Microstructure of $L1_0$ FePt thin films with anisotropic interfacial energy coefficients and anisotropic atomic mobilities

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The microstructure evolution of FePt-X (segregant) thin films were studied by employing a three-dimensional phase field model. Simulated results show that in the absence of substrate constraint related with elastic energy, the morphology of the FePt-X thin films significantly depends on the film thickness, anisotropic interfacial energy coefficients, and anisotropic atomic mobility. Small in-plane anisotropic interfacial energy coefficients or out-of-plane anisotropic mobility of atom diffusion are beneficial to induce columnar shape FePt grains, using any one of parameters of which can make the FePt grains keep columnar shape when FePt-X thin films thickness is 10 nm. Using segregant with both proper in-plane anisotropic interfacial energy coefficients and out-of-plane anisotropic mobility can make the FePt grains keep columnar shape when the FePt-X thin films thickness reaches to at least 15 nm.

Index Terms—FePt, anisotropic mobility, anisotropic interfacial energy coefficients, columnar microstructure, phase field.

I. INTRODUCTION

$L1_0$ FePt thin film, as a candidate for the potential Heat Assisted Magnetic Recording (HAMR) media due to its large magnetic anisotropy ($K_u \approx 7 \times 10^7$ erg/cc), has been extensively studied in many research groups [1-3]. The HAMR media demands that the $L1_0$ FePt thin films must have small grain size, columnar FePt grains, and high coercivity. Especially, the aspect ratio of each columnar FePt grain should be large enough so as to keep the thermal stability of the magnetic moment in the FePt-X thin films [4-5]. Recently, the microstructures of FePt were extensively studied in experiments for preparing the columnar FePt grains by changing various fabrication conditions, e.g., temperature, deposition pressure, the choice of segregants, segregant volume fractions, and thickness of the thin film [6-8]. In order to study the formation mechanisms of microstructure in FePt-X thin films, the phase field model was used to simulate the morphology of FePt-X thin films [9-10]. The simulated results show that the morphology of the FePt-X thin films significantly depends on the interfacial energy, film thickness, and anisotropic atomic mobility. And low interfacial energy can induce small FePt grains, while the high interfacial energy can form big FePt grains when the film thickness is fixed. It also shows that the formation of columnar or the bilayer microstructure of FePt highly depends on the film thickness [10]. However, there are still many challenges lie ahead to fabricate the desired well-separated slim and tall FePt grains from laboratory, the underlying physical mechanisms are still very deficient at present on how to improve the microstructure of the FePt-X thin films media for HAMR [11]. Accordingly, it

is extremely essential to investigate the mechanisms about how to form the columnar FePt grains for future HAMR media through theoretical simulation, and thus to guide the experiments. In this work, based on our previous study about the microstructure of FePt-X thin films [10], the influence of film thickness, anisotropic interfacial energy coefficients and anisotropic atomic mobility to morphology of L_{10} FePt-X thin films were studied in detail by using the three-dimensional phase field model.

II. PHASE FIELD METHOD

Phase field method is a powerful tool to simulate the microstructure evolution in various materials [12-15]. In this work, a conserved field composition $c(\mathbf{r}, t)$ and a nonconserved field order degree $s(\mathbf{r}, t)$ are chosen as the phase field variables to describe the microstructure change of the FePt-X thin films. $c(\mathbf{r}, t)$ describes the concentration of FePt. Thus, the concentration of amorphous segregant is $1-c(\mathbf{r}, t)$ based on the mass conservation. $s(\mathbf{r}, t)$ describes the ordered FePt with $s=1$ and the disordered FePt with $s=0$. The spatial and temporal evolutions of composition and order degree are controlled by the conserved field equation (Cahn–Hilliard equation (1)) and nonconserved field equation (Allen–Cahn equation (2)), respectively as

$$\frac{\partial c(\mathbf{r}, t)}{\partial t} = M \nabla^2 \frac{\delta F}{\delta c(\mathbf{r}, t)} \quad (1)$$

$$\frac{\partial s(\mathbf{r}, t)}{\partial t} = -L \frac{\delta F}{\delta s(\mathbf{r}, t)} \quad (2)$$

where, F is the total free energy of the FePt-X thin films, which is constructed as

$$F = \int [f_{chem} + f_{grad} + f_{elas}] dV \quad (3)$$

M is the atomic mobility determined by both diffusivities of FePt and segregant, L is the kinetic coefficient related to ordering, V is the system volume of the FePt-X thin films, f_{chem} denotes the chemical free energy density. The expression of f_{chem} can be found in Reference [9]. f_{grad} is the gradient energy density calculated by $f_{grad} = \kappa_c (\nabla c_{FePt})^2 + \kappa_s |\nabla s|^2$, where κ_c and κ_s are gradient energy coefficients of composition field $c(\mathbf{r}, t)$ and order degree fields $s(\mathbf{r}, t)$, respectively. f_{elas} is the elastic energy density, which is not considered in the current simulations. M and κ_c are usually assumed that only the diagonal values are non-zero for the tensor matrix. They have the following matrix form for isotropic and anisotropic materials, respectively [16-18],

