Microstructure and magnetic properties of nanocomposite FePtCr–SiN thin films

P. C. Kuo and S. C. Chen
Institute of Materials Science and Engineering, National Taiwan University, Taipei 106, Taiwan

Y. D. Yao
Institute of Physics, Academia Sinica, Taipei 115, Taiwan

A. C. Sun and C. C. Chiang
Institute of Materials Science and Engineering, National Taiwan University, Taipei 106, Taiwan

$[(FePt)_{100-x}Cr_x]_{100-\delta}-[SiN]_\delta$ nanocomposite thin films with $x=0-25$ at. %, and $\delta=0-30$ vol. % were fabricated on a natural-oxidized Si(100) substrate by dc and rf magnetron cosputtering of FePt, Cr, and Si_3N_4 targets. The thickness of the films was kept at 10 nm in order to examine the possibility for applying in high-density magnetic recording media. Transmission electron microscopy (TEM) and electron diffraction analyses indicated that the face-centered-cubic (fcc) γ-phase FePt, body-centered-cubic (bcc) Cr, and amorphous SiN coexisted in as-deposited films. The as-deposited films were annealed in vacuum between 350 and 750 °C for 30 min, and then ice-water quench cooling, in order to transform the soft magnetic fcc γ-FePt phase to the hard magnetic face-centered-tetragonal (fct) γ_1 phase. Cr was added to inhibit the FePt grain growth, and was observed by TEM and energy disperse spectrum analysis in the grain surface area of FePt grains. The TEM observation indicated that the structure of the film was an amorphous SiN matrix with FePtCr particles dispersed in it. The particle size of FePtCr in annealed film was increased with the annealing temperature but decreased with the increase of SiN and Cr contents. Magnetization measurements indicated that the optimum condition for high-density magnetic recording purpose of the film was found with $x=10$ at. % and $\delta=15$ vol. %, annealing at 600 °C for 30 min. The average grain size of the FePtCr in this film is about 9.5 nm, the saturation magnetization is 450 emu/cm^3, in-plane coercivity is 3.7 kOe, and in-plane squareness is about 0.75. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1453352]

I. INTRODUCTION

FePt related magnetic thin films with their high coercivity H_c, relative good remnant magnetization M_r, high magnetocrystalline anisotropy K_u, small grain size, good corrosion resistance, and large energy products $(BH)_{max}$ are attractive media for extremely high-density magnetic recording applications.[1–4] To these metallic films, the most significant problem for magnetic recording media application is the noise that results from magnetic exchange coupling between the grains.[5] The key issue to reducing its noise is the reduction of the intergrain magnetostatic and exchange interactions. Therefore, composite granular films with isolated magnetic grains dispersed in a nonmagnetic matrix are expected to become more suitable for extremely high-density recording media.

Previous investigations have shown that the magnetic properties of the FePt films are sensitive to process parameters,[6] the media noise can be improved by dispersing magnetic FePt grains into a nonmagnetic SiN matrix,[7] and the grain size can be reduced by the addition of Cr into FePt film.[8] In this work, the effects of Cr content, SiN volume fraction, and annealing temperature on the magnetic properties and particle size of the nanocomposite FePtCr-SiN thin films are reported.

II. EXPERIMENT

$[(FePt)_{100-x}Cr_x]_{100-\delta}-[SiN]_\delta$ nanocomposite thin films with $x=0-25$ at. %, and $\delta=0-30$ vol. % were fabricated on a natural-oxidized Si(100) substrate by dc and rf magnetron cosputtering of FePt, Cr, and Si_3N_4 targets. The substrate is rotated at 75 rpm in order to attain uniform composition of the film with a thickness of 10 nm. The magnetic films were covered by a thin Si_3N_4 layer to protect its oxidation.

The chamber base pressure was approximately 3×10^{-7} Torr and films were deposited under an argon pressure of 7 mTorr. The deposition rate was about 0.3 nm/s. The as-deposited film was sealed in quartz capsules and then postannealed in vacuum at various temperatures for 30 min, and the film was quenched in ice water after annealing.

The film microstructure was observed by transmission electron microscopy (TEM) and the average grain size of the film was measured by the TEM bright field image. Magnetic properties at room temperature were measured by a vibrating sample magnetometer and a superconducting quantum interference device, with maximum applied fields of 13 and 50 kOe, respectively. Composition and homogeneity of the films were determined by energy disperse spectrum (EDS). The film thickness was measured by an atomic force microscope.

