Measurement of thermo magnetic property and increase of areal density of $L1_0$-orderd isolated single crystalline FeCuPt grains

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We fabricated FeCuPt grains by using a combination of Rapid Thermal Annealing with Rapid Cooling Process. The isolated grains consist of mostly $L1_0$-ordered and polycrystalline structure. We carried out additional annealing on these isolated FeCuPt grains to achieve single crystallization of grains. The surface morphology and detailed crystal structure of the grains are observed. After additional annealing, FeCuPt grains retained almost the same grain size. As a result, single crystalline $L1_0$-FeCuPt isolated grains were obtained by combining RTA with RCP and by carrying out additional annealing. Furthermore, we characterized the thermo-magnetic properties of single crystalline $L1_0$-FeCuPt isolated grains and increased areal grain density by forming additional grains.

Key words: magnetic recording media, $L1_0$-FeCuPt, single crystalline, isolated grain, thermo magnetic property

1. Introduction

The concept of bit-patterned media (BPM) is considered to be one of the most promising candidates for further increasing areal storage density. Thermally assisted magnetic recording (TAMR) has attracted a great deal of attention as one of the newest recording methods. Ordered FeCuPt alloys have attracted a great deal of attention as promising materials for high density BPM with TAMR because of their high uniaxial magnetic anisotropy $K_u$, adjustable Curie temperatures by changing Cu content, and small-grain-forming capabilities. We previously reported that rapid thermal annealing (RTA) of Pt/Fe and Pt/Cu/Fe multilayered continuous films can effectively obtain perpendicularly magnetized small $L1_0$-FePt and $L1_0$-FeCuPt grains on flat thermally oxidized Si substrates. The areal densities of FeCuPt grains were drastically increased by using nano-structured substrates such as SiO$_2$ substrates with self-assembled nano-pores, self-assembled SiO$_2$ particle (SASP) arrays, and nano-convex patterns (NCPs). We introduced a rapid cooling process (RCP) into RTA to clarify the relations between grain forming processes and annealing conditions. Grain growth was prevented when RCP was combined with RTA. However, a new shoulder peak appeared at a slightly lower angle than the FeCuPt (002) peak in the X-ray diffraction (XRD) profile, which might have corresponded to the disordered structure of FeCuPt.

We investigated the detailed structures of $L1_0$-ordered isolated single-crystalline (LSI) FeCuPt grains fabricated by using a combination of RTA with RCP and additional annealing. Furthermore, we characterized the thermo-magnetic properties of LSI-FeCuPt grains and increased areal grain density by forming additional grains.

2. Experimental method

Films with multilayered structures of Pt/Cu/Fe were fabricated before annealing using DC magnetron sputtering on a flat thermally oxidized Si substrate. Fe, Pt, and Cu layers were fabricated in a 0.18-Pa pressure ArH ($H_2$: 3 vol. %) gas atmosphere. The base pressure was less than $5.0 \times 10^{-5}$ Pa. The total thickness of the deposited multilayer estimated from the deposition rate was 3.75 nm (Pt 1.84 nm/Cu 0.47 nm/Fe 1.44 nm). The composition of the films was Fe$_{43}$Cu$_{14}$Pt$_{43}$.

The crystallization involved annealing Pt/Cu/Fe multilayers using the RTA process in a vacuum chamber at $8 \times 10^{-4}$ Pa, with a 2-kW infrared ray lamp. We subsequently investigated two different cooling processes at the end of RTA that consisted of either natural cooling or quenching, by introducing an N$_2$ gas flow to the atmospheric pressure until the temperature was reduced to room temperature (RT) in order to clarify the effect of RCP. The heating rate ($T_h$) was controlled by varying the lamp power: after it reached the required maximum temperature ($T_m$), the shutter was closed to shut out the optical pass, and the lamp was switched off simultaneously. The temperature was measured with a thermocouple attached to the back side of the Si substrate. The crystalline structures were determined by XRD with Cu-K$_\alpha$ radiation and transmission electron microscopy (TEM). The morphology of grains was observed by scanning electron microscopy (SEM) and TEM. Magnetic images were observed by atomic/magnetic force microscopy (AFM/MFM). The dependence of magnetic properties on temperature was evaluated with vibrating sample magnetometry (VSM).
3. Single crystallization by additional annealing

