

# Atomic Ordering Kinetics of FePt Thin Films

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In the present study, the nucleation and growth kinetics of the  $L1_0$  ordered domains in FePt thin films were studied by using X-ray diffraction (XRD) measurements combined with transmission electron microscopy (TEM). A small nucleation activation energy  $E_n = (0.5 \pm 0.1)$  eV and a relatively large growth activation energy  $E_g = (0.9 \pm 0.1)$  eV for the  $L1_0$  ordered domains are separately determined. The high ratio  $E_g/E_n > 1$  yields an understanding of a high  $L1_0$  ordering temperature for the FePt thin films used for magnetic recording media.

**Keywords:**  $L1_0$  Ordered Domains, Ordering Kinetics, Activation Energy.

## 1. INTRODUCTION

FePt and CoPt thin films with an  $L1_0$  phase are potential candidates for ultrahigh density magnetic recording media since the  $L1_0$  ordered FePt and CoPt crystals have a high magnetocrystalline anisotropy energy density  $K_u = (4 - 7) \times 10^7$  erg/cm<sup>3</sup>.<sup>1-3</sup> As-deposited FePt and CoPt thin films, however, have a disordered face-centered-cubic structure (i.e., A1 structure) and show soft magnetic properties. The phase transformation from A1 disordered structure to  $L1_0$  ordered structure in the films occurs usually at high temperatures above 773 K.<sup>4-6</sup> The high transformation temperature is unfavorable for the practical application of FePt thin films and will lead to a large grain size, e.g.,  $D > 30$  nm, in the thin films,<sup>2,7-9</sup> which will degrade the signal-to-noise ratio of the magnetic recording media. These problems prevent the materials from practical applications and have been the subject of many experimental studies.<sup>10-14</sup> In the past decade, a lot of techniques, e.g., element addition,<sup>10</sup> ion irradiation,<sup>11</sup> gas annealing,<sup>12</sup> the application of the underlayer and the top layer<sup>13,14</sup> etc., have been successfully employed to lower the  $L1_0$  ordering temperature. For further lowering the ordering temperature a fundamental understanding of the high  $L1_0$  ordering temperature of FePt and CoPt thin films for manufacturing magnetic recording media is of particular importance. The disorder (A1)-order ( $L1_0$ ) phase transition is of first order and occurs by nucleation and growth of the ordered domains.<sup>7,15</sup> Therefore, a detailed study on the nucleation and growth kinetics of the  $L1_0$  ordered domains in FePt films is of great significance for the understanding

of the  $L1_0$  ordering transformation process. The  $L1_0$  ordering kinetics in the FePt and CoPt thin films have been investigated by employing differential scanning calorimetry (DSC) and X-ray diffraction (XRD) technique.<sup>15,16</sup> These studies, however, only yield the general information on the phase transformation from the A1 disordered structure to the  $L1_0$  ordered structure in the films, and do not unveil the nucleation and growth mechanisms during the ordering process. In the present study, we report the kinetics studies of the nucleation and growth processes of the  $L1_0$  ordered domains in FePt thin films by employing XRD measurements combined with transmission electron microscopy (TEM) using individual annealing-temperature dependencies of the nucleation and growth parameters.

## 2. EXPERIMENTAL DETAILS

FePt thin films with a thickness of about 20 nm were deposited on natively oxidized Si (100) substrates by dc magnetron sputtering from a binary Fe<sub>50</sub>Pt<sub>50</sub> alloy target at an argon pressure of 2.0 Pa at room temperature. An appropriate temperature range from  $T = 693$  to 773 K was chosen to carry out the isothermal annealing of the as-deposited FePt thin films in a vacuum chamber with a pressure of  $p < 10^{-4}$  Pa. The heating and cooling processes for each annealing treatment were performed in a ramp rate range from 100–150 °C/min, and the holding temperature was kept in the range of  $\pm 1$  °C.

The microstructure of the FePt thin films was studied by employing XRD measurements using a thin film attachment with a grazing incidence diffraction and Cu  $K_\alpha$  radiation and TEM observations. The size of the  $L1_0$  ordered domains was calculated from the broadening of

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the superlattice diffraction peaks of the FePt crystals using the Williamson-Hall method.<sup>17</sup> The volume fraction of the  $L1_0$  ordered domains was determined using the intensity of the diffraction peaks of the  $L1_0$  and fcc phases, and the contributions from different phases to the intensity were carefully deconvoluted.<sup>16</sup>