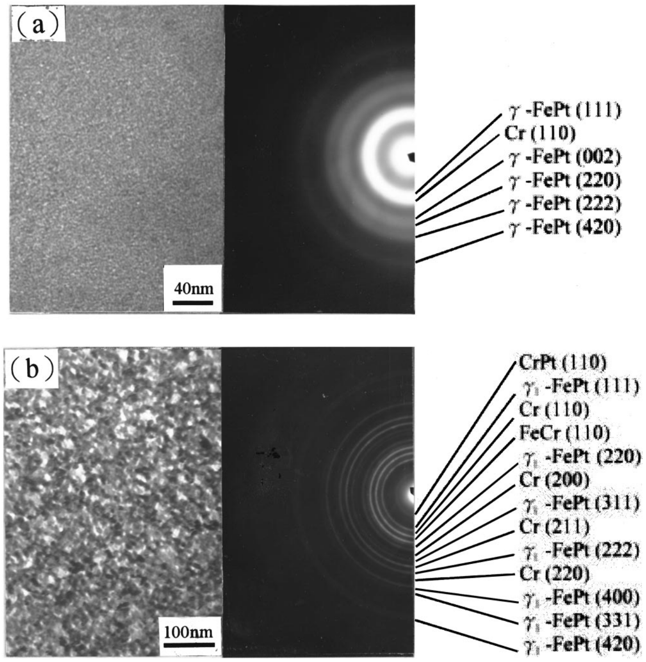

FIG. 1. TEM bright field images and the electron diffraction pattern of (a) the as-deposited $[(FePt)_{90}Cr_{10}]_{85}–[SiN]_{15}$ film and (b) the film after annealing at 600 °C for 30 min.

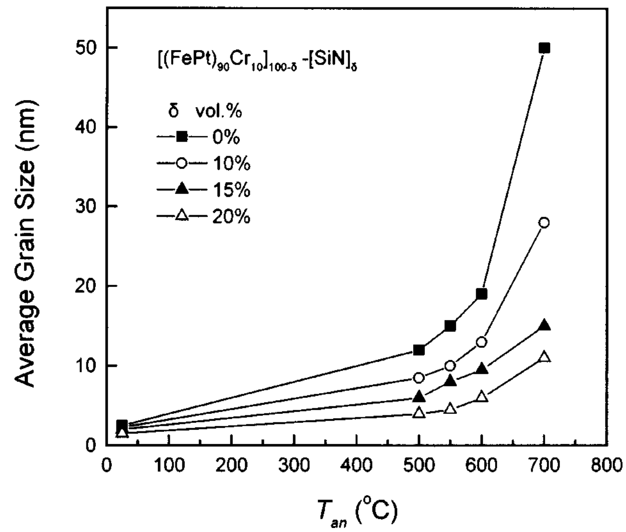

FIG. 2. Variation of the average grain size with annealing temperature of the various $[(FePt)_{90}Cr_{10}]_{100-\delta}–[SiN]_{\delta}$ films; SiN contents of the films are 0, 10, 15, and 20 vol.%, respectively.

III. RESULTS AND DISCUSSION

Figure 1 shows the TEM bright field images and the electron diffraction pattern of (a) the as-deposited $[(FePt)_{90}Cr_{10}]_{85}–[SiN]_{15}$ film and (b) the film after annealing at 600 °C for 30 min. The FePt grains in deeper colors are dispersed in the SiN matrix in lighter or approaching white colors. The size of the FePt grains is about 2 nm in (a) and 9.5 nm in (b). The electron diffraction pattern on Fig. 1(a) shows that the crystal structure of fcc γ FePt and bcc Cr phases have developed in the as-deposited film, and they transform to γ_1-FePt, FeCr, CrPt, and Cr phases after annealing, as shown in Fig. 1(b). The SiN diffraction rings are not observed in both cases, this means that the SiN is in an amorphous state.

From the TEM-EDS analyses of the different areas of the enlarged grain of the film, which annealed at higher temperature and longer times, we observed that Cr existed mainly in the FePt grain surface area and grain boundary.[9] As shown in Ref. 8, the grain size of FePt in FePt metal film will grow up after annealing, and it can be reduced by the addition of Cr. In the FePt–Cr-SiN system, the grain size decreases with increasing Cr content and increases with increasing annealing temperature. Figure 2 shows the average grain size as a function of annealing temperature T_{an} for various $[(FePt)_{90}Cr_{10}]_{100-\delta}–[SiN]_{\delta}$ films with δ up to 20 vol. %. It shows that the SiN can also restrain the growth of FePt grains.

