Effects of non-magnetic phase and deposition temperature on magnetic properties of FePt-MgO granular thin films on single-crystal MgO substrate

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Abstract

(FePt)_{100-x}(MgO)_x granular thin films were fabricated by using co-sputtering on single crystal MgO substrate at different deposition temperatures followed by a high-temperature annealing. The adding MgO was found to have the following effects: 1) high fraction of MgO phase (70 vol.%) degraded the L1₀ phase transformation; 2) FePt grains embedded in MgO matrix were well separated, leading to minimized exchange interaction between the FePt grains; 3) 55 vol.% fraction of MgO was optimal for the L1₀-FePt phase formation with highest coercivity, due to the formation of granular structure, which was also verified by comparing the structure and properties of FePt/MgO bilayer films. Deposition temperature has also effect on the magnetic properties of FePt-MgO films. Room temperature deposition did not result in the L1₀ phase even after annealing at 800°C. However, the magnetic properties of the films deposited at 200°C were better than those deposited at 300°C. The low coercivity resulted from elevated deposition temperature was attributed to the thermal expansion and competitively growth among FePt and MgO grains. The results revealed that adding appropriate amount of MgO (55 vol.%) can be achieved by finding a proper deposition temperature (200°C), well-defined granular L1₀-FePt films with fine grain distribution, high coercivity and small domain size.

Key words:

L1₀-FePt granular film; MgO matrix; Deposition temperature; Exchange coupling effect

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Introduction

L1₀-FePt film has the high magnetocrystalline anisotropy energy (K_u) and high coercivity (H_C), so that it is an ideal high density magnetic recording material ^[1]. However, L1₀-FePt ordered phase is obtained by high temperature annealing or deposition, which is prone to increase the grain size of FePt. The large size grains cause the increase of surface roughness and exchange coupling effects. It is detrimental to the high density recording. Eventually, it is crucial to form the granular or isolated FePt films by adding the non-magnetic matrix, which can decouple the hard phase grains and improve the reliable recording signals. It was reported that some researchers attempted to obtain the granular microstructure by using various non-magnetic materials ^[2-4]. MgO is chosen for its widely application in the tunnel junction devices and has a suitable lattice mismatch with FePt (4%) ^[5-7]. In general, FePt-MgO nano-granular film can be formed by either FePt/MgO multilayer deposition or co-sputtering of FePt and MgO after the annealing ^[8-9]. Nevertheless, excessive amount of MgO non-magnetic phase will degrade the transformation of L1₀-FePt ^[10]. It may be caused by the change of exchange coupling length due to the isolation by MgO matrix. For that reason, it is necessary to control the appropriate concentrations of doping non-magnetic phase.

As the results, the FePt grains embedded in the MgO matrix was studied to minimize the grain size of the FePt grains and segregate the magnetic phase ^[11]. It is significant to enhance the signal-to-noise ratio (SNR) of the film for recording ^[12]. Z. Dong, et al ^[13] showed the magnetization behavior was also affected by different grain size distribution by using 3D micromagnetic studies. However, the mechanisms of the doping amount of MgO in FePt films are still rarely investigated. The optimal amount of MgO non-magnetic phase can effectively decrease not only the exchange coupled effects between the hard magnetic L1₀-FePt grains, but also the grain size after high temperature annealing.

In this paper, the utilization of single crystal MgO substrate can induce the growth of the MgO grains because of the homogeneous structure. The well-isolated FePt grains separated by the MgO matrix, the magnetically decoupled hard phase grains, large magnetic anisotropy constant and high coercivity were obtained ^[14-15]. It provides a novel feasible method to fabricate the granular structure FePt films for high density magnetic recording media with low exchange coupling and high thermal stability.

