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Roles of nanocrystal interfaces on $L1_0$ ordering phase transitions in FePt thin films

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ABSTRACT

Identifying the role of interfaces in nanostructured materials on their phase transitions is of quite interest for microstructure manipulations. In the present study, we demonstrate that the increase of nanocrystal interfaces in as-deposited FePt thin films enhances the nucleation rate of $L1_0$-ordered domains and simultaneously inhibits their growth during the $L1_0$ ordering phase transition in the thin films. The enhanced nucleation rate leads to an accelerated $L1_0$ ordering process for the thin films. The $L1_0$-FePt thin films with small grain and ordered domain sizes are successfully yielded by interface engineering.

1. Introduction

The phase transitions of nanostructured materials have attracted considerable attention since their remarkable functional properties and novel physical phenomena are dictated by the microstructures yielded from the phase transitions [1–4]. Due to tremendous interfaces existing in nanostructured materials [5], these interfaces like the structures (e.g., size) of nanocrystals have quite significant effects on their functional properties and structural stability, e.g., phase transitions. Therefore, a profound understanding of interfacial structures and the role of interfaces on phase transitions in nanostructured solids is intensely required. Although substantial progress has been made in the study of the nature, structural characteristic, diffusion, migration, and kinetics of the interfaces in nanostructured solids [6–11], a good understanding of the effect of nanocrystal interfaces on phase transitions is still required for manipulating the phase transitions and thus the microstructures of the materials.

Equiatomic FePt thin films with an $L1_0$-ordered structure are potential candidates for ultrahigh density magnetic recording media [12]. Room-temperature as-deposited FePt thin films are made of *nanocrystals* with a disordered face-centered-cubic ($A1$) structure, and a disorder–order phase transition is indispensable to produce the $L1_0$-ordered FePt thin films [13,14]. A detailed study of the role of nanocrystal interfaces on the $L1_0$ ordering transition is, therefore, of importance for tuning the phase transition in order to lower the transition temperature and optimize the microstructures of the $L1_0$-FePt thin films. In this work, the effect of nanocrystal interfaces on the $L1_0$ ordering transition in FePt thin films has been studied. We demonstrate that an increased interface fraction in as-deposited FePt thin films enhances the nucleation rate of $L1_0$-ordered domains and simultaneously inhibits their growth, and the $L1_0$-FePt thin films with small grain and ordered domain sizes are successfully yielded by interface engineering.

2. Experimental details

$Fe_{49.5}Pt_{50.5}$ films (named as FePt thin films below) with a thickness of ~40 nm were deposited at room temperature on native-oxidized Si substrates by dc magnetron sputtering from a binary Fe–Pt composite target at argon pressures of $p = 4$ Pa and $p = 2$ Pa. The goal of employing different sputtering pressures is to yield FePt thin films with various nanocrystal size and thus different interface fractions. The initial ratio of the Fe and Pt elements in the composite target was adjusted for different sputtering pressures in order to keep the same composition for the deposited thin films. The composition of the FePt thin films was checked by employing inductively coupled plasma atomic emission spectrometry (ICP-AES). To attain the same thickness for the FePt films deposited under different sputtering pressures, an accurate measurement for the growth rate of the FePt thin film under respective sputtering pressure was carried out by cross-section TEM studies and guided by the growth rate the sputtering time was adjusted. As-deposited FePt thin films were annealed in a vacuum chamber under a pressure of $p < 10^{-5}$ Pa at temperatures from $T = 350$ to 600 °C for 30 min, respectively.

The size of FePt nanocrystals and $L1_0$-ordered domains in as-prepared and annealed thin films was determined by transmission electron microscope (TEM) observations and X-ray diffraction (XRD) studies based on the broadening of diffraction peaks using the Williamson–Hall method [15]. The dark-field images of $L1_0$-ordered domains were made by using the $L1_0$-superlattice (1 1 0) reflection. The size distribution of the grains and the ordered domains was determined from the bright-field and dark-filed TEM images in sample regions of ~20 μm^2, respectively. The interface fraction f_{in} of FePt nanocrystals in as-deposited thin films was calculated according to $f_{in} = 3\Delta/d$ [16], where Δ is the interface thickness and d is

