**Perpendicular Anisotropy and Microstructure of MBE-grown FePt-Ag and FePt-MgO Granular Films**

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In order to study the low temperature process to fabricate (001) oriented FePt granular structure, FePt-Ag and FePt-MgO films were fabricated by the repetition process of deposition of FePt-Ag (0.83 nm) or FePt-MgO (0.83 nm) alloy layer and subsequent annealing at $T_a = 200^\circ C - 400^\circ C$. The FePt-Ag film showed (001) oriented structure together with rather high ordering parameter of 0.5 even at a low process temperature of 250 $^\circ C$. The ordering parameter of FePt-Ag was found to be much higher than that of FePt-MgO. The FePt-Ag prepared by the repetition process at 250 $^\circ C$ had well isolated grain structure with diameters of several nm to several tens nm, and showed a large uniaxial anisotropy of $1.0 \times 10^7$ erg/cc. The application of the repetition process and Ag addition into FePt was found to be quite effective to fabricate well isolated L10 ordered FePt granular structure at a low process temperature.

**Key words:** L10 FePt phase, Perpendicular anisotropy, Granular film, FePt-Ag film, MBE growth

1. Introduction

FePt film with a tetragonal L10 phase is expected to be a potential candidate for high density perpendicular recording media because of its extremely large uniaxial magnetic anisotropy. In order to apply the FePt films to practical recording media, fabrication of nano-sized grains, control of (001) orientation and lowering of ordering temperature of L10 phase are required.  

Mukai et al. reported that the small grains having a diameter of 14.5 nm were produced on (001) oriented MgO polycrystalline layer by the repetition process with the deposition of 0.3-0.5 nm thickness FePt and post-annealing at around 500$^\circ C$. On the other hand, for lowering the ordering temperature of L10 FePt, it was reported that post annealing of multilayered structure containing Ag layer was effective.

In our previous paper, (001) oriented FePt-Ag granular films were fabricated by the repetition of a process: deposition of FePt (0.5 nm) / Ag (0.2 nm) bilayer and subsequent annealing in the MBE chamber, and large perpendicular anisotropy around $1 \times 10^7$ erg/cc resulting from the formation of (001) oriented FePt was reported to be obtained at a process temperature of 250$^\circ C$. In this paper, we report the (001) oriented granular FePt obtained by the repetition process of co-deposition of FePt-Ag, or FePt-MgO and subsequent annealing, and compare the effects of Ag and MgO on the fabrication of (001) oriented FePt granular films.

2. Experiment

FePt-Ag and FePt-MgO films were deposited onto an MgO(001) single crystal substrate by MBE method at a pressure of less than $3 \times 10^{-7}$ Pa. The deposition rate of FePt is about 0.02 nm/sec, which is slower than that of commonly used sputtering process. The films were fabricated by the repetition process of deposition of FePt-Ag (0.83 nm) or FePt-MgO (0.83 nm) layer and subsequent annealing at $T_a = 200^\circ C - 400^\circ C$. During the process, the deposition was carried out at a temperature lower than 100$^\circ C$. The repetition was fixed at 6 times, and the total thickness of the film is 5 nm. We call the samples fabricated by this 6 times repetition process as [FePt-Ag (0.83 nm)]$_6$ and [FePt-MgO (0.83 nm)]$_6$. For comparison, FePt$_{60}$Ag$_{20}$ (5 nm) and FePt$_{40}$MgO$_{20}$ (5 nm) were grown on MgO(001) at various growth temperatures $T_g = 200^\circ C - 400^\circ C$. The film structure was characterized by in-situ reflection high energy electron diffraction (RHEED) and ex-situ X-ray diffraction (XRD). The hysteresis loops and magnetic anisotropy were measured by alternating gradient field magnetometer (AGM) and torque magnetometer, respectively. The torque curves were measured in the temperature range from room temperature to 400$^\circ C$, to investigate the temperature dependence of the anisotropy. The surface topography was checked by atomic force microscopy (AFM).

3. Results and Discussions

Figure 1 shows XRD profiles for [FePt-Ag (0.83 nm)]$_6$ and [FePt-MgO (0.83 nm)]$_6$ prepared at $T_a = 250^\circ C$ and 400$^\circ C$. The profiles of FePt-Ag (5 nm) and FePt-MgO (5 nm) grown at $T_a = 250^\circ C$ and 400$^\circ C$ are also shown in the figure. In all the profiles FePt 002 peak was clearly seen, and the peaks originated from (111) orientation were not confirmed, indicating that all the films are grown with (001) orientation. Moreover, the FePt 001 peak originated from the formation of the L10 phase was seen in all the profiles. The 001 peak intensities increased with increasing $T_g$ and $T_a$, which means the L10 ordering was promoted by increasing the fabrication temperature of $T_g$ and $T_a$. In the case of [FePt-Ag (0.83 nm)]$_6$, the ordering parameter $S$, estimated from the ratio of integrated intensities of 001
and 002 peaks \( I_{001} / I_{002} \) was 0.45 and 0.55 for \( T_a = 250 \)°C and 400 °C, respectively. This ordering parameter is much higher than those of \([\text{FePt-MgO} (0.83 \text{ nm})]_6; 0.2 \) and 0.4 for \( T_a = 250 \)°C and 400 °C, respectively. This implies that the Ag addition into FePt significantly reduces the ordering temperature of L1\(_0\) FePt.