$$M_{iso} = \begin{pmatrix} M_{11} & 0 & 0 \\ 0 & M_{11} & 0 \\ 0 & 0 & M_{11} \end{pmatrix} \quad M_{anis} = \begin{pmatrix} M_{11} & 0 & 0 \\ 0 & M_{11} & 0 \\ 0 & 0 & M_{33} \end{pmatrix}$$

$$\kappa_{iso} = \begin{pmatrix} \kappa_{11} & 0 & 0 \\ 0 & \kappa_{11} & 0 \\ 0 & 0 & \kappa_{11} \end{pmatrix} \quad \kappa_{anis} = \begin{pmatrix} \kappa_{11} & 0 & 0 \\ 0 & \kappa_{11} & 0 \\ 0 & 0 & \kappa_{33} \end{pmatrix}$$

In this simulation, the real interfacial energy coefficients are κ_c times κ_{iso} or κ_{anis} .

The coupled equations (1)–(2) were numerically solved by using the finite difference method under the periodic boundary conditions. The 128×128 discrete grid points were used in the x and y directions, which correspond to real size $50 \times 50 \text{ nm}^2$, and the grid points in z direction vary by choosing different FePt-X thin films thicknesses. The grid spacing in real space is about 0.39 nm . The time step for integration is 10^{-4} . The evolution time of system is 100000 steps. The initial structure of FePt-X thin films with a small conserved composition fluctuation and a small order degree fluctuation are set by using a computer-generated random number. The used calculating parameters are: ordering temperature is 973 K , the atomic ratio is $\text{Fe}_{55}\text{Pt}_{45}$, the interfacial energy γ_s between FePt and segregant X is 1.82 J/m^2 for isotropic case, the composition of FePt is set as

0.4 according to the TEM image in the experimental work [1]. Then, the influences of the film thickness, anisotropic interfacial energy coefficients, and anisotropic atomic mobility on the microstructure of the FePt-X thin films in three-dimensions have been studied.

III. RESULTS AND DISCUSSION

The anisotropic interfacial energy plays an important role in determining the microstructure of material [19,20]. And there are few related reports in study of FePt-X thin films. To this end, we first studied the effect of anisotropic interfacial energy on the structure feature of FePt-X thin films. Figure 1 shows the microstructure of the FePt-X thin films with different values of anisotropic interfacial energy. The film thickness keeps the same as 10 nm for all simulations. The dimensionless atom mobilities in the x , y , and z directions are set as $M_x=M_y=M_z=1.0$, and the dimensionless interfacial energy coefficient κ_z is set as 1.0. The interfacial energy coefficients $\kappa_x=\kappa_y$ in the x and y directions decrease from 1.0 to 0.2 for four simulation cases. The amorphous segregant, the ordered FePt, and the disordered FePt are represented by a blue, yellow, and red color in the figures, respectively. Fig. 1(a) shows the bilayer microstructure due to the thicker FePt-X thin films and isotropic M and κ , and it also shows the large FePt grain size in the FePt-X thin films. Then the number of isolated FePt grains is relatively small when κ is isotropic. When $\kappa_x=\kappa_y$ decreases to 0.6, the FePt grains size decreases and more FePt grains become isolated, and the columnar FePt grains appear as shown in Fig. 1(b). The increase of the columnar FePt grains is more obvious when $\kappa_x=\kappa_y$ decreases to 0.4 as shown in Fig. 1(c). And when $\kappa_x=\kappa_y$ decreases to 0.2, the FePt grains almost completely become columnar shape and smaller further. This is because the evolution of the system tends to decrease the interfacial energy, thus less interfaces is favorable in order to decrease the system energy, which means that the smaller interfacial energy will give more interfaces. This is also consistent with our previous study that the small interfacial energy between FePt and X can result in small FePt grains [10].

Fig. 1 The microstructures of the FePt-X thin films when the film thickness is 10 nm and with $M_x=M_y=M_z=1.0$, $\kappa_z=1.0$, and $\kappa_x=\kappa_y$ are set as: (a) 1.0; (b) 0.6; (c) 0.4; (d) 0.2.