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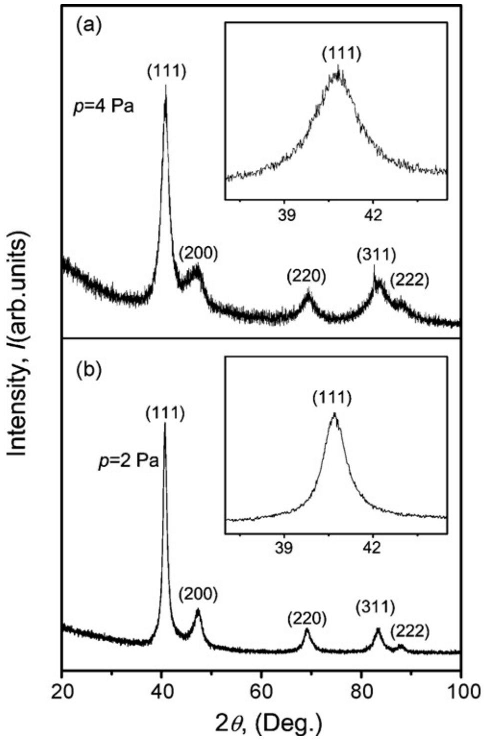

Fig. 1. XRD spectra of FePt thin films deposited at an argon pressure of (a) 4 Pa and (b) 2 Pa. The wider diffraction peaks of the films deposited at 4 Pa indicate a smaller size for FePt crystals in them as compared with those deposited at 2 Pa.

the grain size. The degree of $L1_0$ ordering S was calculated according to the method reported in Ref. [17]. The magnetic properties of FePt thin films were measured by employing a vibrating sample magnetometry with a magnetic field of 21 kOe at room temperature.

3. Results and discussions

XRD studies show that both the FePt thin films deposited at an argon pressure of $p = 2$ and 4 Pa have a disordered face-centered cubic ($A1$) structure [see Fig. 1], and further analyses demonstrate that the films deposited at 4 Pa have a smaller grain size ($d_{as} \sim 4$ nm) than those ($d_{as} \sim 8$ nm) deposited at 2 Pa, which is confirmed by TEM observations [see Fig. 2]. This is attributed to the fact that the high sputtering pressure yields deposited target atoms or clusters with low energy due to the large energy loss of sputtered Ar atoms from atom collisions, and thus makes a reduction in grain growth in the deposited films [18]. The volume fraction of nanocrystal interfaces in the thin films deposited at 4 Pa is determined to be ~75%, higher than that ~37% in the films deposited at 2 Pa.

After 450 °C annealing for 30 min, the FePt thin films with $d_{as} \sim 4$ nm have a smaller average grain size of D~10 nm [Fig. 3(a)] as compared with that $D \sim 17$ nm for the thin films with $d_{as} \sim 8$ nm [Fig. 3(b)]. This anomalous crystallite growth may be attributed to a low interfacial energy for the nanocrystals in the FePt thin films deposited at the high argon pressure of 4 Pa due to a lower energy for deposited atoms. Such an anomalous nanocrystal growth behavior has also been reported in other nanostructured systems [6,19], e.g., Ni–P nanocrystal alloys. Therefore, forming a low-energy interface has been proposed as an approach against grain growth in nanostructured solids [5]. The most striking result of the present studies is that the $L1_0$-ordered domains in the FePt thin films with $d_{as} \sim 4$ nm have a smaller size $d \sim 8$ nm and a higher number density as compared with those in the films with $d_{as} \sim 8$ nm [see Fig. 4(a) and (b)]. This directly demonstrates that the ordering transition in the former films depends dominantly on the nucleation of $L1_0$-ordered domains. The increase of interface fraction enhances the nucleation rate of the $L1_0$-ordered domains since nanocrystal interfaces may act as nucleation sites, as demonstrated in recent studies of FePt nanoparticles [20]. Even after annealing at 600 °C, the ordered domain size ($d \sim 32$ nm) in the thin films with $d_{as} \sim 4$ nm is still smaller than that $d \sim 50$ nm in those with $d_{as} \sim 8$ nm (not shown here). The results yielded above demonstrate that the $L1_0$-ordered domains have a slower growth in the films with more nanocrystal interfaces, which may be attributed to that the nanocrystal interfaces also play a pinning role on the growth of the ordered domains.

