Switching field distribution and magnetization reversal process of FePt dot patterns

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Abstract:

The fabrication of FePt nanodots with a high structural quality and the control of their switching fields are key issues in realizing high density bit pattern recording. We have prepared FePt dot patterns for dots with 15–300 nm diameters by electron beam lithography and re-annealing, and studied the relation between magnetization reversal process and structure of FePt nanodots. The switching field (H_{sw}) of dot patterns re-annealed at 710°C for 240 min showed a bimodal distribution, where a higher peak was found at 5–6 T, and a lower peak was found at ~ 2 T. It was revealed by cross-sectional TEM analysis that the structure of dots in the pattern can be classified into two groups. One group has a high degree of order with well-defined [001] crystalline growth, and the other group includes structurally-disturbed dots like [111] growth and twin crystals. This structural inhomogeneity causes the magnetic switching field distribution observed.

Key words: FePt, Dot Pattern, Magnetization Reversal, Switching Field, Coercivity,

1. Introduction

$L1_0$ FePt is expected to be the most suitable material for bit pattern media because of its large uniaxial magnetic anisotropy. For an assumed coherent rotation of magnetization in an $L1_0$ FePt dot, the coercivity at room temperature is calculated to be over 10 T at uni-axial magnetic anisotropy constant K_u of $\sim 7 \times 10^7$ erg/cm³, with a saturation magnetization M_s of ~ 1100 emu/cm³¹⁻⁴. $L1_0$ FePt films have been fabricated by epitaxial methods²⁻⁶, and by annealing with additives like Cu, Ag, and oxide⁷⁻¹⁰. Such films show excellent magnetic properties with a high order parameter (S) for an $L1_0$ structure and [001] crystalline growth perpendicular to the film plane. Patterning $L1_0$ FePt films into dot arrays has been done mostly by electron-beam lithography and ion-milling using the high quality films mentioned above. Self-organized resist patterns¹¹ can result in dot patterns on full disk substrates. The fabrication of FePt nanodots, with a practical dot size less than 20 nm and high structural quality, has become the focal issue in efforts to make high density bit pattern recording possible.

The coercivity of FePt particles and patterned dots has been reported to be 2–4 T, and generally shows a wide distribution of switching field strengths^{4,9,12-16}. These coercivities may be high enough for bit patterned media and thermally assisted magnetic recording, but their distribution must be controlled. Magnetization switching may take place through a magnetization reversal of nucleation sites and successive domain expansion to the whole dot¹⁷. Structural defects may become nucleation sites for magnetization reversal, and could then be the cause of the observed degradation of H_{sw} and its wide distribution of measured values^{12,13}.

In this paper, FePt dot patterns with 15–300 nm diameter dots were prepared by

electron-beam lithography, ion-milling, and re-annealing, and the correlation between the switching field distribution and the structure of patterned dots has been studied.

2. Experimental Details

2-1 Fabrication of films and patterning

FePt was sputtered on thermally oxidized Si substrates. 10% SiO₂ was added during sputtering to promote *L1₀* ordering at low temperatures⁹⁻¹¹. For practical application to bit-patterned media, a film with less than 10 nm is required. Thus, we used an FePt thickness of 6.1 nm in this experiment. Since FePt films on Si substrate become a particulate film when heated over 600 °C¹⁰, sputtered films were first annealed by rapid thermal annealing (RTA) at about 570 °C for 10 min. The crystal structure of films was studied by X-ray diffraction (XRD) using Cu K α radiation. The degree of order of films annealed at 570 °C for 10 min was found to be $S \sim 0.89$ by fitting the θ -2 θ diffraction patterns. The films showed a [001] crystalline growth, and [111] and [110] diffraction lines were not observed in the film. After the annealing, patterns were formed in 100 μ m squares by electron-beam lithography and ion-milling^{18,19}. We used a negative resist of *p*-chloromethyl-methoxy-calix[4]arene (TEBN-1 provided by Tokuyama Corp.). The concentration of resist solutions was controlled so that the thickness of resist became 15 nm. The resist films were prepared by spin coating the resist solution on a substrate at 2000 rpm, and prebaking at 110 °C for 1 min. The resist films were patterned by e-beam writing system (ELS7500, ELIONIX Inc.) operated at 50 kV and 50 pA. After exposure, the resist patterns were developed in *o*-xylene for 30 s and isopropanol for 30 s. FePt dot patterns were transferred through the resist mask by using Ar ion milling at 200 keV and 60 mA with rotating substrate. A grazing incident angle of Ar ions was set at 60 degrees to film surface. Dot diameters

were designed to be 15~300 nm.

