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Magnetic and Ordering Behavior of Nb-doped FePt Alloy Films

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The magnetic properties and ordering behavior of Nb-doped FePt alloy films prepared by dc-magnetron sputtering were investigated. It was found that Nb addition retarded the ordering reaction from the disordered face-centered-cubic (fcc) *A*1 phase to the ordered face-centered-tetragonal (fct) $L1_0$ phase. The tetragonality (*c*/*a* ratio) of the ordered fct $L1_0$ phase increased with the Nb concentration. Nb addition hampered *c*-axis contraction during ordering, probably because the larger Nb atoms occupy Pt sites. Consequently, the coercivity and magnetocrystalline anisotropic energy of Nb-doped FePt alloy films are lower than those of un-doped FePt film under equivalent annealing conditions.

Key words : FePt alloy film, Nb-doping, magnetic properties, ordering transformation, microstructure

1. Introduction

$L1_0$ phase materials, such as CoPt and FePt alloy films, are potential high-density magnetic recording media because of their high magnetocrystalline anisotropy (>10^7 ergs/cm^3) [1, 2]. High-temperature post-deposition annealing, at temperatures over 500 °C, is usually required to obtain the ordered fct $L1_0$ phase from the magnetically soft disordered fcc phase with sputtering. This high-temperature post-deposition annealing induces grain growth in films, which is detrimental to high-density recording. Recently, the effects of various metallic additives in FePt or CoPt alloy films on aspects of the ordering reactions and microstructural development have been investigated extensively [3-11]. W, Ti [3], and Cr [4] doping significantly restrained the grain growth of the ordered FePt phase, but had no significant effect on accelerating the ordering kinetics or lowering the ordering temperature. Conversely, there have been few attempts to reduce the ordering temperature. Doping with Sn, Pb, Sb [5], and Cu [6-9] reduced the $L1_0$ ordering temperature of CoPt and FePt thin films, but had no effect on inhibiting grain growth. It has been reported that the mechanism for lowering the ordering temperature with Cu addition is alloying with the FePt matrix phase [7, 9], while other analyses have suggested diffusion or a defects mechanism [5, 6, 8]. Recently, we reported that Zr-doped $Fe_{59}Pt_{41}$ films both accelerated the ordering kinetics and restrained grain growth [10]. Zr, which is one of the refractory elements, was chosen because it has large electronegativity and atomic size differences as compared with Fe or Pt. Consequently, it is immiscible with both Fe and Pt, but is prone to form stable compounds. One might expect similar effects with Nb addition. However, it was reported that Nb inhibited the ordering reaction of disordered FePt, although the reason for this was unclear [11].

This study investigated the effects of Nb doping on the ordering behavior and magnetic properties of FePt thin films. Particularly, we analyzed the origin of ordering reaction retardation with Nb-doping in FePt alloy films.

2. Experimental Procedure

FePt and Nb-doped FePt films were prepared by dc-magnetron sputtering using an Fe target composed of Pt and Nb chips. The base pressure of the chamber was less than 7×10^{-7} Torr and the Ar deposition pressure was kept at 2 mTorr with a power of 150 W. Alloy films were deposited on 5-mm-diameter glass (Corning 7059) substrates. The typical film thickness was 70 nm. A series of post-deposition annealing operations was performed *ex-situ* under a vacuum of 1×10^{-5} Torr. Magnetic properties were measured using a vibrating sample magnetometer (VSM). The structure was analyzed using x-ray diffraction (XRD) and transmission electron microscopy (TEM)

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and the composition was analyzed using inductively coupled plasma atomic emission spectrometry (ICP-AES).

3. Results and Discussion

Fig. 1 shows the coercivity (H_c) variation of $Fe_{49}Pt_{51}$ and Nb-doped $Fe_{49}Pt_{51}$ alloy films as a function of the annealing time. All the as-deposited films were magnetically soft, with $H_c < 100$ Oe. On annealing and subsequent transformation to the ordered fct structure, the samples become magnetically hard. The H_c increased very slowly with Nb concentration. The coercivity of the undoped-$Fe_{49}Pt_{51}$film was much higher than that of Nb-doped films in the annealed state. As shown in Fig. 2(a), superlattice peaks in the un-doped film, such as (001), (110), and (201), indicating the formation of an ordered $L1_0$ phase, developed more rapidly than in the 2 at.% Nb-doped film [Fig. 2(b)] as annealing time increased. As the Nb content increased, the superlattice peaks of the ordered $L1_0$ phase were less pronounced, as shown in Fig. 2(c). Consequently, the addition of Nb was found to restrain the ordering of FePt. The H_c of Nb-doped films was obviously lower for the same annealing time, as shown in Fig. 1, and was possibly due to the lower degree of ordering. In the Zr-doped FePt system, a small amount of Zr doping accelerated the $L1_0$ ordering reaction [10]. However, the change in H_c decelerated as the Nb content increased from x = 0 to 2.0 at.%. This is the reverse of the case for the Zr-doped FePt system.

