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Nanostructure and magnetic properties of composite CoPt:C films for extremely high-density recording

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The nanostructure and magnetic properties of composite CoPt:C films at room temperature were investigated as a function of annealing temperature, carbon concentration, and film thickness. CoPt:C films with a variety of carbon concentrations were fabricated by cosputtering Co, Pt, and C onto water-cooled Si(100) substrates followed by annealing. X-ray diffraction and transmission electron microscopy analyses indicate that nanocrystallites of face-centered-tetragonal (fct) CoPt phase, which has a uniaxial magnetocrystalline anisotropy constant of about 5×10^7 erg/cm³, can be formed in carbon matrix when the annealing temperature is higher than 600 °C. The grain sizes of the fct CoPt crystallites are about 10 nm and the coercivities can be as high as 12 kOe. Higher annealing temperature and lower carbon concentration generally lead to larger grain sizes and perhaps more complete formation of the fct CoPt phase, and therefore higher coercivities. The coercivity is insensitive to the film thickness until the thickness is smaller than the CoPt grain size, when the coercivity starts to decrease with film thickness. The magnetic activation volumes are typically around 1×10^{-18} cm³. The nanostructure and the associated magnetic properties of these composite CoPt:C films are promising as potential media for extremely high-density recording. © 2000 American Institute of Physics. [S0021-8979(00)46808-8]

I. INTRODUCTION

Over the past few years the areal density (D_A) of longitudinal magnetic recording media has been increasing at an annual rate of about 60%.^{1–3} 5 and 20 Gb/in.² have been achieved in commercial disk products and lab demonstrations, respectively. Extremely high density recording (EHDR) with D_A of about 100 Gb/in.² is expected in a few years. In order to have adequate signal-to-noise ratio, media with higher D_A require magnetic grains with smaller sizes and higher coercivities. For EHDR media it is estimated that coercivity (H_c) of about 4 kOe⁴ and grain size (d) of about 10 nm is needed.⁵ For such a small grain size the issue of thermal stability must be considered. In the Stoner– Wohlfarth model with particle volume V and uniaxial anisotropy constant K_u , it is usually required at temperature T to have

$$K_u V/k_B T \ge 60 \tag{1}$$

in order for the media to be thermally stable.⁶ The current media materials are Co-based alloys, whose K_u are in the order of 2×10^6 erg/cm³. For $d \approx 10$ nm, Eq. (1) cannot be satisfied for Co-based alloys. The face-centered-tetragonal (fct) CoPt phase has a K_u value of about 5×10^7 erg/cm³ and Eq. (1) can be well satisfied if it is used as the media material. With the high K_u value, it is also easy to develop H_c . High H_c has been obtained in various materials based on the fct CoPt phase.⁷⁻¹⁰

Real magnetic films are usually very different from collections of Stoner–Wohlfarth grains due to intergrain exchange interactions, grain size distributions, and incoherent rotations in magnetization reversal. For such films the magnetic activation volume V^* can be regarded qualitatively as the effective or average unit volume of moments switching together in magnetization reversal.^{11,12} It is V^* instead of V that directly affects the media noise and thermal stability.^{5,13} So V^* is also an important parameter for recording media.

Having small CoPt grains with weak intergrain exchange interactions is essential to reduce media noise. To achieve this goal, a nonmagnetic intergrain material is preferred to isolate the CoPt grains. C is expected to be an ideal choice because there is neither Pt carbide nor stable Co carbide that may affect the formation of fct CoPt phase.

II. EXPERIMENT

All CoPt:C composite films were prepared by cosputtering Co, Pt, and C onto water-cooled Si(100) substrates followed by annealing. The base pressure is better than 3 $\times 10^{-7}$ Torr and the Ar pressure during sputtering is 10 mTorr. The films have equiatomic Co-Pt ratio with various C concentrations. The CoPt:C film thickness δ ranges from 3 to 100 and 20 nm C underlayer and overcoat are used for protection. Vacuum annealing was carried out for one hour at various temperatures. X-ray diffraction (XRD) and transmission electron microscopy (TEM) were used to analyze the structural properties. The average grain sizes d of 100 nm films were estimated from the (111) peak width of the XRD data using Scherrer's formula.¹⁴ An alternating gradient force magnetometer and a superconducting quantum interference device (SQUID) magnetometer were used to characterize the magnetic properties. All magnetic measurements were made in the film plane. The activation volumes V^* were obtained from the magnetic viscosity and irreversible susceptibility data.^{12,15} The activation grain size d^* is defined as $d^* = (V^*)^{1/3}$ in 100 nm films.

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FIG. 1. XRD patterns of 100 nm CoPt:C films with 30 vol % C annealed at various temperatures T_A . The standard XRD patterns for fct and fcc CoPt phases are also plotted as references.

III. RESULTS AND DISCUSSION

In addition to the fct CoPt phase, which is the low temperature phase, there is also a high temperature facecentered-cubic (fcc) CoPt phase around the equiatomic Co–Pt system. The as-deposited CoPt:C films most likely consist of fcc CoPt grains^{7–10} and/or amorphous CoPt plus C matrix. To form the fct CoPt phase, the as-deposited films must be annealed. Because the lattice parameters of the two CoPt phases are very close, the XRD patterns are also very similar except that there are several additional peaks for the fct phase due to reduced lattice symmetry. The standard XRD patterns of 100 nm CoPt:C films with 30 vol % C annealed at various temperatures (T_A) are shown in Fig. 1.