### 3. RESULTS AND DISCUSSION

The XRD spectrum of the as-deposited FePt thin film (see Fig. 1) shows that the film consists of the FePt crystals with a disordered face-centered cubic structure (e.g., A1 structure). The mean grain size of the disordered FePt crystals was determined to be  $\sim 5$  nm calculated by employing the Williamson-Hall method, which can be further confirmed by the TEM analysis shown in Figure 2(a). The appearance of the superlattice (001) and (110) diffraction peaks of the FePt film after annealing at  $T = 723$  K for 1 second indicates the occurrence of the ordering transformation (see Fig. 1). At this annealing condition, the size of the  $L1_0$  ordered domains was determined to be  $d \sim 3.3$  nm, which is in agreement with the TEM image presented in Figure 2(c). The intensity of the superlattice diffraction peaks increases with annealing time, indicating an increase of the  $L1_0$  ordering degree in the FePt thin films.

Figure 3 shows the annealing time dependence of the mean size of the  $L1_0$  ordered domains of the FePt thin

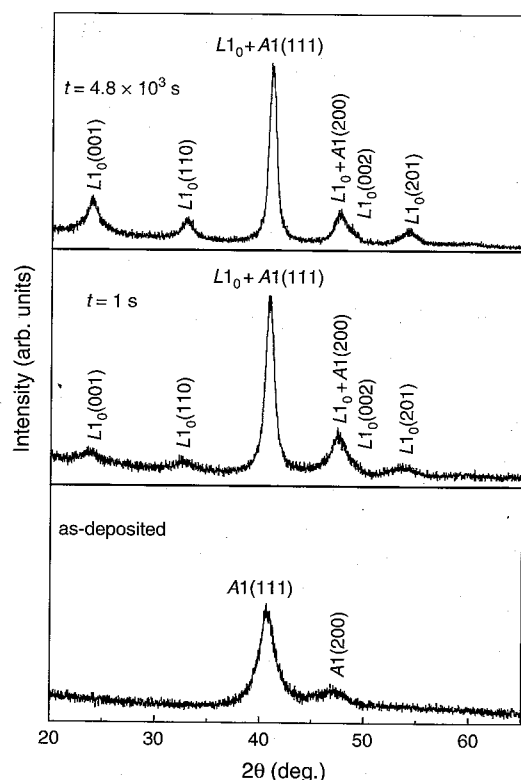


Fig. 1. XRD spectra of FePt thin films before and after annealing at a temperature of  $T = 723$  K for different time.

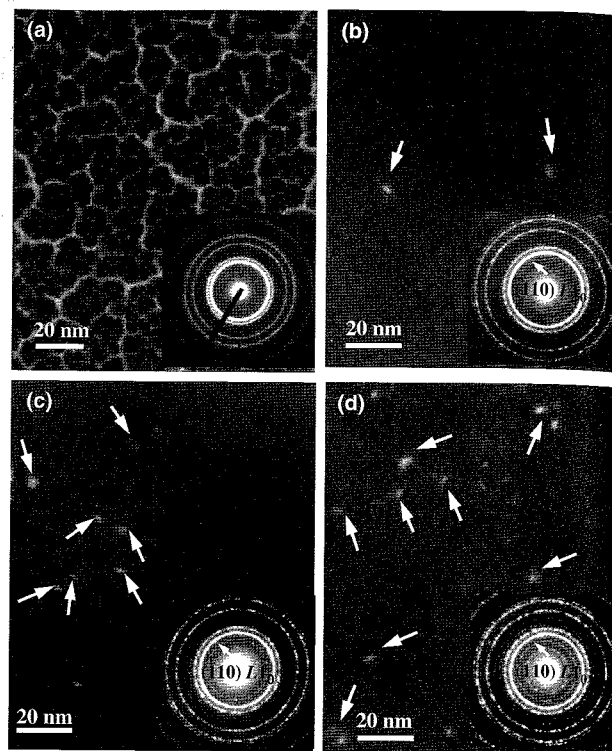


Fig. 2. Bright-field TEM image of the as-deposited FePt thin film (a) and the dark-field TEM images of FePt thin film annealed at  $T = 693$  K (b),  $T = 723$  K (c) and  $T = 753$  K (d) for 1 s made using the (110) super lattice reflections (see the inset).

films after annealing at different temperatures. The size of the  $L1_0$  ordered domains exhibits an exponent-shaped increase with the increase of the annealing time  $t$ . We define the time needed to reach the saturation-state growth rate (i.e.,  $\partial d/\partial t = 0$ ) as the saturation time constant  $t_E$ . It is quite clear that  $t_E$  varies with the annealing temperature.

