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Continuity of perpendicular FePt film with a compositional gradient design

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Abstract. By magnetron sputtering, [Fe(0.6 nm)/Fe_{30.5}Pt_{69.5}(1.9 nm)]₁₀ continuous multilayer films with the nominal composition of Fe₅₀Pt₅₀ and total thickness of 25 nm were deposited on MgO(001) substrates heated to 400°C, and then subjected to a vacuum annealing at temperatures in the range of $T_a = [500, 900]^\circ\text{C}$. The magnetocrystalline anisotropic energy exceeded 2.5×10^7 erg/cc at $T_a \geq 700^\circ\text{C}$, and the morphology kept continuous at $T_a \leq 800^\circ\text{C}$. Though the interlayer diffusion during sputtering was fairly sufficient, a weak periodical compositional gradient was left in the films, which is critical for the morphological continuity of film subjected to subsequently annealing. The slight periodical compositional gradient might suppress the segregation of Pt and sufficiently prevent from the Pt enrichment that would result in the discontinuous boundaries. A modified diffusion-recombination model of carrier was adopted to discuss the morphology continuity of annealed FePt films. The continuous film would fit the need for microfabrication of patterned arrays of $L1_0$ FePt.

1. Introduction

The (001) textured $L1_0$ FePt films with a large magnetocrystalline anisotropy (K_u) above 10^7 erg/cc have drawn great attentions due to the potential applications in data storage [1-3]. FePt films synthesized at room temperature normally adopt its disordered $A1$ phase. Post annealing or synthesis at an elevated temperature is necessary to transform to the desired ordered $L1_0$ phase. It is efficient to induce the (001) texture by depositing the FePt films on heated MgO(001) substrates [4,5]. To suppress the dispersion of grain size, researching on the artificially patterned magnetic arrays microfabricated by the methods such as electron beam lithography and focused ion beam lithography has become popular [6,7]. However, thermal treatment always causes sintering of FePt films and leads to destruction of the morphological continuity, conflicting with the need of uniform and continuous film for microfabrication. In order to keep the morphological continuity of film, the circumstantial temperature is generally limited to about 400°C, at which the ordering is insufficient yet and the films have vanishing small K_u [8]. In this article, we report a so-called compositional gradient method to synthesize [Fe(0.6 nm)/Fe_{30.5}Pt_{69.5}(1.9 nm)]₁₀ continuous multilayer films on MgO(001) substrates, by which the stoichiometric FePt films can undergo an annealing at temperature as high as 800°C without destruction of morphological continuity.

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2. Experiment

By using a magnetron sputtering system with the base pressure better than 8×10^{-6} Pa, multilayer films of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ were sputtered on $\text{MgO}(001)$ substrates heated to 400°C . The nominal composition of film was $\text{Fe}_{50}\text{Pt}_{50}$, and the total thickness was 25 nm. The stander with 9 substrate holders was rotating during deposition to ensure the uniformity of composition and thickness. All the samples were prepared at the same time to further ensure the consistence of composition. The elemental purity of target was 99.99 % for Pt and 99.93% for Fe, respectively. The sputtering chamber was pre-baked at $\sim 100^\circ\text{C}$ for 1 h to remove the water and the gas. The working pressure of argon gas was ~ 2.8 Pa. The sputtering rate was ~ 0.1 nm/s for Fe and ~ 0.3 nm/s for Pt, respectively. As the later discussion on figure 2a, despite the interlayer diffusion was sufficient during sputtering, it is reasonable that a subtle wave of composition (periodical compositional gradient) was left in the films. After sputtering, the samples were encapsulated in the tube of a rapid thermal annealing equipment and annealed for 3 hours at temperatures of $T_a = 500, 600, 700, 750, 780, 800,$ and 900°C , respectively. The crystallographic characterization of the films was performed with x -ray diffraction (XRD) using a Cu-K_α radiation. The morphology was observed by a scanning electron microscopy (SEM). The magnetic properties were measured by using a vibrating sample magnetometer (VSM) with a maximum applied field of ± 30 kOe.