FIG. 2. Annealing temperature T_A dependence of grain size d, activation grain size d^* , coercivity H_c , and saturation magnetization M_s of 100 nm CoPt:C films with 30 vol % C.



FIG. 3. Bright-field TEM picture of a 100 nm CoPt:C film with 30 vol % C annealed at 650 °C.

Excluding the Si(400) peak at $2\theta = 69.2^{\circ}$, all the XRD peaks can be attributed to the fct and/or fcc CoPt phases. This means that in annealed CoPt:C films there are very little or no Co or Pt carbides and C exists only in the form of elemental C matrix. When $T_A < 550^{\circ}$ C, there is no indication of the formation of fct CoPt phase. When $T_A \ge 600^{\circ}$ C, the fct CoPt phase starts to form as indicated by the emergence of the (110) peak at $2\theta = 33.3^{\circ}$. At higher T_A , the conversion from fcc to fct CoPt phase is more complete and larger CoPt grains are formed.

The T_A dependence of d, d^* , H_c , and M_s are plotted in Fig. 2. The increase of d with T_A is expected. The sharp increase of H_c near $T_A = 650$ °C can only be explained by the formation of the fct CoPt phase. The slow increase of H_c as T_A further increases can be attributed to the grain growth. Since the fct CoPt phase has slightly smaller M_s than the fcc CoPt phase,⁷ the gradual decrease of M_s with increasing T_A can also be explained by the increasing conversion from fcc CoPt phase to fct CoPt phase. d^* changes little with T_A and remains at about 10 nm. When $T_A > 650$ °C, d^* is clearly smaller than d, suggesting incoherent rotation.

The bright-field TEM picture of a 100 nm CoPt:C film with 30 vol % C annealed at 650 °C is shown in Fig. 3. It is clear that the film consists of CoPt grains surrounded by C matrix. Electron diffraction analyses confirmed that these CoPt grains have the fct structure. Some CoPt grains are well isolated by C but some are aggregated. The grain size obtained from Fig. 3 agrees with the data in Fig. 2, indicating



FIG. 4. Grain size d, activation grain size d^* , and coercivity H_c of 100 nm CoPt:C films annealed at 650 °C with various C concentrations.

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FIG. 5. Normalized maximum ΔM values $(\Delta M)_{max}$ of 100 nm CoPt:C films annealed at 650 °C with various C concentrations.

that Scherrer's formula is a reasonable method to estimate the average grain size of nanocomposite materials.

Figure 4 shows d, d^* , and H_c of 100 nm CoPt:C films annealed at 650 °C as functions of C concentration. With increasing C concentration, grain size d decreases steadily because there is more C to impede the growth and formation of fct CoPt grains. The direct result is that H_c decreases at almost the same pace as d with increasing C concentration. Again d^* remains almost unchanged, at about 9 nm. When C concentration is less than 50 vol %, d^* is clearly smaller than d, suggesting incoherent rotation.

As the C concentration increases, it is expected that the CoPt grains are better isolated and the intergrain exchange interactions are reduced. Qualitatively the intergrain interaction can be estimated by the ΔM method.¹⁶ As shown in Fig. 5, the trend is apparent that with increasing C concentration, the maximum ΔM value decreases, indicating the decrease of intergrain exchange interaction.

Figure 6 shows the film thickness δ dependence of H_c of CoPt:C films with 50 vol% C annealed at 600 and 650 °C. For the films annealed at 600 °C, H_c remains almost unchanged until $\delta < 5$ nm, when H_c starts to decrease. For those annealed at 650 °C, H_c also remains almost unchanged and starts to decrease when $\delta < 10$ nm. This result can be well explained by the fact that the grain size d of 100 nm films annealed at 600 and 650 °C are no more than 8 and 10 nm, respectively. When $\delta > d$, the nanostructure changes



FIG. 6. Film thickness δ dependence of coercivity H_c of CoPt:C films with 50 vol % C annealed at 600 and 650 °C.

little with δ and therefore H_c is hardly affected. When $\delta < d$, the grain volume decreases with δ due to reduced dimension, resulting in the decrease of H_c . This is exactly what is expected from the particulate nature of the CoPt grains in nanocomposite CoPt:C films. The activation volumes are also insensitive to δ , being about 1.1×10^{-18} and 0.7×10^{-18} cm³ for the films annealed at 600 and 650 °C, respectively.

IV. SUMMARY AND CONCLUSIONS

Nanocomposite CoPt:C films consisting of fct CoPt grains and C matrix can be synthesized with controlled coercivity and grain size. Annealing temperature of at least 600 °C is necessary to form the fct CoPt phase, which has a very high uniaxial anisotropy constant. Coercivity and grain size increase with increasing annealing temperature and decreasing C concentration. Coercivities of up to 12 kOe and grain sizes of 7 to 20 nm can be obtained, depending upon the annealing temperature and C concentration. Due to the particulate nature of the CoPt grains, film thickness appears to have little effect on the nanostructure and therefore the magnetic properties unless it is smaller than the CoPt grain size. The activation volumes are around $1 \times 10^{-18} \text{ cm}^3$ for films with fct CoPt grains. The small grain sizes and activation volumes as well as the adequate coercivity are in the right range for low media noise, and the high uniaxial anisotropy constant provides enough thermal stability. Based on these properties, nanocomposite CoPt:C films appear to be a promising media candidate for extremely high-density recording.

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