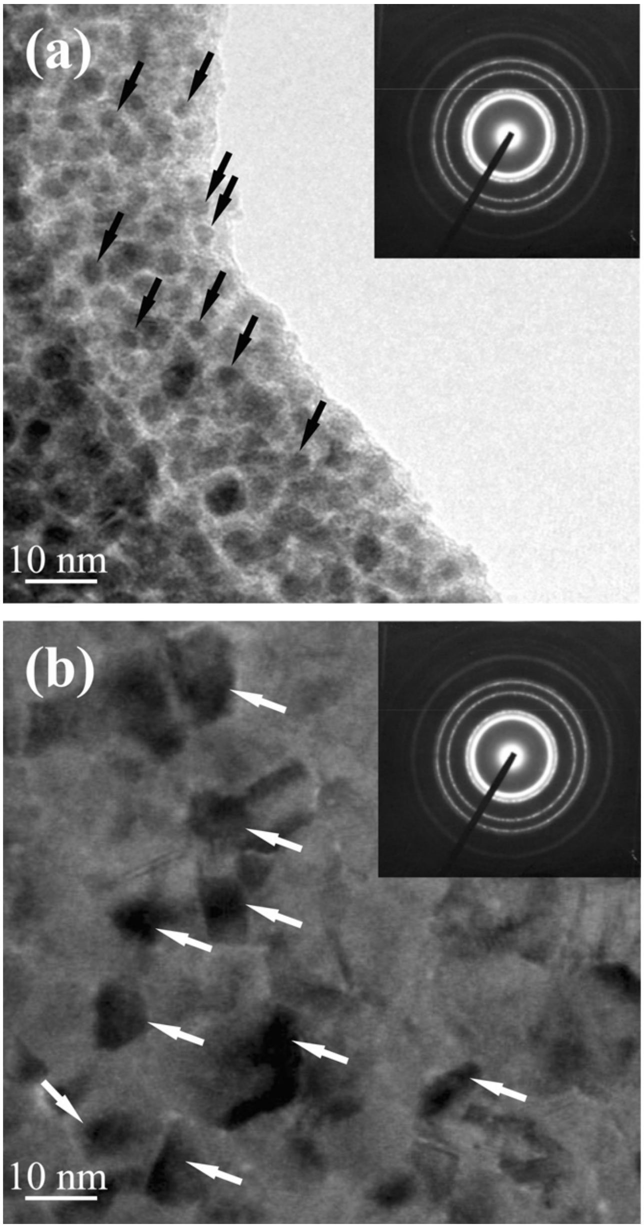

Fig. 2. Bright-field TEM images and electron diffraction (ED) patterns (the insets) of FePt thin films deposited at an argon pressure of (a) $p = 4$ Pa and (b) $p = 2$ Pa.

