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Journal of Alloys and Compounds

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Roles of nanocrystal interfaces on L1₀ ordering phase transitions in FePt thin films

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ARTICLE INFO

Article history: Received 2 December 2009 Received in revised form 20 August 2010 Accepted 26 August 2010

Keywords: FePt thin film Ordering Interface Coercivity

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.

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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 $\sim 20~\mu\text{m}^2$, respectively. The interface fraction f_{in} of FePt nanocrystals in as-deposited thin films was calculated according to $f_{\text{in}} = 3\Delta/d$ [16], where Δ is the interface thickness and d is

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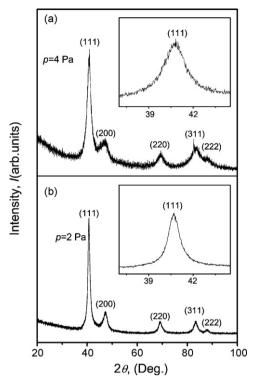


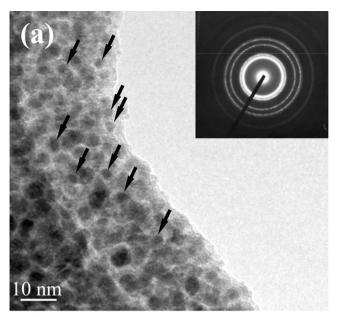
Fig. 1. XRD spectra of FePt thin films deposited at an argon pressure of (a) 4 Pa and (b) 2 Pa. The wider diffraction peaks of the films deposited at 4 Pa indicate a smaller size for FePt crystals in them as compared with those deposited at 2 Pa.

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 $4\,\mathrm{Pa}$ have a disordered face-centered cubic (A1) structure [see Fig. 1], and further analyses demonstrate that the films deposited at $4\,\mathrm{Pa}$ have a smaller grain size $(d_{as}\sim 4\,\mathrm{nm})$ than those $(d_{as}\sim 8\,\mathrm{nm})$ deposited at $2\,\mathrm{Pa}$, 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 $4\,\mathrm{Pa}$ is determined to be $\sim 75\%$, higher than that $\sim 37\%$ in the films deposited at $2\,\mathrm{Pa}$.

After $450\,^{\circ}\text{C}$ annealing for $30\,\text{min}$, the FePt thin films with $d_{as} \sim 4\,\text{nm}$ have a smaller average grain size of $D\sim 10\,\text{nm}$ [Fig. 3(a)] as compared with that $D\sim 17\,\text{nm}$ for the thin films with $d_{as} \sim 8\,\text{nm}$ [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 $4\,\text{Pa}$ 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 $d_{as} \sim 4\,\text{nm}$ have a smaller size



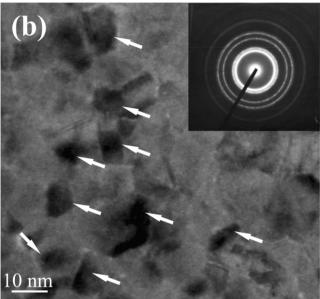


Fig. 2. Bright-field TEM images and electron diffraction (ED) patterns (the insets) of FePt thin films deposited at an argon pressure of (a) p = 4 Pa and (b) p = 2 Pa.

 $d\!\sim\!8\,\mathrm{nm}$ and a higher number density as compared with those in the films with $d_{\mathrm{as}}\!\sim\!8\,\mathrm{nm}$ [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 ($d\!\sim\!32\,\mathrm{nm}$) in the thin films with $d_{\mathrm{as}}\!\sim\!4\,\mathrm{nm}$ is still smaller than that $d\!\sim\!50\,\mathrm{nm}$ in those with $d_{\mathrm{as}}\!\sim\!8\,\mathrm{nm}$ (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.

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