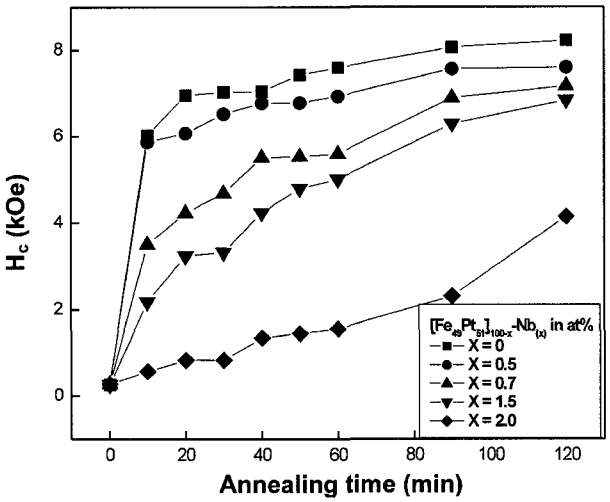

Fig. 1. Coercivity H_c changes of $[FePt]_{100-x}Nb_x$ alloy films as function of Nb content and annealing time. Samples were annealed at 500 °C.

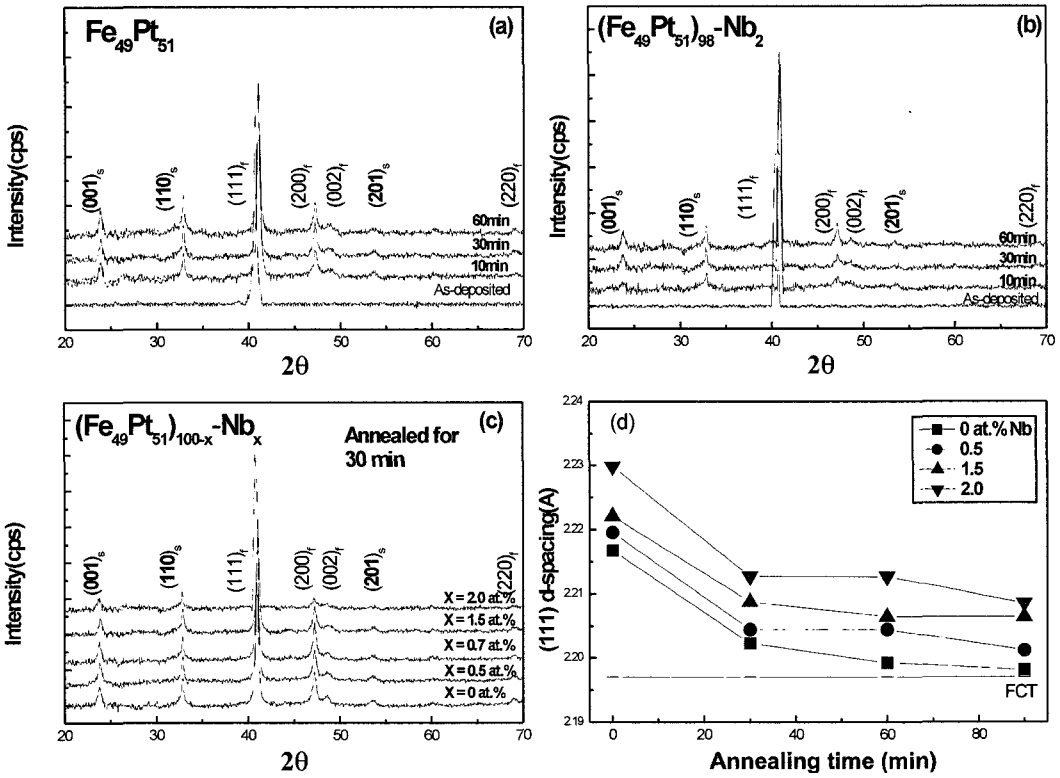

Fig. 2. XRD patterns of (a) $Fe_{49}Pt_{51}$ alloy film as a function of annealing time, (b) $[Fe_{49}Pt_{51}]_{98}Nb_{2.0}$ alloy films annealed at 500 °C as a function of annealing time, (c) $[Fe_{49}Pt_{51}]_{100-x}Nb_x$ alloy films annealed for 30 min as a function of Nb content, (d) (111) d-spacing variations of $[Fe_{49}Pt_{51}]_{100-x}Nb_x$ alloy films as a function of Nb content and annealing time.