The pattern prepared by the above-mentioned process is called pre-annealed patterns hereafter. Finally, patterns were annealed again at 710 °C to eliminate structural damage resulting from nanofabrication and to promote an fcc to $L1_0$ transformation. These patterns are called re-annealed patterns. The schematic diagram for the film preparation and patterning is shown in Fig. 1.

2-2 Structural analyses and magnetic properties

Geometric characterization of patterns was done by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The SEM and AFM images of the pre-annealed pattern (0.71 T bit/inch²) with a 15 nm dot diameter and 30 nm pitches are shown in Fig. 2a and 2b, respectively. The dispersion in dot location is shown in Fig. 2c, and was found to be 5 nm, with a statistical position variance (σ/p) of ~ 10%.

The structure of the pre-annealed patterns was studied by micro X-ray diffraction (μ -XRD) at BL13XU beamline²⁰ at SPring-8, a synchrotron radiation facility. Width of micro X-ray beam is 1.2 x 0.3 μm^2 . The crystal structure of the re-annealed patterns was analyzed by cross-sectional transmission electron microscopy (TEM).

Hysteresis curves of dot patterns were measured by X-ray magnetic circular dichroism (XMCD) at the Pt L_3 edge, with static magnetic fields up to 10 T at BL39XU beamline²⁰ at SPring-8. Remanent magnetization curves of the dot pattern was also measured using the Kerr effect by applying a pulse field of up to 7 T to consider a thermal fluctuation effect. The time-domain half width of the pulse field was 1 ms.

3. Experimental results and discussion

3.1 Crystalline structure of dot patterns

The results of micro-XRD for the pre-annealed pattern are shown in Fig.3, where the diffraction intensity was normalized by the (001) diffraction intensity of the pre-annealed film and corrected by taking account of a change of an areal density of FePt dots. Both (001) superlattice line and (002) fundamental lines were clearly observed for all samples. However the (001) line gradually decreases with reducing dot size to 30 nm diameter and decreases extremely with a further reduction of dot size to 15 nm. At the same time, a decrease of the relative ratio $I_{(001)}/I_{(002)}$ is observed, which suggests a lowering of degree of order (S). The intensity of the fundamental (002) line also shows a decrease with a reduction of dot size, which is typically observed at $d = 15$ nm. Namely, it is considered that the film structure with the [001] growth perpendicular to the film plane collapsed, or even a crystallinity itself was lost during ion-milling. The structure of the disturbed area may be a fcc (or amorphous) state ^{12,13,18,21}, but a coherent lattice relation between the $L1_0$ phase and the induced fcc phase is quite small. When such structurally degradation is assumed to be formed at sidewall of patterned dots, a thickness of the disturbed layer is roughly estimated to be 3-5 nm from the change of the (001) and (002) intensity as a function of dot size.

The crystalline structure of dots in the re-annealed pattern was analyzed by cross-sectional TEM. Cross-sectional TEM analysis was focused on the 30 nm dot patterns re-annealed at 710 °C for 240 min. The structures of 15 individual dots were chosen for analysis. The typical bright field images are shown in Fig. 4. The structure of these dots can be classified into two groups. The first group has a high degree of order with well-defined [001] crystalline growth, which can be clearly seen in the layered bright and dark lattice image in Fig. 4b. In this group, the structural degradation, like an fcc or amorphous phase resulting from ion milling, was not

detected within the resolution of the TEM images. The second group includes mis-oriented dots like [111] growth (Fig.4c), twin crystals (Fig.4d). In total, 8 of the 15 dots observed belong to the first group and the rest belong to the second group.

From the micro-XRD and TEM studies for the pre-annealed patterns and the re-annealed patterns, a structural change in dots is supposed to take place in the following way;

(1) In the pre-annealed pattern, each dot consists of a mixed structure of the $L1_0$ ordered phase and the structurally-disturbed (fcc or amorphous) phase. This structurally-disturbed area is considered to arise from ion-milling of the patterning process and to be formed at sidewall of dots.

(2) By re-annealing process, a structure disturbed phase induced by ion-milling re-transforms into an $L1_0$ phase to some extent. Accordingly, some mis-oriented dots and twins were formed through re-crystallization, and the dots make two groups with regards to a structural quality. The first group has a high degree of order with well-defined [001] crystalline growth. The second group includes mis-oriented dots like [111] growth, twin crystals, and structurally unclear dots. These two groups are expected to have different magnetic properties, which will be shown in the next paragraph.

