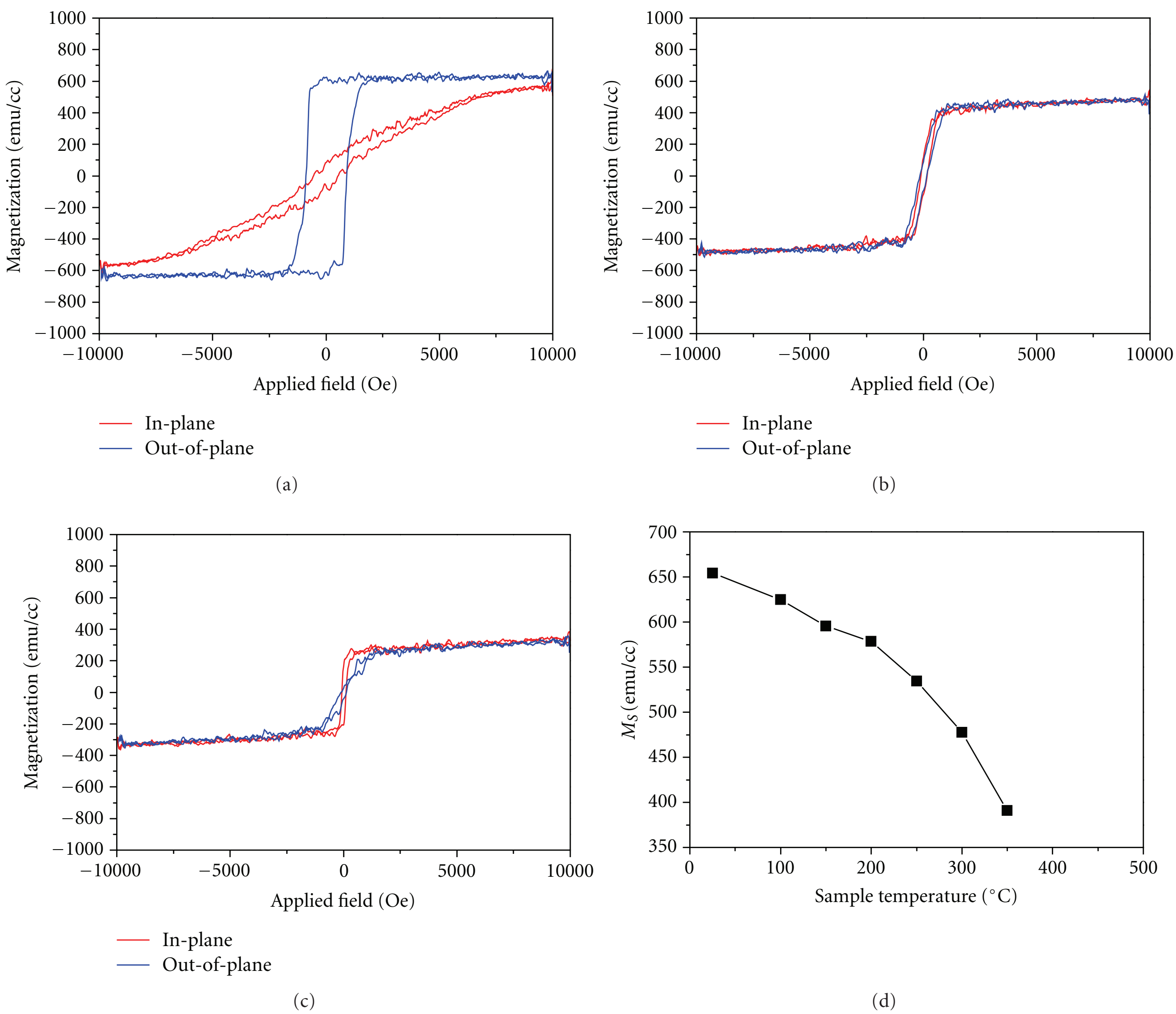

FIGURE 2: In-plane and out-of-plane hysteresis loops of a CoPt/AlN multilayer film with the CoPt layer thickness of 4 nm measured at room temperature (a), 250°C (b), and 350°C (c). The saturation magnetizations (M_S) at various temperatures are shown in (d).

magnetic easy axis will turn to the in-plane direction at high temperatures. Thus, an SRT from out-of-plane to in-plane orientation will be induced. When $d = 2$ nm, the absolute value of the effective volume contribution $-K_V d$ remains lower than the interface contribution $2K_S$, whereas their difference is rapidly reduced. That explains why there is no obvious increase in $M_{\parallel}$ with increasing temperature. When $d = 4$ and 6 nm, CoPt thin films become magnetic isotropy at 230°C and 100°C, respectively (see the intersections in Figure 5). Hence, $M_{\parallel}$ starts a sharp increase near these temperatures for the two films (Figures 1(b) and 1(c)). The volume effect overwhelms the interface effect at room temperature when d is increased to 8 nm. As a result, only IMA is observed in the whole temperature range and SRT should happen below room temperature, as shown in Figure 1(d).