The (111) d-spacings of the as-deposited Nb-doped alloy films were much greater than in the un-doped FePt alloy film, as shown in Fig. 2(d). The lattice expansion indicates that a large part of the Nb additive dissolves and forms a solid solution with the FePt matrix phase. The (111) d-spacings of the Nb-doped and un-doped films gradually decreased as the annealing time increased because the *c*-axis of the fct ordered phase decreased. The (111) d-spacing of the un-doped alloy film closely approached the value of the fct ordered phase with 90 min annealing. Conversely, as the Nb concentration increased, the deviation of the (111) d-spacings of Nb-doped alloy films from the value for the fct ordered phase increased. This clearly indicated that Nb doping retards the ordering reaction. Why does Nb doping retard the ordering reaction in contrast to Zr doping? The electronegativity and atomic size difference between Zr and Pt or Fe are larger than those between Nb and Pt or Fe. Consequently, the amount of dissolution of Zr in the FePt matrix is more limited than that of Nb in the as-deposited state. In addition, Zr has a greater tendency to form compounds with Pt and Fe [12, 13].

Fig.3 shows the variation in the lattice parameters and

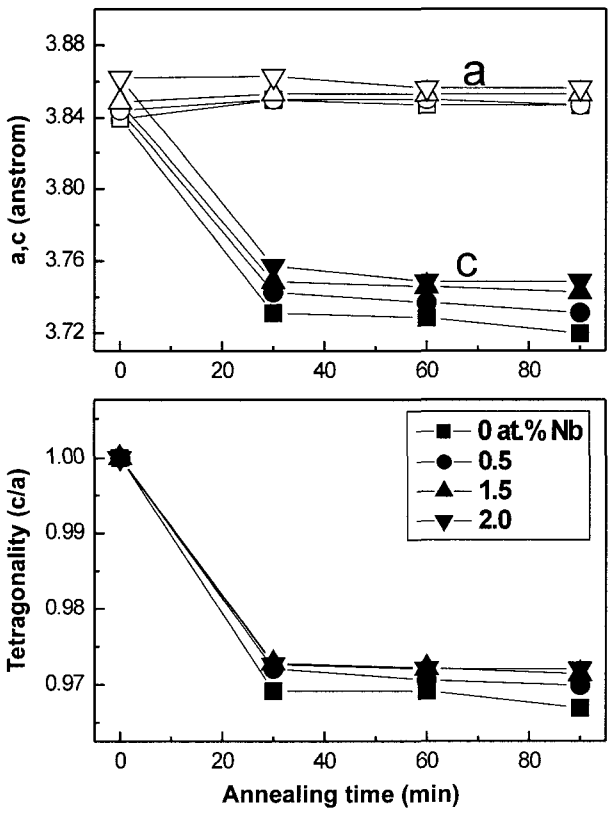

Fig. 3. a) Lattice parameters, *a* and *c*, b) tetragonality (*c/a* ratio) variations of $[FePt]_{100-x}Nb_x$ alloy films as functions of Nb content and annealing time.

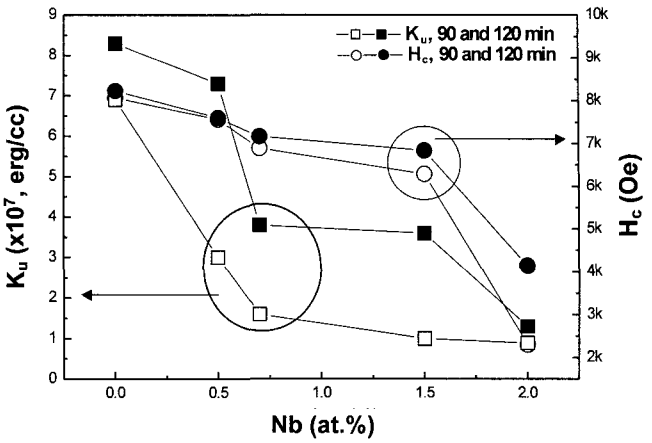

Fig. 4. Magnetocrystalline anisotropy (K_u) changes of $[FePt]_{100-x}Nb_x$ alloy films with various Nb contents and annealing time. Open and solid symbol represent 90min and 120min annealed state, respectively.