3.2 Magnetic properties of patterned dots

Hysteresis curves measured by XMCD are shown in Fig. 5. Coercivity (H_c) depends on both the sample preparation process and the dot size. The maximum H_c for the pre-annealed pattern was about 1.5 T, and the coercivity was increased by re-annealing. The maximum H_c in our experiment was ~ 3.0 T, which was obtained after re-annealing at 710 °C for 240 min.

Increased coercivity through re-annealing was observed for dot diameters of 30 and 100 nm typically, as shown in Fig. 6. However the increased coercivity was always accompanied with a tilting of the hysteresis loops as shown in Fig. 5. The tilting of hysteresis loops indicates that the coercivity of dots is not uniform in the patterns, but has a statistical distribution.

The switching field distribution (SFD) was evaluated through a derivative of hysteresis curves as shown in Fig. 7. The SFD exhibits a single peak in the pre-annealed state, but it changes to bimodal or even trimodal after re-annealing. Here the main peak of the SFD was at ~ 2 T, while an additional peak appeared at ~ 5 T in our experiment. The volume ratio of the higher peak for 30 and 15 nm dots was 30–40% after re-annealing at 710 °C for 240 min.

There are two groups in the re-annealed pattern with regards to the structural quality as described in the previous paragraph. The first group has a high degree of order with well-defined [001] crystalline growth. The second group includes structurally degraded dots. These two groups clearly explain the bimodal distribution of H_{sw} shown in Fig. 7. The dot size in our experiment (15–100 nm) is smaller than the single domain size for FePt (about 150 nm in diameter) ^{17,22}, and coherent rotation of magnetization switching is expected in principle. In this case, the switching field is the highest ($H_k = 2 K_u/M_s$) when the field is applied parallel to the magnetic easy axis, while it is only half this strength when the field is applied at 45 degrees to the easy axis. Although the pattern surface became uneven after re-annealing (Fig. 4a), the inclination of the magnetic easy axis from perpendicular to the film plane was negligible in the first group. It is concluded that the mis-oriented dots, twin crystals, and poorly ordered $L1_0$ dots of the second group are responsible for the lower H_{sw} component, while the first group is

responsible for the higher H_{sw} component shown in Fig. 7.

The H_{sw} distribution of the pre-annealed patterns shows a single peak and its average H_{sw} is lower than re-annealed patterns. This is understood in such a way that a $L1_0$ FePt core is surrounded by the structure disturbed area (fcc or amorphous) in every dots. The disturbed area is expected to have a soft magnetic property and its thickness is very thick in the pre-annealed pattern compared to the re-annealed pattern, resulting in the low H_{sw} in the pre-annealed patterns.

3.3 Magnetization reversal mechanism of FePt dots

In the first group of the re-annealed pattern, $L1_0$ structure looks almost perfect as seen in Fig.4b with the [001] axis perpendicular to the film surface. The switching field for this group is, however, 4–6 T and is rather smaller than the expected value from the magnetocrystalline anisotropy ($K_u \sim 4\sim 7 \times 10^7$ erg/cm³). Despite intensive research on FePt dot patterns, and on epitaxial and granular films, the maximum coercivity of $L1_0$ FePt dots is always found to be around 4–6 T^{9,12-16,23-26}. Our results agree with such reports. There seems to be two possible reasons for the reduction of coercivity from the expectation. One is a very thin disturbed area remaining even after re-annealing and the other is an effect of thermal fluctuations of magnetization.

The micro-magnetic simulation using the Landau-Lifshitz- Gilbert equation (LLG simulation) was carried out to estimate a volume of structurally disturbed soft magnetic phase. An assumed model structure is shown in the inset of Fig.8, where a soft magnetic layer is surrounding hard magnetic core. The dot shape is 15nm~100nm in diameter and 6 nm in height. The values of magnetocrystalline anisotropy of the inner hard magnetic core and the soft magnetic sidewall are assumed to be 6×10^7 erg/cm³ and zero, respectively. The magnetic easy axis of the inner core is set to parallel to dot axis.

Magnetization is 1100 emu/cm³, and cell size is 1 nm³. The hysteresis curves was square except dots with the soft layer thickness of 3 and 4 nm, in which small steps appeared in the magnetization reversal process. It was confirmed that the magnetization reversal starts at a soft magnetic sidewall and a reversed domain expands to the whole dot successively. The coercivity obtained by the simulation is shown in Fig.8. One can see the even 1 nm soft magnetic layers of sidewalls deteriorates H_{sw} from over 10 T to 5-6 T and the coercivity decreases to less than 2 T at thickness of 3-5 nm. Although structurally-disturbed area around dots were not observed in our TEM experiments within resolution, a structural degradation surrounding dots may remain even after the re-annealing ^{6,27}.