3. Results and discussions

Figure 1 shows the SEM images of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ multilayer films annealed at different temperature [as-deposited (a), $T_a = 700^\circ\text{C}$ (b) and 900°C (c)], and the SEM image of FePt alloy film co-deposited at 700°C (d) for comparison. The thickness of all films was 25 nm. The as-deposited film had a continuous morphology with a number of small holes (the dark areas in figure 1a). At $T_a = 700^\circ\text{C}$ (figure 1b), the small holes disappeared and the surface of film became smoother. Compared to figure 1d, the morphological continuity of this multilayer film was obviously improved. No obvious change of roughness or continuity was observed for the films annealed at T_a in the range of $[700, 800]^\circ\text{C}$. After annealed at 900°C (figure 1c), the continuity was seriously destroyed.

Figure 2 shows the XRD spectra of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ multilayer films annealed at different temperature [as-deposited (a), $T_a = 700^\circ\text{C}$ (b) and 900°C (c)]. The strong (002) peak from MgO substrate suggests that the x -ray penetrated the FePt film. For the as-deposited film, only the (200) peak attributed to disordered A1 FePt was seen. There is no evidence for the presence of Pt and Fe phases, indicating that the interlayer diffusion during deposition was fairly sufficient. FePt (200) peak had a wider full width at half maximum (FWHM), which should be resulted from the difference of lattice constant at different depth of film. The distortion of lattice implies the existence of compositional fluctuation. Therefore, it is reasonable that a subtle wave of composition (periodical compositional gradient along the perpendicular direction) was left in the film which was deposited layer by layer. Annealed at 700°C and 900°C (figure 2b and 2c), the onset of (001) and (003) reflections from superlattice appeared, indicating that the film became ordered $L1_0$ phase with significant (001) orientation. The position of FePt (200) fundamental peak also shifted to a bigger angle [labeled as (002)], too, as a result of shortened lattice constant of c . In figure 2c ($T_a = 900^\circ\text{C}$), two additional unlabeled peaks respectively split from MgO (002) and FePt (002) appeared. They are likely to be caused by the weaker Cu-K_β radiation. The FePt (002) peak split due to its narrower FWHM, as the periodic composition fluctuation had vanished. However, the split of MgO (002) peak might be the result of the exposure of substrate surface to the x -ray radiation directly. This indicates that the morphological continuity of FePt film was seriously destroyed due to the sintering. As a result, the discontinuous FePt film could not completely absorb the Cu-K_β diffraction by MgO substrate any more. The change of XRD pattern agrees with the morphology evolution as seen in figures 1a-1c.

Figure 3 shows the hysteresis loops of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ multilayer films. The loops of as-deposited FePt film with disordered A1 phase (figure 3a) showed typical soft behaviors with an in-plane easy axis controlled by the shape anisotropy; the in-plane and out-of-plane

coercivities were both small. At $T_a = 500^\circ\text{C}$ (figure 3b), the shape anisotropy became weaker and the coercivities increased regardless along in-plane and out-of-plane directions, indicating the occurrence of nucleation of $L1_0$ phase. The film was a mixture of A1 and $L1_0$ phases with dominant A1 phase. The in-plane and out-of-plane coercivities reached maxima at $T_a = 600^\circ\text{C}$ (loops were not given here). At $T_a = 700^\circ\text{C}$ (figure 3c), the volume of $L1_0$ phase became dominant, and the perpendicular coercivity began to decrease with an out-of-plane magnetic easy axis. In the range of $T_a = [700, 780]^\circ\text{C}$ (figures 3c and 3d), the slope of out-of-plane curve near the coercive force became more abrupt, companied by an increased curve squareness and an decreased perpendicular coercivity. The loops showed obvious hard magnetic behaviors of continuous $L1_0$ films with a significant (001) texture, referring the morphology change as showed in figure 1 and the XRD change as showed in figure 2. The slope of perpendicular curve relaxed contrarily as T_a was slightly elevated from 780°C to 800°C (figure 3e). The maximum of positive magnetization was somewhat different from that of negative

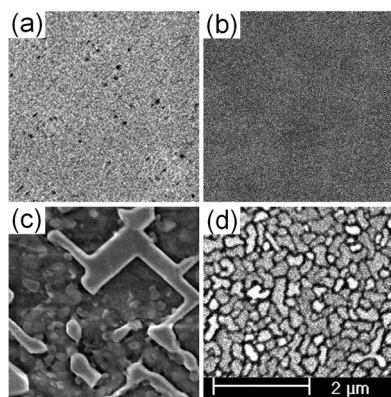


Figure 1. SEM images of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ film [(a) as-deposited, (b) $T_a = 700^\circ\text{C}$, (c) $T_a = 900^\circ\text{C}$], and FePt film co-deposited at 700°C (d).