Experimental

The FePt-MgO granular thin films and FePt/MgO bilayer thin films were fabricated by dc magnetron sputtering system at various temperatures (room temperature, 200°C and 300°C). (FePt)_{100-x}(MgO)_x films were prepared by co-sputtering. A Pt target was symmetrically distributed with fan shaped Fe chips. The purity of the Fe, Pt and MgO were higher than 99.99%. The substrate was MgO<100> single crystal material. The base pressure of the sputter chamber was higher than 7.0×10^{-5} Pa and the argon pressure was fixed at 0.7 Pa. The chemical composition of FePt phase was Fe₅₅Pt₄₅, which was determined by Energy dispersive spectrometry (EDS, NOVA NANOSEM 430). The as-deposited film was annealed in vacuum at 800°C for 1 hour. The total thicknesses of the FePt–MgO film were fixed at 60 nm. The FePt layer was 27nm and MgO layer was 33nm in the FePt/MgO bilayer film. The total thickness of bilayer thin film was consistent with the [FePt]₄₅[MgO]₅₅ film by varying the deposition rate and time of FePt and MgO. The RF sputtering powers of MgO were used to control the volume fraction of MgO in the FePt–MgO film. Since, the MgO content of the film was obtained by using the formula ^[16]: $[R_{\text{MgO}}/(R_{\text{FePt}}+R_{\text{MgO}})] \times 100$ vol. %, where R was the sputtering rate. The structure, magnetic properties and domains of the FePt films were characterized by X-ray diffraction (XRD, Philips X'Pert Pro, Cu-K), atomic/magnetic force microscope (AFM/MFM, Asylum Cypher), and vibrating sample magnetometer (VSM setup: PPMS-9 Quantum Design Co.) with a maximum applied field of 9T. The plane-view morphology and selected area electron diffractions (SAEDs) were measured by Transmission electron microscopy (TEM, JEOL JEM2100F).

Result and discussion

1.1 Phase and microstructure

Fig. 1 showed the XRD patterns for different composition FePt-MgO films annealed at 800°C. It was recognized that the L1₀ phase formation was prohibited by the deposition of FePt-MgO films at room temperature, because the superlattice diffraction peak (001) and fundamental peak (002) of L1₀ phase were not observed at the room temperature deposited films.

The L1₀-FePt phase transformation can be enhanced with increasing the deposition temperature from room temperature to 200°C. The films with small fraction of FePt(30%) were difficult to form L1₀-phase even after high temperature annealing, as shown in Fig. 1a. When the deposition

temperature was as high as 300°C, the FePt was almost decomposed into Fe₃O₄ and PtO for the higher diffusivity of O atoms at high temperature deposition. Also adding higher fraction of MgO(vol. 70%) was prone to generate the oxidation phase (Fe₃O₄ and PtO). Based on the theory of thermodynamics, the excessive density of oxygen element and sufficient thermal energy in the system provided the possibility to form the FeO_x, which was certified by the previous studies on FePt-ZnO films^[17]. The existence of fcc-FePt (200) peak was found in the annealed films and the PtO phase was disappeared for the increasing fraction of FePt, shown in Fig. 1b and Fig. 1c. Additionally, the strong L1₀-FePt superlattice diffraction peak (001) was appeared when the composition of the films was (FePt)₄₅(MgO)₅₅ and (FePt)₆₀(MgO)₄₀.

However, even if for the films with larger fraction of FePt, the oxidation phase can also be formed by increasing the deposition temperature. It is logical to evaluate the L1₀-FePt transformation degree by calculating the ordering parameter, which is obtained by^[18]:

$$S \cong 0.85 \left[\frac{I_{001}}{I_{002}} \right]^{1/2} \quad (1)$$

where $I(001)$ and $I(002)$ represent the integrated intensities of the (001) and (002) diffraction peaks from the XRD patterns (Fig. 1).

