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Invited Paper

$L1_0$ FePt thin films with [0 0 1] crystalline growth fabricated by SiO_2 addition—rapid thermal annealing and dot patterning of the films

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ABSTRACT

FePt films that have a high degree of order S in their $L1_0$ structure ($S > 0.90$) and well-defined [0 0 1] crystalline growth perpendicular to the film plane were fabricated on thermally oxidized Si substrates by the addition of an oxide and successive rapid thermal annealing (RTA). The mechanism of $L1_0$ ordering and [0 0 1] crystalline growth perpendicular to the film plane arising through the oxide addition and RTA process is also discussed. The $L1_0$ ordering ($S > 0.90$) and the [0 0 1] crystalline growth were achieved by (1) lowering the activation energy due to in-plane tensile stress and the initiation of $L1_0$ ordering at a low temperature, (2) [0 0 1] crystalline growth through in-plane tensile stress, and (3) enhancement of atomic diffusion via the addition of an oxide and the resultant lowering of the ordering temperature. Effect (1) was observed in the case of SiO_2 addition, effect (2) was generally observed in the case of oxide addition and the RTA process, and effect (3) was prominent in the case of ZnO addition. With the addition of ZnO, the $L1_0$ ordering started at below 400 °C and was completed at 500 °C. Finally, dot patterns were successfully fabricated down to a diameter of 15 nm using electron beam lithography, and the magnetic state of the dot pattern was observed by magnetic force microscopy.

1. Introduction

$L1_0$ FePt has a large magnetocrystalline anisotropy ($K_u \sim 7.0 \times 10^7$ erg/cm^3) due to the alternating arrangement of Fe and Pt layers along the c axis [1]. $L1_0$ FePt is expected to have applications in high-density perpendicular magnetic recording media, bit-patterned media, and spin electronic devices [2–15]. In recent decades, numerous studies have been conducted to investigate the fabrication of $L1_0$ FePt films with a well-defined [0 0 1] crystalline texture perpendicular to the film plane, focusing on technical applications as well as elucidation of the basic mechanism of $L1_0$ ordering.

Epitaxial growth on heated substrates has frequently been used to prepare [0 0 1]-oriented $L1_0$ FePt films. In many cases, single-crystal MgO (0 0 1) substrates were used in addition to Pt, Cr, CrRu, and MgO under-layers to enhance [0 0 1] crystalline growth perpendicular to the film plane [4,6,7,10,16–19]. The use of metals [20–23] and oxide additives [24–33] for non-epitaxial films has also been investigated. Non-epitaxial FePt films are usually deposited on non-crystalline substrates at room temperature and then successively treated by a rapid thermal annealing (RTA) process. Typical experimental conditions are listed in Table 1 [24–38]. This oxide addition-RTA (OA-RTA) process is very attractive for practical applications because it makes the fabrication process quite simple and yields a high throughput during manufacturing. However, despite numerous studies, a fabrication process for $L1_0$ FePt thin films with a [0 0 1] crystalline texture through the OA-RTA process has not been developed to date. Understanding the OA-RTA process is the key factor involved in obtaining high-quality films.

We fabricated $L1_0$ FePt films using the OR-RTA process. To clarify the mechanism involved, we used SiO_2, CuO, and ZnO as oxide additives and changed the heating rate from 30 to 50 °C/s. We then fabricated $L1_0$ FePt dot arrays with diameters of 15–300 nm and observed their magnetic structures by magnetic force microscopy.

2. Experimental

FePt-SiO_2 films were prepared by either co-sputtering or alternative sputtering of three targets. For practical application to high-density perpendicular magnetic recording media and bit-patterned media, a film with a thickness of less than 10 nm having high magnetic anisotropy with a magnetic easy axis perpendicular to the film plane is required. Therefore, the thickness

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Table 1
$L1_0$ FePt films fabricated by the OA-RTA process [23–38].

Additives	Thickness of FePt films (nm)	Heating	Annealing conditions		Reference
			Temperature(°C)	Time (s)	
MgO	14–20	–	600	1800	[24]
	16–20	RTA	350–600	10–1800	[25]
B_2O_3	5–60	RTA	550	1800	[26]
	16	–	550	1800	[27]
SiO_2	~50	–	450–650	1800–7200	[28,29]
	6.1	RTA	300–400	2–60	[30,31]
MnO	4.0–10	RTA	650–750	600	[32]
HfO_2	4.0–10	RTA	650	600	[32]
ZrO_2	20, 50	–	550	1800	[33]
No additives	6–24	–	550	2	[34]
	10.4	RTA	350–600	2–600	[35]
	1–4	RTA (80 °C/s)	500	1	[36]
	14.8	RTA(21.5 °C/s)	300–600	6–1800	[37]
	10	RTA (100 °C/s)	600	600	[23]
	20	RTA (30 °C/s)	360–930	5–6000	[38]

of FePt was set to 6.1 nm and the thickness of the oxide additives was varied. The films were deposited on a thermally oxidized silicon substrate at room temperature using magnetron sputtering. The Ar gas pressure during sputtering was about 0.5 Pa. The sputtered films were successively annealed by RTA, which was conducted under a vacuum of $\sim 10^{-4}$ Pa. The heating rate was 30–50 °C/s, the annealing temperature was 400–800 °C, and the films were kept at these temperatures for 10–120 min. We used SiO_2, CuO, and ZnO as oxide additives.