The abrupt decrease of K_S with temperature is quite different from the linear dependence observed in some other systems, for example, Fe/Cu(100) and Gd/W(110) [7, 10], but similar to the results in Ni/Mo multilayers [22], denoting the importance of stress effect. K_S always contains the Néel interface anisotropy K_N, which originates from the broken symmetry at the interface [26]. Our previous study found the existence of a tensile stress in CoPt layers after postannealing and demonstrated that such tensile stress promotes a large PMA through magnetoelastic effect [23]. The magnetoelastic anisotropy energy K_{me} can be expressed as $K_{\text{me}} = -3\lambda\sigma/2$, where λ is the magnetostriction constant and σ is the internal stress [27]. When the parameters are independent of the film thickness, K_{me} can be identified with a volume contribution K_V. However, in some cases, σ is proportional to $1/d$ due to the accommodation of misfit dislocations [27]. Hence, K_{me} is proportional to $1/d$ and $K_{S,\text{me}}$, defined as $K_{S,\text{me}} = K_{\text{me}}d/2$, becomes a part of the interface contribution K_S. It has been revealed that for the 500°C annealed CoPt/AlN multilayer films, K_{me} contributes to K_S when $d \geq 2$ nm [28]. Based on the above discussion, K_S in the present work should consist of the Néel interface and magnetoelastic anisotropy terms $K_S = K_N + K_{S,\text{me}}$. At room temperature, K_S is 1.0 erg/cm^2, while K_N

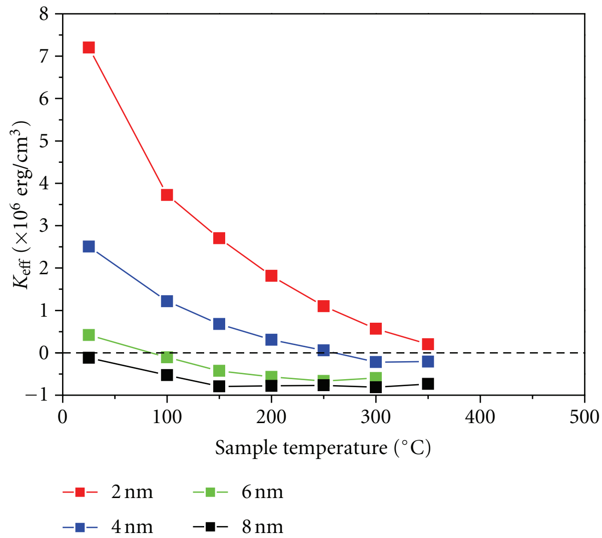

FIGURE 3: Magnetic anisotropy energy K_{eff} versus temperature for CoPt/AlN multilayer films with the CoPt layer thickness as 2, 4, 6, and 8 nm. $K_{eff} > 0$ and $K_{eff} < 0$ correspond to perpendicular and in-plane magnetic anisotropy, respectively.

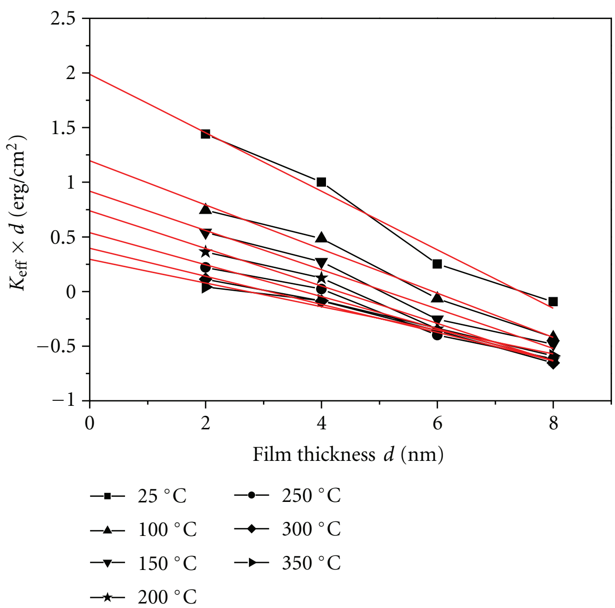

FIGURE 4: $K_{eff}d$ as a function of CoPt film thickness d at various measuring temperatures. Red lines are linear fit lines to the experiment data.