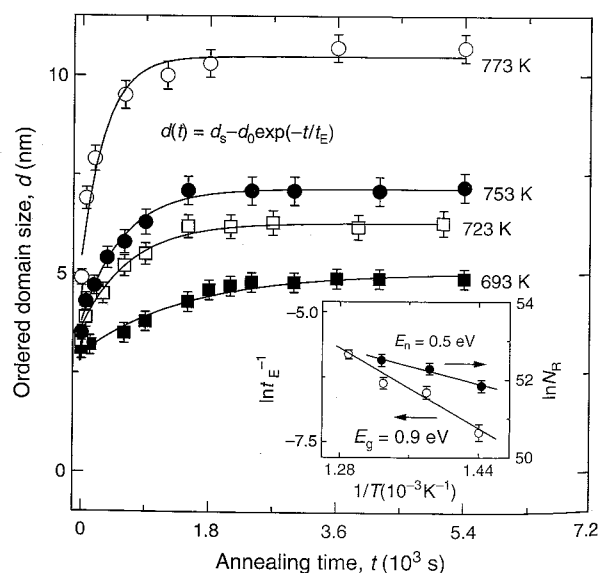


Fig. 3. Annealing time dependence of the size of the  $L1_0$  ordered domains in FePt thin films at different temperatures.

To determine the time constant  $t_E$ , the experimental data shown in Figure 3 were fitted by

$$d(t) = d_s - d_0 \exp(-t/t_E) \quad (1)$$

where  $d_s$  is the size of the ordered domain with  $\partial d/\partial t = 0$  and  $d_0$  is a parameter.

The saturation time constant  $t_E$  decreases with increasing the annealing temperature  $T$  (see the inset in Fig. 3), which indicates that the growth of the  $L1_0$  ordered domain is a thermally activated process. Assuming the saturation rate  $t_E^{-1}$  has an Arrhenius temperature dependence,<sup>18–20</sup> we have

$$t_E^{-1} = t_{E,0}^{-1} \exp(-E_g/k_B T) \quad (2)$$

where  $E_g$  is the activation energy for the growth of the  $L1_0$  ordered domain,  $t_{E,0}$  is the preexponential factor, and  $k_B$  is the Boltzmann's constant. An Arrhenius plot yields a growth activation energy  $E_g = (0.9 \pm 0.1)$  eV for the  $L1_0$  ordered domain in the FePt films (see the inset in Fig. 3).

The size of the  $L1_0$  ordered domain is almost unchangeable, e.g.,  $d \approx 3.2$ – $3.5$  nm (see Fig. 3), after annealing in the temperature range from 693 K to 753 K for 1 s, which is confirmed by TEM analyses [see Figs. 2(b), (c), and (d)]. Therefore, the increase of the volume fraction of the  $L1_0$  ordered domain in the annealing temperature range depends dominantly on the nucleation of the  $L1_0$  ordered domain. This allows the nucleation process of the ordered domain to be investigated independently. According to the Arrhenius temperature dependence of the nucleation rate  $N_R$ ,<sup>18,21</sup> a nucleation activation energy  $E_n = (0.5 \pm 0.1)$  eV for the  $L1_0$  ordered domain is determined from the slope of the plot of  $\ln(N_R)$  versus  $1/T$  shown in the inset of Figure 3.

The present kinetics studies of the nucleation and growth of  $L1_0$  ordered domains can be directly employed for an immediate understanding of a high  $L1_0$  ordering temperature for the practical application, e.g.,  $T \geq 773$  K in FePt thin films.<sup>2,4–8,15</sup> The high equilibrium ordering temperature ( $T = 1573$  K) together with the large magnitude of the transformation enthalpy,  $H = (-10.2 \pm 2.1)$  kJ/g-atom,<sup>15</sup> for the FePt thin films will lead to a large driving force and thus a lower nucleation barrier for the formation of the  $L1_0$  ordered domains in the films, e.g.,  $E_n = 0.5$  eV determined above. However, the high ratio  $E_g/E_n > 1$  indicates a difficult growth for the  $L1_0$  ordered domains, which constrains the  $L1_0$  ordering transformation in the FePt thin films. As a result, a high temperature is required for the phase transformation.

#### 4. CONCLUSIONS

The nucleation activation energy  $E_n = (0.5 \pm 0.1)$  eV and the growth activation energy  $E_g = (0.9 \pm 0.1)$  for the

$L1_0$  ordered domains in FePt thin films are determined.  $E_g/E_n > 1$  yields a fundamental understanding of a high  $L1_0$  ordering temperature for the FePt thin films. The present work is of importance for reducing the  $L1_0$  ordering temperature of the FePt thin films and thus is of wide interest.

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