Figure 2 shows the variations of microstructure of the FePt-X thin films with different film thicknesses, where $M_x=M_y=M_z=1.0$, $\kappa_x=\kappa_y=0.2$, and $\kappa_z=1.0$. The film thickness varies from 8 to 15 nm. As shown in Fig. 2(a) and Fig. 2(b), the FePt-X thin films with the film thickness 8 and 10 nm have the columnar shape FePt grains. When the thickness of FePt-X thin films increases to 12 nm, the number of columnar shape FePt-X grains decrease as shown in Fig. 2(c). Fig. 2(d) clearly shows the number of bilayer FePt grains increases and the amount of columnar FePt grains decreases largely when the film thickness increases to 15 nm. Therefore, the columnar shape grains are not favorable in thicker films.

Fig. 2 The microstructures of the FePt-X thin films with different thicknesses when $\kappa_x=\kappa_y=0.2, \kappa_z=1.0$ and $M_x=M_y=M_z=1.0$. (a) 8 nm; (b) 10 nm; (c) 12 nm; (d) 15 nm.

Our previous studies demonstrated that selecting the materials with the anisotropic mobility as a segregant is vital to prepare the columnar microstructure of the FePt-X thin films [10]. In this work, we study the effect of film thickness on the microstructure of FePt-X thin films with anisotropic mobility. The gradient coefficients are $\kappa_x=\kappa_y=\kappa_z=1.0$. The anisotropic mobilities are set as $M_x=M_y=1.0, M_z=0.01$. The film thickness varies from 8 to 15 nm. Figure 3(a) and Fig. 3(b) show that the columnar shape FePt grains distribute in the segregant matrix in the FePt-X thin films when t is 8 and 10 nm, and the FePt grains become more isolated in Fig. 3(b) compared with Fig. 3(a). However, when the film thickness increases to 12 nm, the bilayer and semi-spherical FePt grains begin to be formed, as shown in Fig. 3(c). Fig. 3(d) shows that the FePt grains are still layer by layer or semi-spherical shape when the FePt-X thickness reaches to 15 nm.

Fig. 3 The microstructures of the FePt-X thin films with $\kappa_x=\kappa_y=\kappa_z=1.0, M_x=M_y=1.0$ and $M_z=0.01$ when film thicknesses is (a) 8 nm; (b) 10 nm; (c) 12 nm; (d) 15 nm.

Figure 4 shows the microstructure change of the FePt-X thin films with different film thickness when $M_x=M_y=1.0, M_z=0.01, \kappa_x=\kappa_y=0.2, \kappa_z=1.0$. The film thickness changes from 8 to 18 nm. Figure 4(a) shows the microstructure of the FePt-X thin films when the film thickness is 8 nm. It can be found that FePt grains become smaller and well isolated compared with cases of using anisotropic interfacial energy in Fig. 2(a) and anisotropic mobility in Fig. 3(a). As shown in Fig. 4(b) and Fig. 4(c), when the FePt-X film thickness increase to 12 and 15 nm, the FePt grains can still keep the good columnar shape. Fig. 4(d) shows that some FePt grains are still columnar shape but some FePt grains are bilayers microstructure when the FePt-X film thickness reaches to 18 nm. It can be seen that the effect of anisotropic energy and anisotropic mobility on columnar FePt grains in FePt-X thin films is restricted by the film thickness. These results demonstrate that using the doping materials with the anisotropic mobility of atom diffusion and anisotropic interfacial

energy coefficients are essential to fabricate FePt thin films with columnar shape grain.

Fig. 4 The microstructures of the FePt-X thin films with $\kappa_x=\kappa_y=0.2$, $\kappa_z=1.0$, $M_x=M_z=1.0$, and $M_y=0.01$ when film thicknesses is (a) 8 nm; (b) 12 nm; (c) 15 nm; (d) 18 nm.

IV. CONCLUSION

The microstructural evolutions of the FePt-X thin films were studied by using a three-dimensional phase field model. The simulation results demonstrate that both the anisotropic interfacial energy and anisotropic atom mobility play important roles in determining the microstructure of the FePt-X thin films. When using segregant with small in-plane anisotropic interfacial energy coefficients, the FePt grains are inclined to form small and columnar shape due to low interfacial energy and which results in more number of FePt grains. In the case of using segregant with small out-of-plane anisotropic mobilities, it is also beneficial to form the columnar FePt grains since the diffuse of atoms in the in-plane direction is faster than the out-of-plane direction. While using segregant with both small in-plane anisotropic interfacial energy coefficients and out-of-plane anisotropic mobilities, the FePt-X thin films thickness reaches up to 15 nm can still keep the FePt grains in columnar shape. These results indicate that using the segregant with the anisotropic interfacial energy coefficients and the anisotropic atom mobility are beneficial to fabricate FePt thin films with columnar shape grain. The effect of substrate constraint on the microstructure of the FePt-X thin films will be studied in our future work.

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