and $K_{S,me}$ have been determined to be 0.47 and 0.53 erg/cm^2 in our previous work, respectively [28]. According to a Néel mean filed model, K_N is proportional to M_S^2, $K_N = AM_S^2$ [22, 29]. The coefficient A is chosen to match the model with the experiment at room temperature. Using the M_S values at various temperatures (Figure 2(d)), the change of K_N with temperature is estimated (Figure 6). Subsequently the temperature dependence of $K_{S,me}$ is obtained from $K_{S,me} = K_S - K_N$ (Figure 6). K_N exhibits a much milder temperature

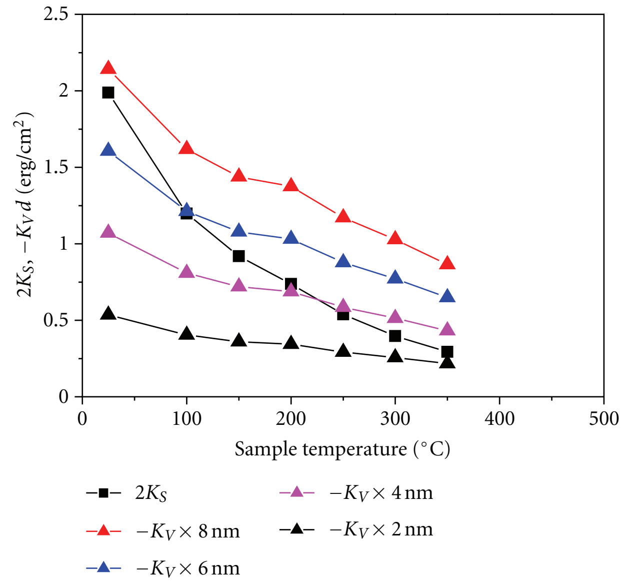

FIGURE 5: $2K_S$ and $-K_V d$ of CoPt/AlN multilayer films with d as 2, 4, 6, and 8 nm against temperature. $2K_S$ is the interface anisotropy energy density and $-K_V d$ is the absolute value of the volume anisotropy energy density.

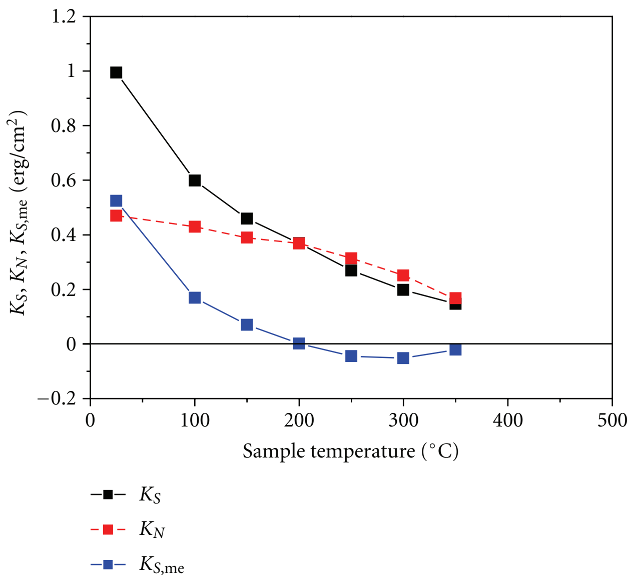

FIGURE 6: K_S, K_N and $K_{S,me}$ versus temperature. The dash line of K_N denotes it is estimated from a Néel mean field model. $K > 0$ and $K < 0$ correspond to perpendicular and in-plane magnetic anisotropy, respectively.

dependence than $K_{S,me}$, and thus the rapid decrease of K_S with increasing temperature is dominated by the rapid degradation of $K_{S,me}$.

The change of $K_{S,me}$ should be correlated to the change of stress σ, because $K_{S,me}$ arises from the magnetoelastic effect. At room temperature there exists a large tensile stress in the CoPt film and a compressive strain in the direction of film normal [23]. When the temperature is increasing, the lattice expands along film normal freely [30]. By contrast, the lattice expansion in the film plane would be restrained to a great extent by adjacent AlN layers through interfacial restriction because of the smaller thermal expansion coefficient and higher elastic modulus of AlN than CoPt [23]. Thus with increasing temperature, the compressive strain along film normal will degrade and further changes to a tensile strain. Therefore, as shown in Figure 6, $K_{S,\mathrm{me}}$ rapidly decreases with temperature and reaches nearly zero at 200°C. After that, $K_{S,\mathrm{me}}$ becomes negative and favors in-plane magnetization.