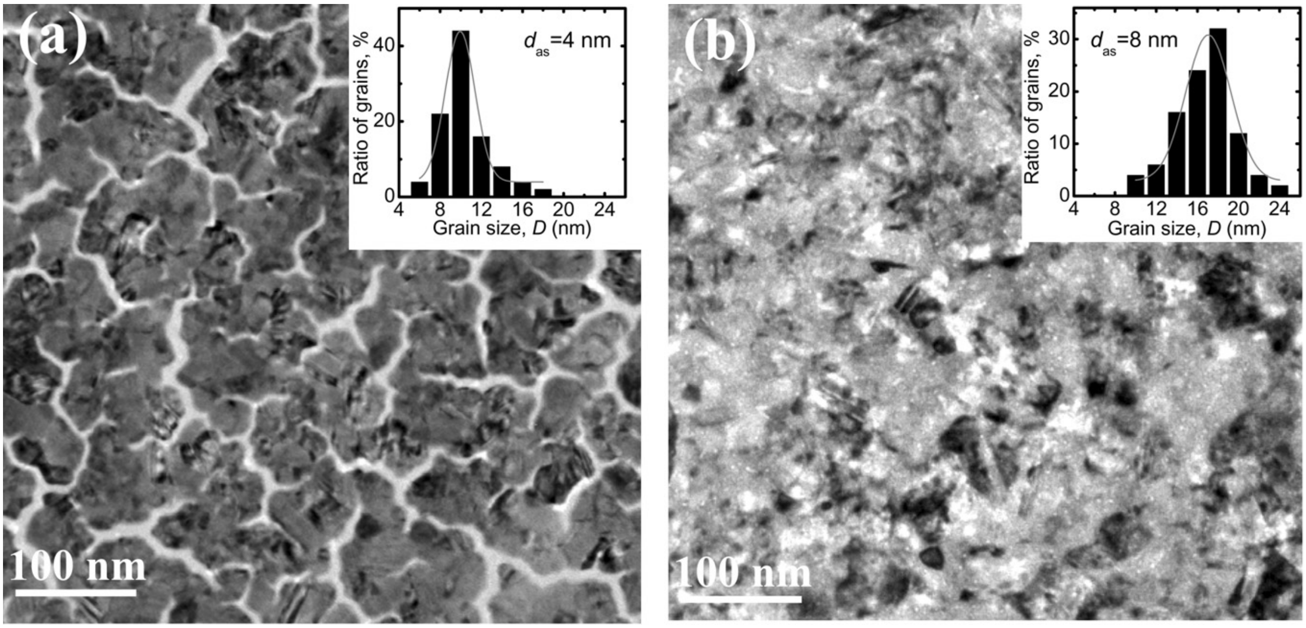

Fig. 3. Bright-field TEM images and grain size distributions of FePt thin films deposited at an argon pressure of (a) $p = 4$ Pa ($d_{as} \sim 4$ nm) and (b) $p = 2$ Pa ($d_{as} \sim 8$ nm) after 450 °C annealing for 30 min. The statistic of grain size distribution (see the insets) demonstrates that the average grain size of the annealed thin films with $d_{as} \sim 4$ nm is still smaller than that of the films with $d_{as} \sim 8$ nm.

The FePt thin films with $d_{as} \sim 4$ nm present an accelerated $L1_0$ ordering process as compared with those with $d_{as} \sim 8$ nm [see Fig. 5]. The ordering parameter $S = 0.71$ and the coercivity $H_c = 6.3$ kOe for the thin films with $d_{as} \sim 4$ nm after 450 °C annealing are similar to those $S = 0.70$ and $H_c = 6.7$ kOe for the films with $d_{as} \sim 8$ nm after 600 °C annealing, indicating that the ordering temperature of the FePt thin films is significantly reduced $\Delta T \sim 150$ °C by varying nanocrystal size from $d_{as} \sim 8$ nm to 4 nm in as-deposited FePt thin films. Obviously, the accelerated ordering process results from the enhanced nucleation rate of the $L1_0$-ordered domains by increasing nanocrystal interfaces as demonstrated above.

We want to point out here that XRD stress measurements demonstrate a neglectable strain value ($\varepsilon < 10^{-4}$) in both the thin films with $d_{as} \sim 4$ and 8 nm, indicating that the observed accelerated ordering process is unrelated to the stress of the thin films.

