Low-temperature Ordering in FePt Films Deposited by Gas Flow Sputtering

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FePt films were deposited by a low energy sputtering method, i.e., gas flow sputtering, without heating the substrate. The effects of annealing on the $L1_0$ order parameter, morphology, and magnetic properties of the films were studied. The order parameter increased significantly after annealing at 413°C; the order parameter of a 76-nm-thick film was found to be 0.84. The coercivity of the film was 15.6 kOe, which was in agreement with the high order parameter. Scanning electron microscopy observations showed that the ordering in the films proceeded with the growth of grains. The XRD pattern of an unannealed thick (950 nm) film showed a 110 superstructure peak. The small grain size in the as-deposited film and the existence of nuclei of the $L1_0$ phase may play key roles in inducing the ordering at a relatively low temperature. These factors may promote discontinuous ordering, wherein ordering proceeds with grain boundary diffusion and growth of grains with an $L1_0$ structure.

Key words: FePt film, gas flow sputtering, annealing, $L1_0$ structure, ordering

1. Introduction

Granular thin films or thin films with a columnar structure, which consist of a material with high magnetic anisotropy, can potentially be used in the fabrication of high density magnetic recording media. One such material is FePt alloy with an $L1_0$ ordered structure. However, $L1_0$ ordering can be achieved generally at high temperatures, which may cause coalescence and growth of FePt grains. Therefore, low-temperature ordering is more suitable for practical applications of FePt films. Some techniques have been proposed for decreasing the ordering temperature of FePt alloys, such as deposition of Fe/Pt multilayers^{1,2)}, addition of a third element^{3,4)}, and the use of underlayers^{5,6)}.

We have adopted a different approach for lowering the ordering temperature, by which FePt films are deposited by low energy sputtering; this method is called gas flow sputtering (GFS). GFS is carried out at a high pressure of around 100 Pa using a hollow cathode discharge. The mean free paths of both Ar atoms and sputtered atoms are as short as 0.1 mm due to the high pressure. Therefore, energetic particles, including sputtered atoms and recoiled Ar atoms, lose their energies on collisions with ambient Ar atoms. We have successfully carried out the deposition of Co thin films having a pure hcp structure⁷⁾ and the deposition of Co-Pt thin films having a nanosized columnar structure⁸⁾ by GFS. The pure hcp structure, which is the low-temperature phase of Co, is presumably obtained as a result of low energy deposition. GFS is expected to have the same effect on the formation of the $L1_0$ structure in FePt films.

In this study, we investigate the effect of annealing on the $L1_0$ order parameter, the morphology, and

magnetic properties of GFS-deposited FePt films. Further, we discuss the mechanism of $L1_0$ ordering occurring at a relatively low temperature.

2. Experiment

FePt films were deposited on SiO₂ glass substrates by GFS, unless otherwise specified. The sputtering target was a tube made of Fe (inner diameter of 3 cm and length of 6 cm), in which Pt plates were placed. The GFS chamber was evacuated to below 2.6×10^{-5} Pa prior to sputtering, and film deposition was carried out at a pressure of 130 Pa and an Ar flow rate of 600 sccm. The sputtering power was approximately 360 W. During sputtering, the substrate temperature was maintained below 80°C. The films were annealed in a flow of 3% H₂/Ar gas for 30 min. The temperature inside the annealing furnace was corrected by placing an additional thermocouple at the sample position. Information about the composition and thickness of films is important for the estimation of their order parameter; the composition and thickness of the films were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) and scanning electron microscopy (SEM), respectively. The composition and thickness of FePt films annealed at various temperatures are summarized in Table 1. The crystal structure was characterized by X-ray diffraction (XRD) with θ - 2θ scan using Cu-K α radiation. For a quantitative analysis of the XRD peak intensity, the sample size and slit widths of the diffractometer were set such that over the entire range of diffraction angles, the X-ray irradiation area did not fall outside the sample. The magnetic properties of the film were determined using a superconducting quantum interference device magnetometer.

Table 1 Composition x (in $\text{Fe}_x\text{Pt}_{1-x}$) and thickness of FePt films annealed at various temperatures T_a .

T_a (°C)	Composition x	Film thickness (nm)
613	0.46	48
563	0.50	68
513	0.46	56
463	0.47	64
413	0.45	76
363	0.50	48
313	0.56	50
as-deposited	0.50	63

3. Results

The XRD patterns of FePt films annealed at various temperatures are shown in Fig. 1. Peaks originating from the $L1_0$ superstructure can be observed in the XRD pattern of the film annealed at 313°C; their intensities increase with increasing annealing temperature. The peak-to-peak intensity ratios calculated in the case of all samples were found to be almost equal to that of a powdered sample. Since it is well known that in a powdered sample, crystallites are oriented randomly, the above result clearly suggests that even in the thin films, the crystallites are randomly oriented. The Bragg-Williams order

Fig. 1 XRD patterns of FePt films annealed at various temperatures. The peaks labeled with asterisks indicate superstructure peaks.

parameter⁹⁾ was estimated from the intensity ratio of a superstructure peak to a fundamental peak, i.e., that of the 001 peak to the 002 peak. The integrated intensity was corrected using the Lorentz-polarization factor for randomly oriented crystallites⁹⁾, thermal factor¹⁰⁾ (where the overall isotropic atomic displacement parameter was assumed to be $0.5 \text{ \AA}^2$), and film-thickness factor¹⁰⁾, where the linear absorption coefficient was assumed to be the mean value of those of Fe and Pt. The order parameter as a function of annealing temperature is shown in Fig. 2. The order parameter drastically increases as the annealing temperature increases from 363°C to 413°C; at 413°C, the order parameter becomes 0.84, which is close to the upper limit of the order parameter for the composition shown in Table 1 (the upper limit of the order parameter is $2x$ and $2(1-x)$ when $x \leq 0.5$ and $x \geq 0.5$, respectively⁹⁾). It should be noted that this value of the order parameter is considerably high for a film with a thickness of less than 100 nm. Takahashi *et al.* reported that the order parameter of an FePt film deposited at 300°C increased drastically from 0 to 0.8 as the film thickness increased from 50 nm to 200 nm¹¹⁾; this result indicated the existence of a critical thickness for the ordering. The thickness of the samples used in the present study was just within the critical range.