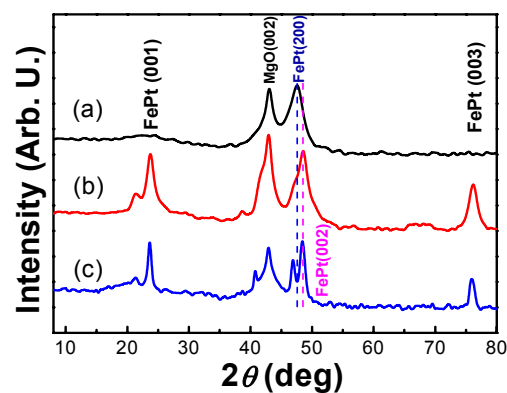


Figure 2. XRD spectra of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ film. (a) as-deposited, (b) $T_a = 700^\circ\text{C}$, (c) $T_a = 900^\circ\text{C}$.

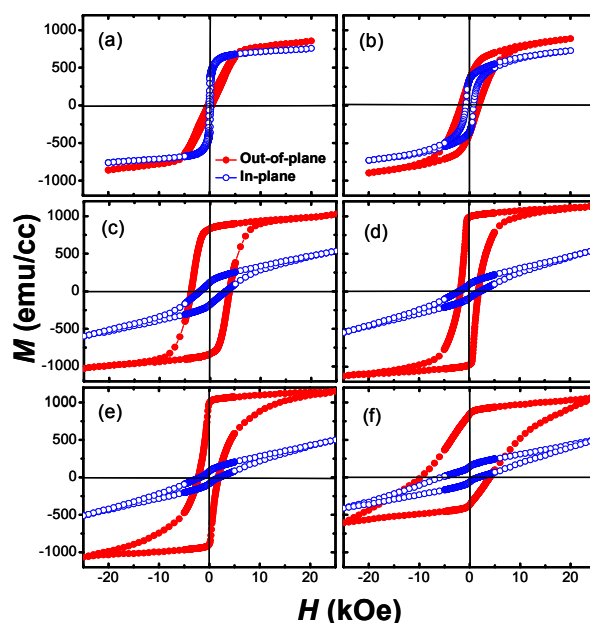


Figure 3. Hysteresis loops of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ film. (a) as-deposited, (b) $T_a = 500^\circ\text{C}$, (c) $T_a = 700^\circ\text{C}$, (d) $T_a = 780^\circ\text{C}$, (e) $T_a = 800^\circ\text{C}$, and (f) $T_a = 900^\circ\text{C}$.

magnetization, suggesting a subtle ruin of the morphological continuity. A small number of separated small $L1_0$ FePt particles with multi-domain magnetic configurations were magnetized to saturation along the direction of *ab initio* applied field, and the maximum of opposite field was not enough to overcome the barrier of large K_u to reverse the magnetization in them. The faint discontinuity should merely have taken place at the edges or locations of defect. At $T_a = 900^\circ\text{C}$, the out-of-plane loop was obviously asymmetric (figure 3f), due to the serious morphological discontinuity. The maximal magnetization of ~ 1070 emu/cc was less than that of ~ 1150 emu/cc for the film annealed at 800°C . This could be explained by the presence of fine single-domain particles with large switching field, which would contribute nothing to the magnetization as their magnetic moments aligned randomly upward and downward. Detailed analysis to this phenomenon can be found in references 9-12. Table 1 summarizes the T_a dependence of K_u determined from the enclosed area of the prolonged out-of-plane and in-plane curves.

Table 1. K_u of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ film annealed at different T_a .