Fig. 1d showed the ordering parameter as the function of FePt deposition temperature. It was apparent to find that the ordering parameter was not able to calculate at the room temperature deposition due to the disappearance of L1₀-FePt (001) peak. When the deposition temperature was 200°C, L1₀-FePt ordering parameter was much higher than the films deposited at 300°C. It implied that the L1₀-FePt phase was beneficial to form at 200°C deposition. Fig. 2a showed the shift of FePt (111) peak from $2\theta=40.1^\circ$ to 41.6° as the deposition temperature elevating to 200°C. The inset indicated the L1₀-FePt (001) peak of the film deposited at 200°C had 2θ angle of 24° , which was slightly higher than that (23.5°) of the film deposited at 300°C. It indicated the L1₀ phase ordering was enhanced at 200°C deposition. For the (FePt)₆₀(MgO)₄₀ films, the L1₀-FePt (111) peak of the film deposited at 200°C also had higher 2θ angle than the film deposited at 300°C, shown in Fig. 2b. However, the FWHM of the L1₀-FePt (001) and (111) peaks was decreased, which indicated the increase of L1₀-FePt grain size with increasing FePt fraction.

The grain size was calculated by scherrer equation from XRD patterns and the results were shown in Fig. 2c. It demonstrated that the size of FePt grains became larger as the deposition

temperature increased. The grain size of 200°C or 300°C deposited films were about 23-26nm after 800°C annealing. Fig. 3 showed the plane-view morphologies of (FePt)₄₅(MgO)₅₅ films. In Fig. 3a, the average grain size was 25nm and the major phases were L1₀-FePt and MgO shown in the inset SAED, whereas, the granular structure can not be obviously seen in plane-view TEM dark field image. For the 300°C deposited film, shown in Fig. 3b, the FePt grain size was also about 24nm and the phase was not changed, which was consistent with the calculation results. The grain size of the different temperature deposited films was only slightly changed and the growth of FePt grains was effectively limited by MgO matrix. Additionally, the grain size of (FePt)₆₀(MgO)₄₀ film was larger than the other films, which was related to the not well-separated structure and the agglomeration of the FePt grains.

The decrease of ordering parameter for the FePt-MgO films at 300°C deposition was caused by the different thermal expansion coefficients of FePt and MgO. According to the previous research, both the thermal expansion coefficient and melting point of the materials reflects the atomic bonding and interaction force ^[19]. It concludes that if the the melting point of the material is high, it will reduce thermal expansion coefficient. The bonding force is stronger between Mg and O atoms because of higher melting point (2800°C) compared with FePt(about 1300°C). As the result, the linear thermal expansion of FePt is much easier based on the low melting point, which can also be calculated by the formula:

$$\alpha = \frac{L_2 - L_1}{L_0(t_2 - t_1)} = \frac{\Delta L}{L_0 \Delta t} \quad (2)$$

Also, Winkler, et al ^[20] found that the thermal expansion coefficient was related to the binding energy by referring the equation: $\alpha \propto 1/E_B$, where E_B represents the binding energy of the alloys. The weak bonding energy will induce the large thermal oscillation, which increases thermal expansion.

The formation of L1₀-FePt and the expansion of MgO matrix mechanisms depending on the deposition temperature from room temperature to 300°C are illustrated in Fig. 4. At room temperature deposition, both FePt and MgO grains have no sufficient energy for diffusion and expansion. FePt grains are embedded in the MgO matrix. It suggests that it is difficult to generate the L1₀ phase. As the result, the ordering parameter is relatively low and grain size is small at room temperature deposition.

After heating the substrate to 200°C, the size of both MgO matrix and FePt grain becomes larger and L1₀-FePt transformation process is started due to the enough thermal energy supply for the diffusion of Fe and Pt atoms. The competitive growth of both of the grains is started, but the FePt grain expansion and L1₀-ordering process is dominated due to the large thermal expansion coefficient. Charles, et al ^[21] also explained that the thermal expansion of MgO was anisotropic expansion, which extended the in-plane direction axis. Consequently, both the expansion directions of FePt and MgO are in-plane, which would cause the compression of perpendicular axis. However, when the deposition temperature is further increased to 300°C, the competitively growth among the grains are more obvious. The size of MgO grains became large in the films and the FePt grains were surrounded by MgO matrix. It is known that the L1₀ phase transformation is the process of in-plane axis expansion and perpendicular axis compression. The growth of FePt grains during ordering process is suppressed by the increasing number of growing MgO grains. Additionally, the atomic diffusivity is increased by elevating the deposition temperature, especially for the O atoms. As the result, the ordering parameter is decreased and oxidation phase (Fe₃O₄) is easily appeared, which has been observed by the XRD results.