SEM images of an as-deposited film and films annealed at various temperatures are shown in Fig. 3. The as-deposited film consists of small grains with diameters of approximately 10 nm. The grain size increases with increasing annealing temperature, and the morphology changes drastically with the drastically increasing order parameter at 413°C. This indicates that ordering is associated with the growth of grains.

In addition, coercivity also increases drastically with increasing order parameter. Figure 4 shows coercivity

Fig. 2 Order parameter as a function of annealing temperature. The horizontal line at each temperature denotes the upper limit of order parameter calculated from the composition shown in Table 1⁹⁾.

Fig. 3 SEM images of as-deposited FePt film and FePt films annealed at various temperatures.

as a function of the annealing temperature. A large coercivity of 15.6 kOe is observed at an annealing temperature of 413°C; this value of coercivity is in agreement with the high value of order parameter at this annealing temperature. The increase in coercivity at an annealing temperature below 413°C is ascribed to an increase in the magnetocrystalline anisotropy of the films with the development of the L_{10} phase. On the other hand, the decrease in coercivity at an annealing temperature above 413°C is ascribed to the change in the magnetization process from magnetization rotation to domain wall displacement with the growth of grains.

Fig. 4 In-plane coercivity of FePt films measured at room temperature. The inset shows the magnetization curve of a film annealed at 413°C.

4. Discussion

In this section, the mechanism of L_{10} ordering occurring in GFS-deposited FePt films at a relatively low temperature is discussed. From the equilibrium phase diagram, the ordering temperature of equiatomic FePt alloy is found to be approximately 1300°C¹²⁾. At this temperature, ordering occurs homogeneously with lattice diffusion¹³⁾. On the other hand, at temperatures considerably below the ordering temperature, ordering proceeds with nucleation and growth of L_{10} domains from A1 (disordered) matrix^{14),15)}. The mechanism of ordering in the GFS-deposited FePt film on annealing corresponds to the latter case.

Takahashi *et al.*¹⁵⁾ termed the ordering mechanism involving nucleation and growth “discontinuous ordering,” which was originally used to describe an ordering mechanism observed in some alloys other than FePt^{16),17)}. In discontinuous ordering, an ordered phase nucleates at the grain boundaries and grows into the interior of disordered grains¹⁷⁾. Since atoms composing the alloy migrate by grain boundary diffusion, this type of ordering proceeds at low temperatures¹⁵⁾.

In the GFS-deposited FePt films, the ordering at a relatively low temperature may proceed under conditions whereby discontinuous ordering can occur easily; this is suggested by two experimental facts. One is the small grain size in the GFS-deposited film. A small grain size is preferable for the discontinuous ordering, since the grain boundary energy in the case of a small grain size is high¹⁵⁾. Figure 5 shows the transmission electron microscopy (TEM) image of an unannealed FePt film deposited on a carbon TEM grid. The film consists of distinct grains having a diameter of approximately 5 nm. The small grain size is a result of the low energy deposition in GFS, which impedes the growth of grains.

The other experimental fact is the presence of nuclei of the L_{10} phase in the as-deposited film. Since the nucleation of the L_{10} domain requires a certain amount

Fig. 5 TEM image of as-deposited FePt film on carbon grid. The deposition time is 20 s and nominal thickness is 10 nm.

Fig. 6 XRD pattern of as-deposited film. The film thickness is 950 nm, and the substrate is glass.

of energy, the presence of $L1_0$ nuclei before annealing facilitates the ordering process. Figure 6 shows the XRD pattern of an as-deposited thick (950 nm) film. This pattern shows the presence of a 110 superstructure peak; however, this peak is not observed in the XRD pattern of the thin as-deposited film, as shown in Fig. 1. In the case of the as-deposited thick film, the detection of the 110 superstructure peak was possible because of the large mass of the film and the preferred orientation of $\langle 110 \rangle$ in the film, which is the direction normal to the film plane. However, it is speculated that the as-deposited thin film may also contain $L1_0$ nuclei.

Another possible mechanism for low-temperature ordering in FePt films, proposed in other studies is not applicable to the GFS-deposited films. The low-temperature ordering of FePt films deposited on an underlayer is induced by stress produced as a result of the lattice mismatch between FePt and the underlayer^{5),6)}. Similarly, the low-temperature ordering in FePt films sputtered at high pressure¹⁸⁾ is also induced by stress produced in the films. This stress-induced low-temperature ordering is possible only in films having an aligned crystal orientation (in most cases, the c -axis is perpendicular to the film plane) and when compression stress is produced along the c -axis. However, GFS-deposited FePt films have randomly oriented crystallites; therefore, if stress is produced in a direction parallel or normal to the film plane, it presumably has no effect on the net ordering temperature of the film.

5. Summary

GFS-deposited FePt thin films undergo $L1_0$ ordering

on annealing at relatively low temperatures; this ordering proceeds with the growth of grains. The XRD pattern of a thick FePt film shows a superstructure peak even though it is not subjected to annealing. Possible reasons for low temperature ordering are the small grain size and the existence of $L1_0$ nuclei formed during film deposition. They may promote discontinuous ordering, which is a key mechanism in low-temperature ordering.

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