T_a ($^\circ\text{C}$)	700	750	780	800
K_u ($\times 10^7$ erg/cc)	2.5	2.8	3.0	3.5

4. Analysis of the reason for improvement of morphological continuity

The kinetics for the polymorphic phase transition $A1$ to $L1_0$ is rather complex. Recently, the compositional profiles by atom probing indicated a preference of Pt to the grain boundaries in $L1_0$ FePt films, even though the films were Fe-rich [13], and modeling predictions also suggested the same preferential segregation events for changes in crystal symmetry [14-16]. These reports demonstrate that the phase transition provides an additional driving force for Pt migration to induce the segregation and subsequently the ruin of morphological continuity. The critical technique to improve the continuity of FePt film is how to control the segregation of Pt. The ordering in hot FePt film was suggested to initiate at the grain boundaries [17]. The subtle compositional fluctuation at boundaries assists in the atomic rearrangement and therefore is essential for the initiation of phase transition. In this paper, the film was designed as a multilayer structure of $[\text{Fe}(0.6 \text{ nm})/\text{Fe}_{30.5}\text{Pt}_{69.5}(1.9 \text{ nm})]_{10}$ with interlayer diffusion during temperature-elevated sputtering. As a subtle periodic compositional fluctuation was designed into the film along the perpendicular direction (assumed as z axis), the nucleation of $L1_0$ phase during annealing would mainly occur in the interior of film, and the periodical compositional gradients of Fe and Pt would suppress the segregation of Pt which can result in grain boundaries. Conveniently, we discuss the diffusion of Fe atoms. The Fe concentration can be roughly expressed as the atom density, n_{Fe} ,

$$n_{\text{Fe}} = n_0 + n'_0 \cos(\omega z), \quad (1)$$

where $\omega = 2\pi/2.5$, n_0 represents the averaged n_{Fe} , and n'_0 the fluctuation amplitude of n_{Fe} . In the subsequent ordering by annealing, Fe can suppress the segregation of Pt by preferentially migrating toward Pt-rich directions. Due to lack of grain boundaries, the film can maintain its morphological continuity at a higher temperature, until the depletion of periodical compositional gradient. By referring to the diffusion and recombination of carriers in semiconductor [18], considering the migration of Fe atoms in z direction as non-equilibrium diffusion with an activation energy Q , and the ordering jumping as recombination with an activation energy Q' ($Q' > Q$) and with an ordering probability $p = \exp(-Q'/k_B T)$, where k_B is the Boltzmann constant, and T is the temperature, the equilibrium in every half of the period of (1) will satisfy the following formula,

$$-\frac{d}{dz} \left(-\frac{1}{6} \frac{c^2}{\tau} e^{-\frac{Q}{k_B T}} \frac{dn}{dz} \right) - \frac{1}{\tau} n e^{-\frac{Q'}{k_B T}} = 0. \quad (2)$$

In (2), $n = n_{\text{Fe}} - n_0$, c is the diffusion length, and $1/\tau$ is the frequency of thermal vibration. The first item represents the accumulation of n , caused by the diffusion with a diffusion coefficient of $c^2 \exp(-Q/k_B T)/(6\tau)$. The second item represents the loss of n , caused by the ordering with a

modification of n replacement by np . The 'loss' means that the ordered atoms will no longer contribute to diffusion. z starts at every position of $n = n'_0$ and always toward the Pt-rich direction. The solution of (2) is

$$n = (n_{\text{Fe}} - n_0)_0 \exp \left[-\sqrt{6} \left(\frac{z}{c} \right) / e^{\frac{Q'-Q}{2k_B T}} \right]. \quad (3)$$

Therefore, the factor of $\exp[(Q'-Q)/2k_B T]$ decides the new diffusion length. (3) tells us that overlong annealing time is no use to urge the ordering as the initiation time is in the scale of seconds [17]. With the increase of T , the diffusion length becomes shorter, indicating that more volume of the film transits into $L1_0$ phase. $(n_{\text{Fe}} - n_0)_0$ should be modified with the increase of T . At $k_B T \gg (Q'-Q)/2$, $n \rightarrow 0$, as $(n_{\text{Fe}} - n_0)_0 \rightarrow 0$.

5. Conclusions

[Fe(0.6nm)/Fe_{30.5}Pt_{69.5}(1.9nm)]₁₀ continuous multilayer films were sputtered on MgO(001) substrates heated to 400°C. By interlayer diffusion during the deposition, a subtle periodical compositional gradient was left in the films along the perpendicular direction. By annealing at $T_a \geq 700^\circ\text{C}$, the films became ordered $L1_0$ phase with a significant (001) orientation and a magnetocrystalline anisotropic energy exceeding 2.5×10^7 erg/cc. The morphology of film kept continuous at $T_a < 800^\circ\text{C}$. The compositional gradient design is an effectual method to suppress the segregation of Pt in the phase transition and provide a convenience for microfabrication with continuous $L1_0$ FePt films.

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