tetragonality (*c/a* ratio) for $[Fe_{49}Pt_{51}]_{100-x}Nb_x$ films as a function of Nb content and annealing time. The $L1_0$ ordering reaction accompanies the crystal structure change from fcc to fct. The bulk value of the tetragonality (*c/a*) of equiatomic FePt alloy is 0.963 [14]. As the Nb concentration increased, the *c*-lattice parameter increased, *i.e.*, the tetragonality (*c/a*) increased as compared with the bulk value, as shown in Fig. 3b. This suggests that Nb is incorporated by substitution along the *c* axis of $L1_0$ FePt. If larger Nb atoms replace Pt at Pt sites, such as 0,1/2,1/2, 1/2,1/2,1/2, 1,1/2,1/2, or 1/2,0,1/2 sites, the reduction in the *c*-axis during ordering will be hampered. Consequently, the increased Nb content would retard the transformation from disordered fcc-FePt to ordered fct-FePt phase. Another possible reason for the decrease in H_c with increasing Nb concentration is the decreasing anisotropic energy resulting from the tetragonality change of the $L1_0$ phase.

Fig. 5(a) shows the variation in the saturation magnetization (M_s) for $[Fe_{49}Pt_{51}]_{100-x}Nb_x$ films as a function of annealing time. The M_s in the as-deposited state decreased as the Nb concentration increased, probably because of dilution of the magnetic moment. The M_s of both Nb-doped and un-doped FePt films gradually decreased as annealing time increased. When un-doped samples were annealed at 500 °C for 120 min, the M_s decreased about 33.3% (1184 → 789 emu/cc). The structural transformation of a disordered fcc A_1 phase to an fct $L1_0$ phase also accompanies a change in magnetic ordering from ferromagnet to ferrimagnet. The net magnetic moment of the ferrimagnetically ordered $L1_0$ fct phase is known to be smaller, by up to about 30% depending on FePt composition, than that of the

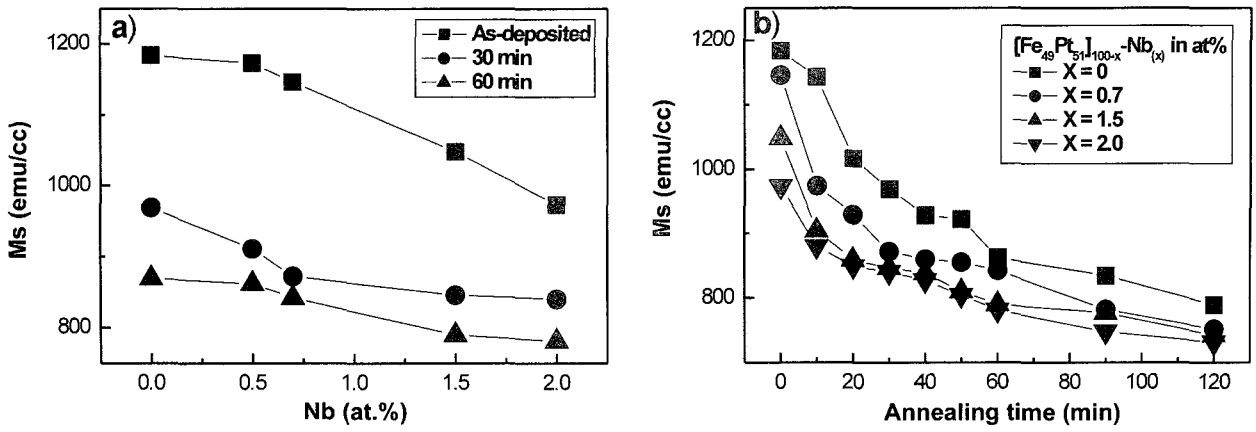

Fig. 5. M_s changes of $[FePt]_{100-x}Nb_x$ alloy films as function of Nb contents and annealing time. Samples were annealed at 500 °C.