Overall, the single layer (5 nm) grown on heated substrate exhibits slightly higher ordering parameter, e.g., \( S \) for FePt-Ag (5 nm) at \( T_a = 250 \)°C and 400 °C were 0.65. The FePt-Ag (5 nm) has good (001) orientation compared to \([\text{FePt-Ag} (0.83 \text{ nm})]_6\), which will be described later, and it is considered that there exist grains with (100) orientation in \([\text{FePt-Ag} (0.83 \text{ nm})]_6\), and that the 002 peak comprises of the overlap of 002 and 200 peaks. This will reduce the intensity ratio \( I_{001} / I_{002} \), resulting in the reduced \( S \) in \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) even at the same process temperature.

As shown in Fig. 1, the XRD profiles was not significantly dependent on the heating method, e.g., \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) and \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) exhibited similar profiles each other, however, the surface morphology was quite different between the two heating process. Figures 2 (a) and (b) show AFM images taken for \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) prepared at \( T_a = 250 \)°C and \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) grown at \( T_a = 250 \)°C, respectively. In both images, the grains having diameters from several nm to several tens nm are seen, but, as shown in Fig. 2 (a)', the height of the grains was about 3 – 5 nm for \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) at \( T_a = 250 \)°C, which is comparable to the total film thickness. The electric resistivity of the film, checked by two-point probe method, suggests that the \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) is an insulating film, indicating that the grains are physically isolated. Mukai et al. also reported that such a repetition process promotes the island growth of FePt due to the thermally assisted surface migration \(^3\). On the other hand, the surface of the FePt-Ag (5 nm) grown at \( T_a = 250 \)°C was smooth compared to \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) at \( T_a = 250 \)°C, which is comparable to the total film thickness. The electric resistivity of the film, checked by two-point probe method, suggests that the \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) is a metallic film, indicating that the grains are physically isolated. Mukai et al. also reported that such a repetition process promotes the island growth of FePt due to the thermally assisted surface migration. 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The AFM image of the \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) prepared at \( T_a = 250 \)°C is shown in Fig. 2 (c). In case of Ag addition during FePt deposition, highly ordered and well separated FePt grains were obtained. However, the FePt-MgO was found to have grains with a height of

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Fig. 1 X-ray diffraction profiles of \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) and \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) processed at \( T_a = 250 \)°C and 400 °C. The profiles of FePt-Ag (5 nm) and FePt-MgO (5 nm) grown at \( T_g = 250 \)°C and 400 °C are also shown in the figure.

Fig. 2 AFM images of (a) \([\text{FePt-Ag} (0.83 \text{ nm})]_6\) processed at \( T_a = 250 \)°C, (b) FePt-Ag (5 nm) grown at \( T_a = 250 \)°C, and (c) \([\text{FePt-MgO} (0.83 \text{ nm})]_6\) processed at \( T_g = 250 \)°C. Z range of all the images was fixed at 5 nm. The lower right figures (a)', (b)', and (c)' are typical line profiles of Figs (a), (b), and (c), respectively.
around 1 nm (Fig. 2 (c’)), and confirmed to be metallic from the two-point probe resistivity measurement, which means that the [FePt-MgO (0.83 nm)]₆ at Tₐ = 250°C is regarded as a continuous film. Thus the Ag addition is considered to be effective not only to reduce the L1₀ ordering temperature but also to stimulate the grain growth.

Figure 3 shows M-H loops of (a) [FePt-Ag (0.83 nm)]₆ prepared at Tₐ = 250°C and (b) FePt-Ag (5 nm) grown at Tₐ = 250°C, and (c) [FePt-MgO (0.83 nm)]₆ prepared at Tₐ = 250°C. As shown in Fig. 3 (a), the [FePt-Ag (0.83 nm)]₆ exhibits a large coercivity more than 5 kOe even at a process temperature of 250°C. The coercivity of this sample is much higher than that of previously reported FePt-Ag granular film fabricated from FePt/Ag multilayer strucre. This suggests that the co-deposition of FePt-Ag instead of the previously FePt/Ag multilayer deposition is effective to obtain magnetically isolated granular structure. The magnetization of the [FePt-Ag (0.83 nm)]₆, Mₐ = 700 emu/cc, is smaller than the value expected from the mixture of L1₀ FePt and non-magnetic Ag. Using the Mₐ of L1₀ FePt, 1140 emu/cc, the Mₐ of (FePt)₉Ag₂₀ is simply calculated to be 880 emu/cc. One of the reasons why the small Mₐ for [FePt-Ag (0.83 nm)]₆ was obtained is insufficient field to saturate the sample. One can see the relatively large Hₑ in the in-plane M-H loop shown in Fig. 3 (a). This implies that the c-axis of some FePt grains are along the film plane, which makes hard to saturate magnetization even when the external field is applied in the film normal direction. From the XRD profiles shown in Fig. 1, one can see that the diffraction angle of 002 peak of [FePt-Ag (0.83 nm)]₆ at 250 °C is slightly lower than that of FePt-Ag (5 nm) at 250 °C, implying the overlap of 002 and 200 peaks in [FePt-Ag (0.83 nm)]₆.