In order to further illustrate the phase structure affected by MgO matrix, the FePt/MgO bilayer thin film was fabricated to make a comparison with (FePt)₄₅(MgO)₅₅ film. Fig. 5a showed the L1₀-FePt phases were appeared after high temperature annealing in FePt/MgO bilayer thin film. The FePt grain size of as-deposited FePt/MgO bilayer film was only 15 nm by calculation. However, the FePt grain size was sharply increased to 40 nm after 800°C annealing. Fig. 5b displayed that the FWHM of FePt/MgO bilayer film was much smaller than that of (FePt)₄₅(MgO)₅₅ film. It indicated adding MgO matrix between the FePt grains can minimize the grain size of FePt and form the well-separated granular structure.

1.2 Magnetic properties

After 800°C annealing, Fig. 6a showed the in-plane coercivity of (FePt)₃₀(MgO)₇₀ film deposited at 200°C was 3.1 kOe. It means the L1₀ phase transformation was started, but the loop showed the obvious two-phase magnetic behavior ^[22]. When the deposition temperature was increased to 300°C, the film had very low coercivity, shown in Fig. 6b. Since, the FePt decomposed due to the large MgO fraction (vol. 70%) and high oxygen atom diffusivity at elevated deposition temperature, which had been explained before. In Fig. 6e, the coercivity showed an upward trend as the

deposition temperature increased to 200°C and then dropped down at 300°C deposition. Both the in-plane and out-of-plane coercivity were the highest (6.5kOe and 4.8kOe, respectively) in the (FePt)₄₅(MgO)₅₅ film deposited at 200°C. Moreover, Fig. 6c showed the appearance of shoulder in the second quadrant, which was originated from the co-existed L1₀-FePt and fcc-FePt phases. It was investigated that adding non-magnetic phase would lead to the existence of two phases behavior after heat treatment due to the large exchange length ^[23]. In Fig. 6d, the shape of loop for the film deposited at 300°C was similar with that of the film deposited at 200°C. However, when the fraction of FePt increasing to vol. 60% in the film, the FePt grains were not well-separated by MgO matrix and the exchange coupling effects were enhanced, which caused the decrease of coercivity.

Meanwhile, the M-H loop of FePt/MgO bilayer film deposited at 200°C indicated that the in-plane coercivity was slightly decreased (5kOe) compared with that of (FePt)₄₅(MgO)₅₅ film, shown in Fig. 6e. The slope of the hysteresis loop (α_s) at the coercive point is used to reflect the coherency of magnetic reversal process and characterize the exchange coupling effects among FePt grains of in-plane hysteresis loop ^{[12],[16]}. α_s was obtained by the formula:

$$\alpha_s = 4\pi \left(\frac{dM}{dH} \right)_{H=H_C} \quad (3)$$

The bilayer film showed a behavior like typical rigid magnetic reversal process and the α value was 3, which represented the enhancement of exchange coupling effect. But the α value was 2.2 for the (FePt)₄₅(MgO)₅₅ film deposited at 200°C, suggesting the exchange coupling among the FePt grains was largely reduced and the domain wall continuous motion was blocked due to adding MgO. As the result, the high coercivity of the (FePt)₄₅(MgO)₅₅ film was achieved by the isolated structure.