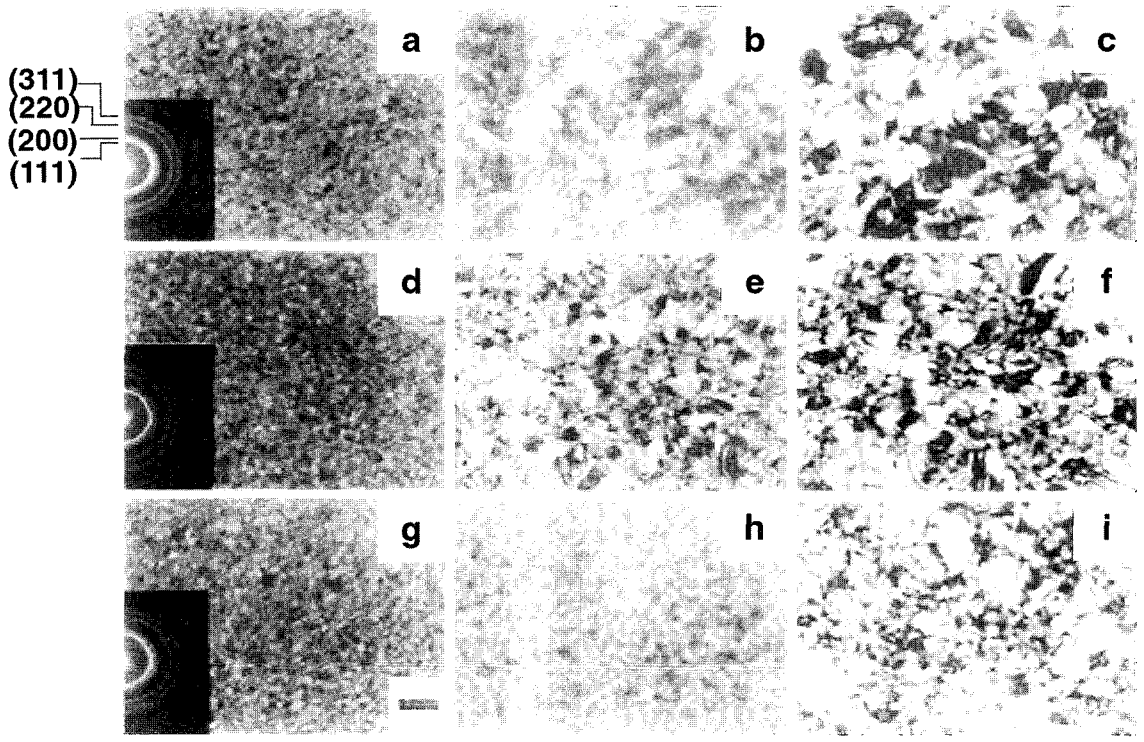

Fig. 6. TEM micrographs of $Fe_{59}Pt_{41}$ (a)-(c), $[FePt]_{99.3}Nb_{0.7}$ (d)-(f), and $[FePt]_{98}Nb_{2.0}$ (g)-(i) alloy films as a function of annealing time. (a)-(d)-(g): as-deposited, (b)-(e)-(h): annealed for 10 min, and (c)-(f)-(i): annealed for 60 min.

ferromagnetically disordered fcc phase [14]. The M_s difference between the as-deposited and annealed state decreased as the Nb concentration increased. This also indicates that the ordering reaction was restrained as the Nb concentration increased.

Bright-field TEM images of both as-deposited and annealed FePt and FePt-Nb alloy films are shown in Fig. 6. The grain growth of Nb-doped films was relatively slow as compared to the un-doped FePt film during post-deposition annealing. In the as-deposited state, the average grain size was less than 5 nm. When the films were annealed at 500 °C for 60 min, the average grain size of the un-doped and 2 at.% Nb doped films was about 35 and 25 nm, respectively. Compared with the previous result on the effect of Zr addition on grain growth retardation, Nb addition had a small effect on grain growth retardation. Zr has a greater effect on grain growth retardation because it has more limited solubility in the FePt matrix and is effectively segregated in the grain boundaries.

4. Conclusion

We studied the effect of Nb doping on the ordering reaction and magnetic properties of FePt films. Nb addition retarded the phase transformation from disordered fcc phase to ordered fct phase reaction. Based on the tetragonality (*c/a*) changes of the ordered fct $L1_0$ phase with Nb concentration, it is thought that the Nb atoms hampered *c*-axis contraction of the $L1_0$ face-centered-tetragonal phase, probably because larger Nb atoms filled Pt sites. The primary reason for the decrease in H_c with increasing Nb concentration is the decreasing K_u value resulting from incomplete transition from the fcc FePt phase to the fct FePt phase.

Acknowledgments

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