We fabricated FeCuPt grains by RTA with RCP under a variety of $T_m$ and $T_R$ conditions in an experiment. The smallest isolated grains were fabricated at $T_R = 150{\degree}C/s$ and $T_m = 460{\degree}C$ under various $T_R$ and $T_m$ conditions. Differences in contrast can clearly be seen at the rim of each grain in the SEM image in Fig. 1. Moreover, new peaks in the XRD profile were found near the $L1_0$-FeCuPt (002) peak shown in Fig. 2. This may have been caused by the electron channeling contrast produced by the existence of disordered FeCuPt, which corresponded to a new peak at a slightly lower angle of the (002) peak. Then, measurements with TEM and electron beam diffraction were carried out to investigate the local crystal structures in single grains, which are indicated by the inhomogeneous brightness in the SEM image. Differences in brightness were also observed in the grains from TEM bright field images and electron beam diffraction measurements of typical grains, as shown in Fig. 3(A). The dark field images from spots a), b), and c) are shown in Fig. s 3(C)-(E). Clear (110) super lattice peaks appeared; however, dark field images from diffraction spots a) and b) mean grains still have a poly crystalline structure. Further, the rims of grains may belong to other phases from lower intensity spot c), as seen in Fig. 3(B). An $L1_0$-ordered structure is a major part of the grains, but other phases with crystal structures can be found around the edges of the grains. Thus, FeCuPt grains with mostly $L1_0$-ordered poly-crystalline structures were obtained by RTA with RCP.

Therefore, we carried out additional annealing on these isolated FeCuPt grains by using the same annealing chamber as that for RTA in order to crystallize these poly-crystal grains to form single crystalline grains. The annealing conditions were set to $600{\degree}C$ for 1 h. The ordering temperature of FePt alloy was $\sim 600{\degree}C$ and the estimated atomic diffusion length was on the order of the third nearest neighbor distance in FePt under annealing conditions. A comparison of the SEM images of FeCuPt grains shows that the inhomogeneity of brightness in each grain disappeared after additional annealing as shown in Fig. 4. The average annealed grain diameter of 53.4 nm was almost the same as the average pretreatment grain diameter of 53.2 nm. A comparison of the XRD profiles showed that the intensity of the (001) super lattice peak that indicated the formation of an $L1_0$-ordered phase increased, and the shoulder peak at a slightly lower angle of (002) disappeared, as shown in Fig. 5. Thus, FeCuPt grains retained almost the same grain size and were expected to form single crystalline grains.

Furthermore, we observed a crystal structure of a typical single grain after additional annealing with TEM. Octagonal symmetrical shapes appeared in most grains, as shown in Fig. 6(a). The edges of the octagonal grains were derived as (100), (010), (110), and (110) facets of $L1_0$-FeCuPt by comparing the bright field images and electron beam diffraction patterns in Fig. 6.

A series of [110] super lattices and [200] lattice spots is observed with fourfold-symmetry from electron beam diffraction measurements of single grains, as shown in Fig. 6(b). The {110} spots indicate the formation of $L1_0$-ordered structures. Thus, we found the grains had c-axis oriented single crystalline structures from complementary results of XRD that covered macroscopic areas and from localized electron beam diffraction. Namely, c-axis orientated LSI-FeCuPt grains were obtained from the use of RTA combined with RCP and additional annealing.

![Fig. 1](image1.png)  
**Fig. 1** SEM planer view of isolated FeCuPt grains prepared by RTA with rapid cooling at $T_m = 460{\degree}C$ and $T_R = 150{\degree}C/s$.

![Fig. 2](image2.png)  
**Fig. 2** XRD profile of isolated FeCuPt grains prepared by RTA with rapid cooling at $T_m = 460{\degree}C$ and $T_R = 150{\degree}C/s$. The peak positions of two-theta angle for the corresponding crystal structures are estimated by using the data quoted from the previous work.