Fig. 7 displayed the 2D surface morphologies and domain distribution of 200°C deposited (FePt)₄₅(MgO)₅₅ film and FePt/MgO bilayer film annealed at 800°C. The clear granular structure can be observed by AFM in (FePt)₄₅(MgO)₅₅ film, shown in Fig. 7a. Meanwhile, Fig. 7b displayed the smoother surface for FePt/MgO bilayer film, despite of some large grains. Additionally, combining analyzing the domain structure, it can be seen that small size domains were uniformly distributed in the (FePt)₄₅(MgO)₅₅ film, shown in Fig. 7c. It indicated that the large grains were MgO matrix in Fig. 7a, with mean size of approximately 100nm. However, Fig. 7d displayed that the large size domains was appeared due to the agglomeration of FePt grains, which were exceed the single domain size. These large domains were formed by exchange coupling effects. The strong coupled

effects between the grains made their moments deviate from the magnetic easy axis, aligning to the same direction. Since, the large size coupled domains may decrease the coercivity. It demonstrated that the domain size and the magnetic properties can be adjusted by adding MgO non-magnetic matrix.

Conclusion

We have investigated the effects of adding various fraction of non-magnetic MgO phase on the phase structure, morphology and magnetic properties of the FePt-MgO granular thin films. It is found that the fraction of MgO phase has significant effects on the microstructure and magnetic properties of the FePt phase. It is also revealed that the room-temperature co-deposition does not lead to formation of ordered $L1_0$ -FePt hard magnetic phase even after a high temperature annealing. The optimal deposition temperature for the $L1_0$ phase formation is at 200°C with the composition of $(\text{FePt})_{45}(\text{MgO})_{55}$, which is attributed to the well-separated granular structure with reducing exchange coupling effects. Compared with FePt/MgO bilayer film, it indicates that the MgO phase indeed segregates the FePt grains, which gives rise to high coercivity of the FePt-MgO granular film.

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Figure Captions:

Fig. 1 XRD patterns and the ordering parameter for FePt-MgO films annealed at 800°C, (a) (FePt)₃₀(MgO)₇₀; (b) (FePt)₄₅(MgO)₅₅; (c) (FePt)₆₀(MgO)₄₀; (d) ordering parameter depended on deposition temperature

Fig. 2 Magnified XRD patterns and the grain size variation of FePt-MgO films annealed at 800°C, (a) (FePt)₄₅(MgO)₅₅; (b) (FePt)₆₀(MgO)₄₀; (c) Average FePt grain size depended on deposition temperature

Fig. 3 Plane-view TEM images and SAEDs for (FePt)₄₅(MgO)₅₅ film, (a) deposited at 200°C; (b) deposited at 300°C

Fig. 4 Schematic of the competitively growth of FePt and MgO grains depended on deposition temperature

Fig. 5 (a) XRD patterns for FePt/MgO bilayer films deposited at 200°C and annealed at 800°C; (b) Magnified XRD patterns for 200°C deposited FePt/MgO bilayer films and (FePt)₄₅(MgO)₅₅ film annealed at 800°C

Fig. 6 Hysteresis loops and in-plane coercivity variations of FePt-MgO granular films and FePt/MgO bilayer film annealed at 800°C, (a) (FePt)₃₀(MgO)₇₀ film deposited at 200°C; (b) (FePt)₃₀(MgO)₇₀ film deposited at 300°C; (c) (FePt)₄₅(MgO)₅₅ film deposited at 200°C; (d) (FePt)₆₀(MgO)₄₀ film deposited at 200°C (e) FePt/MgO bilayer film deposited at 200°C; (f) in-plane coercivity variations corresponding to deposition temperature of FePt-MgO granular films

Fig. 7 AFM and MFM images of FePt-MgO granular film and FePt/MgO bilayer film annealed at 800°C, (a) AFM image of (FePt)₄₅(MgO)₅₅ film deposited at 200°C; (b) AFM image of FePt/MgO bilayer film deposited at 200°C; (c) MFM image of (FePt)₄₅(MgO)₅₅ film deposited at 200°C; (d) MFM image of FePt/MgO bilayer film deposited at 200°C

Figure 1

Figure 2

Figure 3

Figure 4

Room temperature
deposition

200 °C
deposition

300 °C
deposition

Figure 5

Figure 6

Figure 7