4. Conclusions

Temperature dependence of CoPt-AlN interface anisotropy in 500°C annealed CoPt/AlN multilayer films has been investigated. The interface anisotropy in the present work consists of Néel interface anisotropy and the magnetoelastic contribution. Néel interface anisotropy shows a mild temperature dependence, estimated from a Néel mean field model. By contrast, with increasing temperature, the magnetoelastic contribution decreases abruptly and changes its sign above 200°C. Such temperature dependence is owing to the involvement of the stress state in CoPt films. Accordingly, the total interface anisotropy exhibits a rapid degradation with temperature. Due to the milder temperature dependence of volume anisotropy than that of interface anisotropy, interesting temperature driven spin reorientation transition phenomena from out-of-plane to in-plane direction are clearly observed for CoPt/AlN multilayer films with appropriate CoPt layer thickness.

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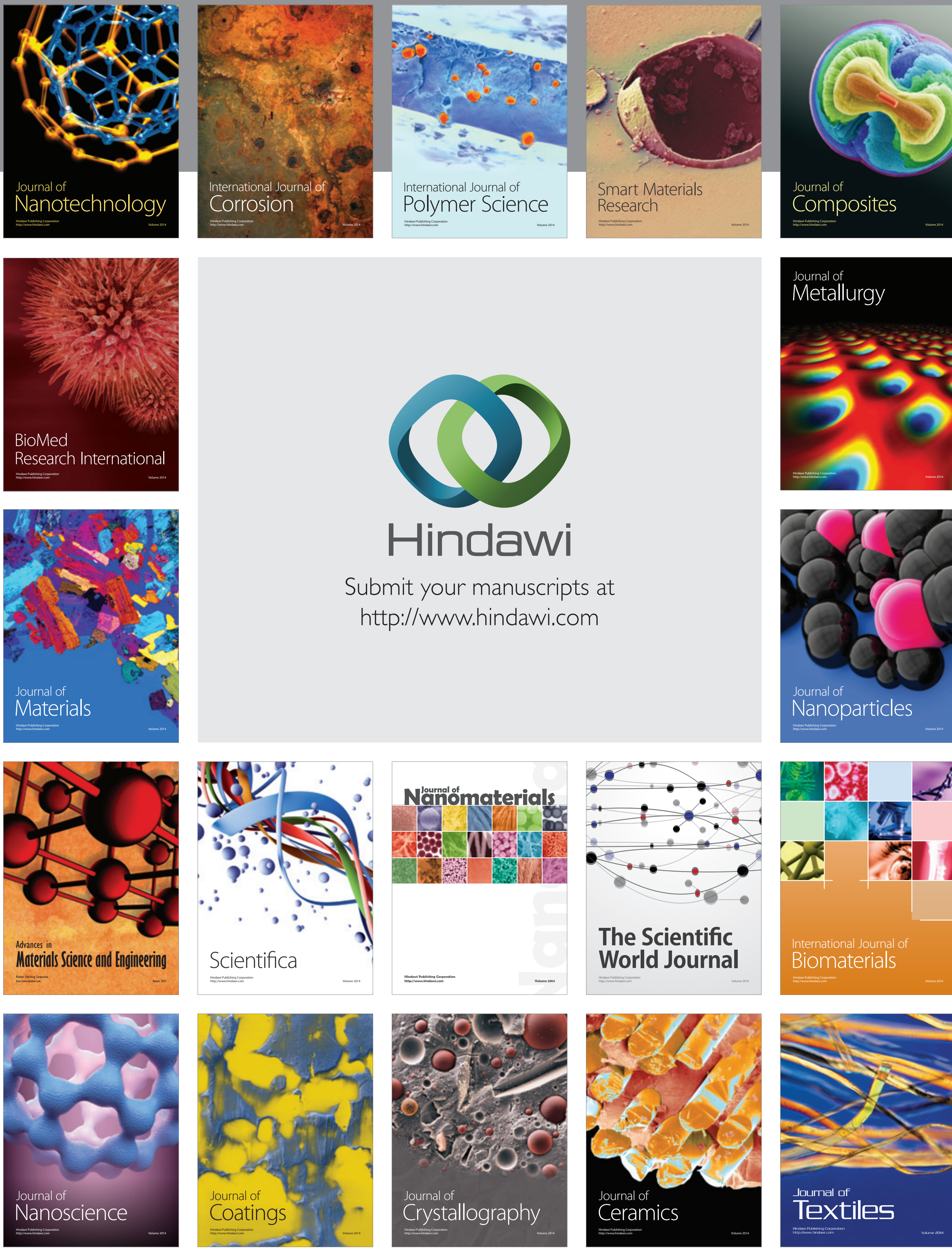
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