The Mₛ of FePt-Ag (5 nm) at Tₐ = 250°C was estimated to be 900 emu/cc as shown in Fig. 3 (b), which is well agree with the simple estimation mentioned above. The FePt-Ag (5 nm) exhibits higher Hₑ than that of [FePt-Ag (0.83 nm)]₆. This may be due to the higher ordering parameter in FePt-Ag (5 nm) as discussed in Fig. 1. However, the slope of the M-H loop near Hₑ, which is a good measure of the exchange coupling between the grains, for FePt-Ag (5 nm) is steeper than that for [FePt-Ag (0.83 nm)]₆. This is consistent with the difference of the microstructure between FePt-Ag (5 nm) and [FePt-Ag (0.83 nm)]₆, i.e., [FePt-Ag (0.83 nm)]₆ is considered to have well magnetically isolated grain structure, while FePt-Ag (5 nm) is regarded as a continuous film.

Compared with the [FePt-Ag (0.83 nm)]₆, [FePt-MgO (0.83 nm)]₆ exhibits a low perpendicular anisotropy; the easy axis of the magnetization lies in the film plane as shown in Fig. 3 (c). The low perpendicular anisotropy is considered to be related to the low ordering parameter S of 0.2. The Mₛ of [FePt-MgO (0.83 nm)]₆ was estimated to be 900 emu/cc, which agrees with the value (860 emu/cc) estimated for the mixture of L1₀ FePt and non-magnetic MgO.

Figure 4 (a) shows the dependence of the perpendicular anisotropy constant Kᵤ on the process temperature for [FePt-Ag (0.83 nm)]₆, FePt-Ag (5 nm), [FePt-MgO (0.83 nm)]₆, FePt-MgO (5 nm). For comparison, the Kᵤ of previously reported FePt/Ag multilayer deposition is shown in the figure. The uniaxial anisotropy was estimated as follows. If the sample magnetization was saturated by the external magnetic field (15 kOe), which is the maximum field of the torque magnetometer, the Kᵤ was estimated simply by the sum of the effective anisotropy Kₑ and the demagnetizing energy 2πMₑ² (5.1 x 10⁶ erg/cc for Mₑ = 900 emu/cc). However, some of the samples exhibit quite large anisotropy and the external field is not enough to rotate the magnetization from easy to hard axis. In such case (typically for Kₑ > 1 x 10⁷ erg/cc), the Kₑ at room temperature was estimated from the linear extrapolation of Kₑ at measured at high temperature. In Fig. 4 (b), the temperature dependence of the torque amplitude of FePt-Ag (5 nm) grown at Tₐ = 300 °C is shown as a example. From rotational hysteresis in the torque curve, it can be distinguished whether the external field of 15 kOe was enough to saturate the magnetization. In the case of Fig. 4 (b), the saturated Kₑ was obtained at temperatures > 200 °C. Thus, we estimated the Kₑ at room temperature by extrapolating...
In this paper, we presented the experimental results of the growth of well isolated FePt granular films with (001) orientation. We found that the [FePt-Ag (0.83 nm)]$_6$ fabricated by the repetition process at $T_a = 250{\degree}C$ has well isolated granular structure and exhibits a large uniaxial anisotropy of $1.0 \times 10^7$ erg/cc as well as high coercivity of > 5 kOe. From the experiment, it can be concluded that the Ag addition and applying a repetition process the deposition and subsequent annealing are effective to fabricate highly ordered and well isolated L1$_0$ FePt grains at a low process temperature.

4. Summary

In this paper, we presented the experimental results of the growth of well isolated FePt granular films with (001) orientation. We found that the [FePt-Ag (0.83 nm)]$_6$ fabricated by the repetition process at $T_a = 250{\degree}C$ has well isolated grain structure and exhibits a large uniaxial anisotropy of $1.0 \times 10^7$ erg/cc as well as high coercivity of > 5 kOe. From the experiment, it can be concluded that the Ag addition and applying a repetition process the deposition and subsequent annealing are effective to fabricate highly ordered and well isolated L1$_0$ FePt grains at a low process temperature. The process temperature of 250 °C reported here is much lower than those in the previous reports. However there are remaining issues to be addressed. One is why the Ag addition and the repetition process were effective. In order to discuss it in detail, further studies including transmission electron microscope (TEM) observation and compositional analysis by Auger depth profile will be necessary. Moreover, in this study, we used MBE method to fabricate the samples. For practical application, comparison between sputtering deposition and the present MBE growth should be discussed.

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