To consider the effect of a thermal fluctuation, the SFD curve for the re-annealed 100 nm dots was measured by the Kerr effect in a pulse field of up to 7 T. The time-domain half width of the pulse field was 1 ms. The SFD curve obtained is shown in Fig.9 together with the SFD obtained with XMCD. It is noted that the SFD curve measured using the pulse field method is shifted about 10 % to a higher field side compared with the result from XMCD. When the magnetization switching is expressed in terms of the coherent rotation, the activation volume, V , and an intrinsic reversal field ($H_{sw}^0 = 2 Ku/M_s$) are related by Scharrock's equation^{28,29},

$$\frac{H_{sw}(\tau)}{H_{sw}^0} = 1 - \left[\frac{k_B T}{KuV} \cdot \ln\left(\frac{A^* \cdot \tau}{\ln 2}\right) \right]^{\frac{1}{2}},$$

where A^* is the exchange stiffness constant ($\sim 10^{-9}$ s), T the measurement temperature, τ is the time kept at T , and k_B is the Boltzmann factor. The values of $KuV/k_B T$ and H_{sw}^0 can be evaluated through switching field measurements with different values for τ . We, therefore, evaluated H_{sw}^0 using Scharrock's equation. The resultant value for H_{sw}^0 is

5.9 T for the lower peak and 9.4 T for the higher peak in Fig. 9, respectively. H_{sw}^0 for the higher peak is almost the same as the expected value (over 10 T) for the coherent rotation model. This result may suggest the coercivity reduction is attributable to the thermal fluctuation effect too. In conclusion, we suppose both the structurally disturbed soft magnetic area remaining even after the re-annealing and the thermal fluctuations of magnetization are responsible for the reduction of coercivity from the expectation.

4. Summary

FePt dot patterns with diameters of 15–100 nm were prepared by electron beam lithography. The switching field, H_{sw} , of re-annealed dot patterns showed a bimodal distribution, with a higher H_{sw} peak at ~ 5 –6 T and a lower H_{sw} peak at ~ 2 T. It was revealed by cross-sectional TEM analysis that the structure of patterned dots could be classified into two groups. The dots in the first group were found to have a high degree of order and [001] crystalline growth, while the second group included mis-oriented dots and twin crystals. We conclude that the first group has an H_{sw} value of 4–5 T, while the second group is responsible for lower (~ 2 T) H_{sw} component.

The coercivity at room temperature is calculated to be over 10 T at uni-axial magnetic anisotropy constant K_u of $\sim 7 \times 10^7$ erg/cm³ based on the coherent magnetization rotation model. The switching field observed for the first group is however 4–6 T, and is only about a half of the expected value. There seems to be two possible reasons of this discrepancy. One is the soft magnetic structurally-disturbed area which remains even after re-annealing and the other is the effect of thermal fluctuation of magnetization. The mechanism for the reduction of coercivity from the expectation was discussed based on the LLG simulation and the thermal fluctuation

measurement.

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Fig.1 Schematic diagram of sample preparation

Fig.2 SEM (a) and AFM (b) images of the pattern with a 15 nm dot diameter and 30 nm pitches (bit density = 0.71 T bit/inch²). The dot location dispersion is shown in (c).

Fig.3 Micro-XRD pattern for the pre-annealed film and patterns. The diffraction intensity is normalized by the (001) intensity of the film and corrected by taking account of a change of an areal density of FePt dots.

Fig.4 Cross sectional TEM images of patterned dots. The dot diameter is 30 nm and the pitch is 100 nm. (b) is the typical dot image of the group (1). The dot (c) and (d) belong to the group (2).

Fig.5 Magnetization curves for the pre-annealed and re-annealed patterns measured by XMCD.

Fig.6 Coercivity of the pre-annealed and re-annealed patterns as a function of dot diameter.

Fig.7 Switching field distributions of FePt dot patterns.

Fig.8 Dependence of the coercivity on the thickness of assumed soft magnetic layer formed at sidewalls. An assumed model structure is shown in the inset. Simulation parameters are written in the text.

Fig.9 Switching field distribution (SFD) of the re-annealed pattern measured by XMCD and by Kerr effect with a pulse field. A dot size is 100 nm in diameter.