Figure 3 shows the variations of in-plane coercivity $H_{c\parallel}$ with annealing temperature of the $[(FePt)_{100-x}Cr_x]_{85}–[SiN]_{15}$ thin films with different Cr contents. When $T_{an} > 400$ °C, the $H_{c\parallel}$ value of the $(FePt)_{85}$-$[SiN]_{15}$ thin film (Cr = 0 at. %) will step up rapidly as T_{an} is increased. $H_{c\parallel}$ value reaches its maximum value of about 10 kOe at $T_{an} \sim 700$ °C, and then steps down quickly as T_{an} keeps going up. When $T_{an} < 700$ °C, $H_{c\parallel}$ increases as T_{an} is increased because the soft magnetic γ-FePt phase transforms to the hard magnetic γ_1-FePt phase, which has an extremely high magnetocrystalline anisotropy constant. When $T_{an} > 700$ °C, the growth of γ_1-FePt grains and the reaction of FePt with Si substrate will cause $H_{c\parallel}$ to decrease as T_{an} is increased.[7] At $T_{an} = 600$ °C, the $H_{c\parallel}$ value is 8 kOe for the $(FePt)_{85}–[SiN]_{15}$ film (Cr = 0 at. %) but it will drop to 3.7 kOe as Cr content increases to 10 at. %, because increase of Cr content inhibits the growth of FePt grains during annealing and makes the grain size deviate considerably from the single domain size [which is about 90 nm (Ref. 6)] and some grains become superparamagnetic particles. Moreover, the diffusion of Cr into the FePt grain surface area and lowering the degree of the ordering γ_1-FePt phase will also decrease the crystal anisotropy constant of FePt. As shown in

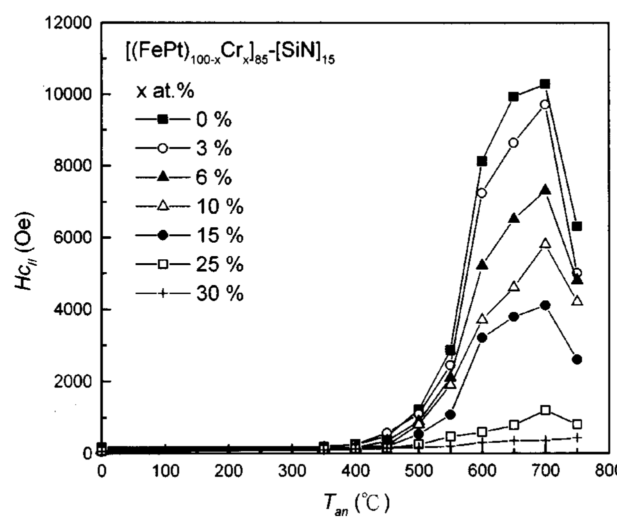

FIG. 3. Variations of $H_{c\parallel}$ with annealing temperature of various $[(FePt)_{100-x}Cr_x]_{85}–[SiN]_{15}$ films; Cr contents of the films are 0, 3, 6, 10, 15, 25, and 30 at. %, respectively.

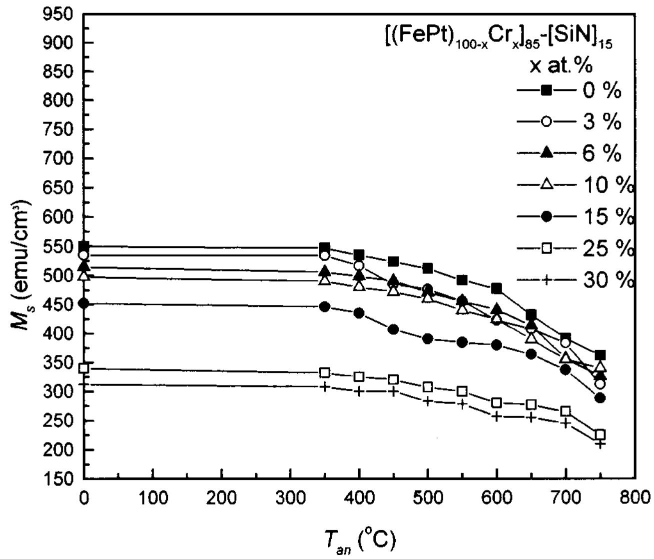

FIG. 4. Variations of M_s with annealing temperature of various $[(FePt)_{100-x}Cr_x]_{85}-[SiN]_{15}$ films; Cr contents of the films are 0, 3, 6, 10, 15, 25, and 30 at. %, respectively.