![Fig. 3](image3.png)  
**Fig. 3** (A): Bright field image of TEM for a typical isolated grain prepared by RTA with rapid cooling process at $T_m = 460{\degree}C$ and $T_R = 150{\degree}C/s$. (B): Electron beam diffraction pattern of the grain. The spots labeled as a) and b) correspond to the $L1_0$-FeCuPt (110) super lattice and (200) lattice diffraction, spot c) belong to other phase. (C),(D),(E): Three dark field images from spots a), b) and c).
4. Magnetic properties

The (001)-oriented LSI-FeCuPt grains were expected to have perpendicular magnetic anisotropy. Observed MFM images of as-deposited grains were quantized to the two kinds of contrast shown in Fig. 7, which were expected to be perpendicularly magnetized particles. We then measured the thermal properties of the same samples, which are important characteristic parameters of TAMR media. The dependence on temperature of saturation magnetization ($M_s$), coercivity ($H_c$), and saturation magnetic field ($H_s$) of LSI-FeCuPt grains are plotted in Fig. 8 with the application of an out-of-plane magnetic field. As can be seen in the figure, $M_s$, $H_c$, and $H_s$ decreased with increasing temperature. Furthermore, Curie temperature $T_c$ was ~290°C and $H_s$ was less than 10 kOe at 200°C. Thus, we confirmed that a reasonable decrease in $T_c$ and $H_c$ occurred by adding Cu to FePt.

5. Increase of areal density of grains

Important issues to achieve high density recording media are to miniaturize grains and increase their areal density. We focused attention on the dependence of average grain diameter ($D_a$) on initial deposition thickness ($T_s$) as a method of miniaturizing the grains. The results in Fig. 9 indicate that the value of $D_a$ monotonically decreased and the areal density increased by reducing $T_s$. The $D_a$ was 7.05 nm, and the areal density of grains was 2.9 T particles/inch$^2$ for $T_s = 1.25$ nm. Furthermore, Fig. 10 shows that these grains with high areal density are also single crystalline grains similar to those discussed in Section 3.

The same deposition and rapid thermal annealing procedures were repeated because we expected further increases in the areal density of grains with nucleation. The $D_a$ is almost the same in Figs. 11(a) and (b), and particle density is increased. However, $X_d$ is increased and particle density is decreased in Figs. 11(b) and (c). Areal grain density was increased due to nucleation between original grains when the relations of particle density with $D_a$ and average distance between nearest grains ($X_d$) were compared, after the grain fabrication procedure was repeated for samples that had $X_d$ larger than $D_a$. However, when it was repeated with $X_d$ that was already smaller than the original $D_a$, areal grain density decreased with the coalescence of grains during growth.
Isolated FeCuPt grains were obtained by applying RTA with RCP to Pt/Cu/Fe multilayer continuous films that were fabricated on thermally oxidized Si substrates. We carried out additional annealing on these isolated FeCuPt grains to achieve single crystallization in them. After additional annealing, grains were almost the same size in the SEM images, the intensity of the (001) super lattice peak increased, and the peak of disordered structures disappeared in the XRD profiles. Furthermore, the series of {110} super lattice spots were observed with fourfold-symmetry from electron beam diffraction measurements of single grains. As a result, single crystalline L1₀FeCuPt isolated grains that were c-axis oriented were obtained by combining RTA with RCP and by carrying out additional annealing.

The MFM images of individual grains were obtained and quantized to two kinds of contrast to characterize their magnetic properties, which we expected to contain perpendicularly magnetized particles. The results of measuring the thermo-magnetic properties of the same samples indicated that the Curie temperature was ~290°C and $H_{\text{c}}$ was less than 10 kOe at 200°C. Thus, we confirmed a reasonable decrease in $T_c$ and $H_c$ by adding Cu to FePt.

Furthermore, we studied the increased areal density in grains. The value of $D_a$ monotonically decreased, and areal density increased due to the decrease in initial deposition thickness $T_s$. We confirmed through additional grain formation processes that areal density only increased when the average grain distance was larger than the average grain diameter.

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**References**


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