The crystal structure was studied by X-ray diffraction (XRD) with CuKα radiation and a transmission electron microscope (TEM). The XRD was measured by a 2θ–θ method for $2\theta = 20°$–$118°$. The degree of order (S) and the interference depth (Λ) of the $L1_0$ ordered crystallite were calculated by fitting the θ–2θ diffraction patterns using

$$I[Q] = \int L(Q)|F(Q)|^2 L_p e^{-2M} \frac{1}{1+k^2(Q-q)^2} dq \quad (1)$$

where L(Q) is the Laue function, $F(Q)$ is the structure factor, L_p is the polarization factor, e^{-2M} is the temperature factor, k is the device correction, and $Q = 4\pi/\lambda \sin\theta$. In this equation, λ is the wavelength of the CuKα radiation. The interference depth Λ is taken into account through the Laue function [39,40]. The order parameter S is included in the structure factor $F(Q)$. The details of Eq. (1) and the fitting procedure are provided elsewhere [41]. Energy decentralized spectroscopy (EDX) was used for the composition analysis and a photoelectric spectrum device (XPS) was used for the composition analysis along the depth direction.

3. Results and discussion

3.1. Effect of oxide additives and heating rate on $L1_0$ ordering and [0 0 1] crystalline growth

3.1.1. SiO_2 additive

Fig. 1 shows the XRD patterns for FePt–SiO_2 films heated at 30 °C/s and kept at 700 °C for 120 min. When the SiO_2 volume was 0 vol%, only a very weak superlattice (0 0 1) peak belonging to the $L1_0$ phase (face centered tetragonal) and fundamental (0 0 2) and (1 1 1) reflection peaks were observed. The (0 0 1) and (0 0 2) reflection peaks increased with increasing SiO_2 content to 10 vol%, while the (1 1 1) reflection peak disappeared rapidly. The (0 0 1) maximum intensity was observed at 10 vol% SiO_2, where the superlattice (0 0 3) peak and fundamental (0 0 4) lines were observed. As the SiO_2 increased from 12 to 22 vol%, the (0 0 1) intensity decreased and the [0 0 1]-oriented crystalline growth began to degrade. These findings indicate that the addition of SiO_2 enhances $L1_0$ ordering only when the proper amount of SiO_2 is added to the FePt film. The optimum amount of SiO_2 (~10 vol%)

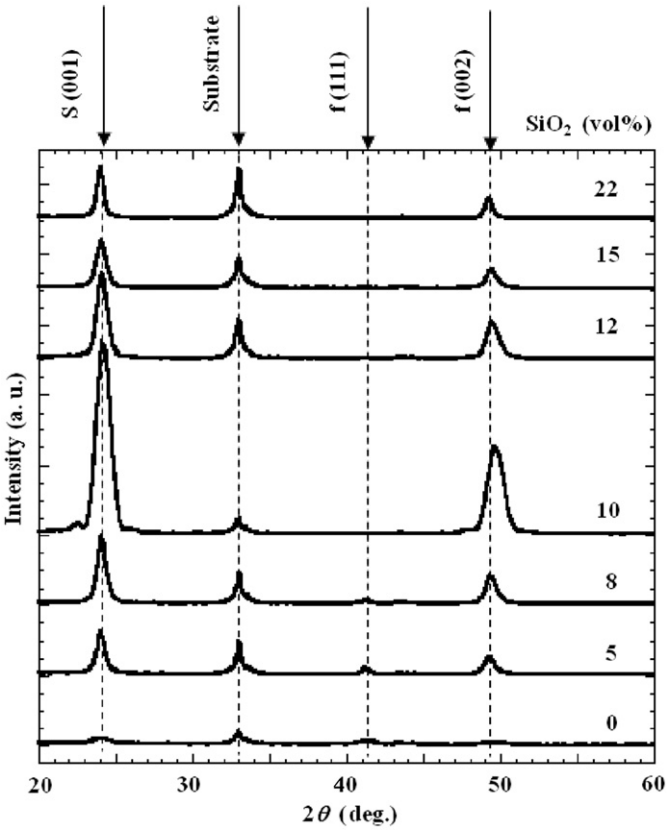

Fig. 1. XRD patterns of FePt films prepared by SiO_2 addition and the RTA process. Films were heated at 30 °C/s and kept at 600 °C for 120 min.