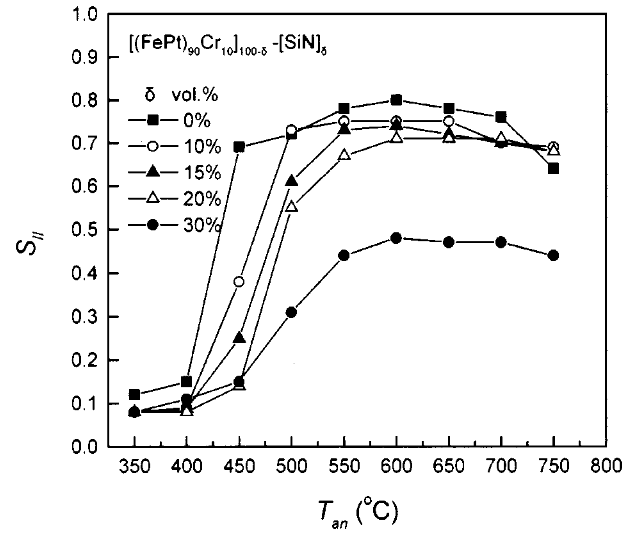

FIG. 5. Variations of $S_{\parallel}$ with annealing temperature of various $[(FePt)_{90}Cr_{10}]_{100-\delta}-[SiN]_{\delta}$ films; SiN contents of the films are 0, 10, 15, 20, and 30 vol. %, respectively.

Fig. 1(a), we can see that only the γ-FePt and Cr phases were found in the as-deposited film, but they transform to γ_1-FePt, FeCr, CrPt, and Cr phases after annealing at 600 °C for 30 min, as shown in Fig. 1(b). This indicates that some Cr are diffused into FePt grains.

Figure 4 shows the relations between saturation magnetization M_s and T_{an} of the $[(FePt)_{100-x}Cr_x]_{85}-[SiN]_{15}$ films with different Cr contents. We find the M_s value of $(FePt)_{85}$-$[SiN]_{15}$ film (Cr=0 at. %) decreases as T_{an} is increased because of the reaction of the magnetic layer with the Si substrate at higher T_{an}.[7] On the other hand, Cr is an antiferromagnetic substance, and the increase of Cr will dilute the M_s values of the film. The M_s value of the $(FePt)_{85}-[SiN]_{15}$ film (Cr=0 at. %) which annealed at 600 °C, is about 490 emu/cm^3, but it will decrease to 450 emu/cm^3 as Cr content increases to 10 at. %.

Figure 5 shows the relationships between in-plane squareness $S_{\parallel}$ and T_{an} of the $[(FePt)_{90}Cr_{10}]_{100-\delta}-[SiN]_{\delta}$ thin films with different SiN contents; they have the same tendency as that of $H_{c\parallel}$ vs T_{an}. The maximum $S_{\parallel}$ occurs at $T_{an}\sim 600$ °C. The $S_{\parallel}$ at $T_{an}=600$ °C are about 0.8 and 0.75 for the $(FePt)_{90}Cr_{10}$ film (SiN=0 vol. %) and $[(FePt)_{90}Cr_{10}]_{85}-[SiN]_{15}$ film, respectively, but it decreases as SiN content is increased. It will drop to about 0.5 as SiN content increases to 30 vol. %. This indicates that the magnetic easy direction of FePtCr particles is changed from the parallel film plane to random and the interparticle interactions are reduced as SiN content is increased. The $S_{\parallel}$ value for randomly oriented noninteracting Stoner-Wohlfarth particles is 0.5.[10] This suggests that the magnetic FePtCr particles in this film are almost randomly oriented and isolated by the SiN.

IV. CONCLUSION

FePtCr–SiN granular films consisting of the ordered fct FePtCr particles embedded in an amorphous SiN matrix have been successfully prepared. The particle size of FePtCr in annealed film was increased with annealing temperature but decreased with increasing SiN and Cr contents. A granular $[(FePt)_{90}Cr_{10}]_{85}-[SiN]_{15}$ film with an $H_{c\parallel}$ value of 3.7 kOe and the magnetic particle size about 9.5 nm was obtained after annealing at 600 °C for 30 min. Its in-plane squareness is about 0.75. This FePtCr–SiN nanocomposite film can be a promising candidate for extremely high-density recording media.

ACKNOWLEDGMENT

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