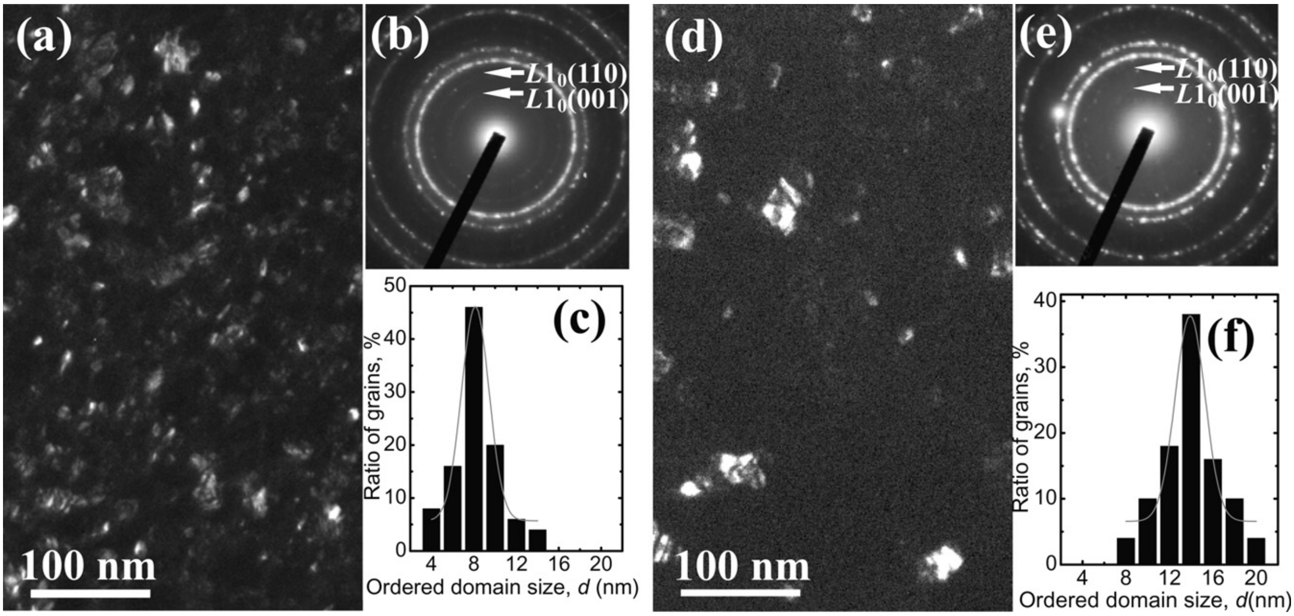

Fig. 4. Dark-field TEM images, ED patterns and the statistical size distributions of $L1_0$-ordered domains of the FePt thin films with (a–c) $d_{as} \sim 4$ nm and (d–f) $d_{as} \sim 8$ nm after 450 °C annealing for 30 min. The dark-field images are made by using the (1 1 0) reflection of the $L1_0$-FePt phase. The average size of the $L1_0$-ordered domains in the films with $d_{as} \sim 4$ nm is obviously smaller than that in those with $d_{as} \sim 8$ nm, indicating that the increase of nanocrystal interfaces enhances the nucleation rate of the $L1_0$-ordered domains and simultaneously inhibits their growth.

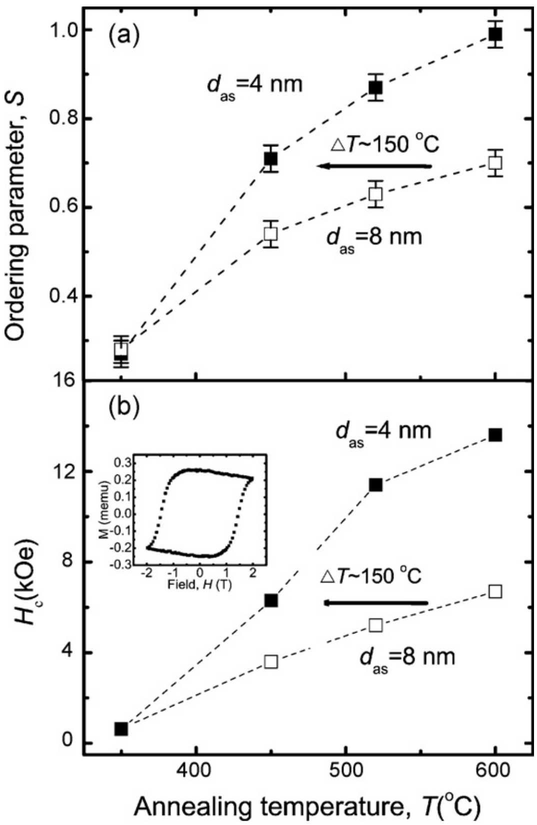

Fig. 5. Annealing temperature dependence of (a) ordering parameter S and (b) coercivity H_c of the FePt thin films with $d_{as} \sim 4$ and 8 nm. A typical magnetic hysteresis loop for the FePt film with $d_{as} \sim 4$ nm annealed at 600 °C for 30 min is given as inset of (b). The ordering process of the FePt thin films with $d_{as} \sim 4$ nm is accelerated as compared with that of the films with $d_{as} \sim 8$ nm, and a decrease of annealing temperature $\Delta T \sim 150$ °C is obtained for yielding a similar ordering parameter S and coercivity H_c.

4. Conclusions

In conclusion, the present studies demonstrate that the increase of nanocrystal interfaces in as-deposited FePt thin films enhances the nucleation rate of the $L1_0$-ordered domains and simultaneously inhibits their growth during the ordering phase transition, which results in an accelerated ordering transition with a reduced size for both the ordered domains and the FePt grains in the films. The present study provides an alternative approach to prepare the $L1_0$-FePt thin films with controlled microstructures and thus is of wide interest. The insights into the effects of nanocrystal interfaces on the ordering phase transition can be further applied in the microstructure manipulations of other nanostructured systems that require structural phase transitions.

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