Enhancement in coercivity of Pulsed Laser Deposition-fabricated Fe-Pt thick film magnets by reducing droplets

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A high-speed Pulsed Laser Deposition method with the deposition rate of several ten microns per hour followed by a post-annealing process enabled us to obtain isotropic Fe-Pt thick film magnets with the $L1_0$ order phase. In the study, we investigated the relationship between droplets and coercivity of the post-annealing films. It was found that the reduction in the numbers of droplets enabled us to increase the coercivity although the average deposition rate decreased from 40 to 15 μm per hour. In the samples with R_a less than 0.5 μm , the coercivity exceeded 450 kA/m at the annealing temperature of 673 K. A SEM-EDX analysis of droplets revealed that they tended to have iron-rich or platinum-rich compositions compared with the average one of $\text{Fe}_{50}\text{Pt}_{50}$. © 2012 American Institute of Physics. [doi:10.1063/1.3679566]

I. INTRODUCTION

Although the high price of platinum is disadvantage in Fe-Pt magnet as an industrial material, the magnet thicker than several microns has been used by several researches who have focused on the outstanding biocompatibility.^{1,2} Watanabe *et al.*³ prepared rapidly-quenched isotropic Fe-Pt ribbons with the $(BH)_{\text{max}}$ of approximately 160 kJ/m³, and Makino *et al.*⁴ added Zr and B elements to enhance the coercivity of melt-spun isotropic Fe-Pt alloys up to approximately 650 kA/m. Givord *et al.*⁵ reported Fe-Pt multilayered foils with the coercivity higher than 800 kA/m by using a cold-deformation. In addition, sputtering-made isotropic Fe-Pt film magnets thicker than several microns were reported by Aoyama⁶ and Liu,⁷ respectively.

In the paper, isotropic Fe-Pt thick films prepared by using a high-speed Pulsed Laser Deposition (PLD) method with the deposition rate of several-ten-microns per hour.⁸ It is generally said that micrometer-sized droplets are ejected as a whole from a target during a PLD deposition process. In this study, two types of films with different numbers of droplets were fabricated by taking advantage of controlling the focus distance from an optical lens to a target together with the distance between a target and a substrate. This contribution reports that the reduction in the numbers of droplets is effective to increase the coercivity value for the PLD-made isotropic Fe-Pt thick film magnets.

II. EXPERIMENTAL PROCEDURE

A Fe-Pt target was ablated with a Nd-YAG pulse laser (wave length: 355 nm) at the repetition rate of 30 Hz in a vacuum atmosphere. A laser power was measured with a power meter in front of the entrance lens of a chamber. Before the ablation, the chamber was evacuated down to approximately 4×10^{-7} Torr with a rotary pump together

with a molecular turbo pump. The distance between a target and a Ta substrate (T-S distance) varied from 10 to 20 mm, and the area of all the obtained films were $5 \times 5 \text{ mm}^2$. The numbers of droplets in each film were controlled by changing the T-S distance and a DF (Defocus) rate⁹ which was the focus distance between an optical lens and a target. The T-S distance, DF rate and the compositions of targets enabled us to vary the ratio of Fe to Pt elements in each film. Average deposition rate exceeded 10 $\mu\text{m}/\text{h}$, which was almost as twice as that of a sputtering method.⁶

(a). $R_a = 0.44 \mu\text{m}$

(b). $R_a = 1.1 \mu\text{m}$

FIG. 1. Surface observations of two samples with the R_a of 0.44 and 1.1 μm , respectively. A lot of droplets could be observed in the film with the R_a of 1.10 μm .

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FIG. 2. Coercivity of samples with the Ra less than $0.5 \mu\text{m}$ and larger than $1.0 \mu\text{m}$, respectively, as a function of post-annealing temperature. At the temperature of 673 K, the value became higher than 400 kA/m in a sample with Ra less than $0.5 \mu\text{m}$.

All the as-deposited film did not have hard magnetic properties because of the existence of the disorder phase, therefore the post-annealing was carried out to obtain the $L1_0$ order phase. In the annealing process, the rate of temperature rise and holding time were 50 K/min and 10 min, respectively, under the range of temperature from 623 to 773 K. After an annealed-sample was magnetized up to 9 T with a pulse magnetizer, M-H loops were measured with a vibrating sample magnetometer (VSM) which could apply a magnetic field up to approximately 1800 kA/m reversibly. In the experiment, in-plane magnetic properties were only shown because all the films had isotropic magnetic properties. An average thickness was measured with a micrometer. In addition,

FIG. 3. Compositions of droplets for a $\text{Fe}_{50}\text{Pt}_{50}$ film were evaluated by using a line analysis in SEM-EDX. Droplets with Fe-rich or Pt-rich composition compared to $\text{Fe}_{50}\text{Pt}_{50}$ could be observed.

FIG. 4. $(BH)_{\text{max}}$ values as a function of Fe compositions in each sample. The samples were prepared by using 3 T-S distances of 10, 15, and 20 mm, respectively.

estimation in the difference of the numbers of droplets was carried out by measuring the centerline average roughness, Ra.

III. RESULTS AND DISCUSSION

In order to examine the relationship between the numbers of droplets and magnetic properties, two types of films with the different Ra values were prepared by controlling the DF rate and T-S distance. Figure 1 shows the surface views of samples with two Ra values of 0.44 and $1.10 \mu\text{m}$, respectively. A lot of droplets could be observed in the film with the Ra of $1.10 \mu\text{m}$. (see Fig. 1 (b)) Coercivity values of samples with two kinds of Ra as a function of post annealing temperatures were displayed in Fig. 2. Here, the compositions and thicknesses of all films were kept at approximately $\text{Fe}_{50}\text{Pt}_{50}$ and thicker than $10 \mu\text{m}$, respectively. At each temperature, the coercivity values of samples with Ra less than $0.5 \mu\text{m}$ were higher than those of Ra larger than $1.0 \mu\text{m}$. In particular, reduction in the numbers of droplets enabled us to obtain samples with the coercivity higher than 400 kA/m at relatively low temperature of 673 K. An evaluation on compositions of droplets for a $\text{Fe}_{50}\text{Pt}_{50}$ film was carried out by using a line analysis as shown in Fig. 3. In Fig. 3(a), a

FIG. 5. $(BH)_{\text{max}}$ values as a function of Ra in each sample. The samples were prepared by using 3 T-S distances of 10, 15, and 20 mm, respectively. As the value of Ra decreased, $(BH)_{\text{max}}$ increased.

FIG. 6. M-H loop of a Fe-Pt thick film magnet with the Ra of approximately $0.16 \mu\text{m}$. The values of coercivity, remanence and $(BH)_{\text{max}}$ were 424 kA/m, 1.0 T, and 115 kJ/m^3 , respectively.

droplet with Fe-rich composition compared to $\text{Fe}_{50}\text{Pt}_{50}$ could be observed. In other droplets, Pt-rich compositions were also observed as shown in Fig. 3(b). The deterioration of magnetic properties due to the increase in the numbers of droplets is considered to be the reduction in the magnetic anisotropy of each film.

Figures 4 and 5 show $(BH)_{\text{max}}$ as a function of Fe compositions and Ra of each sample prepared by 3 T-S distances of 10, 15 and 20 mm, respectively. In the range of Fe compositions from 48 to 58 at %, the $(BH)_{\text{max}}$ values were independent on the composition. On the other hand, the values of $(BH)_{\text{max}}$ decreased with the reduction in Ra. These results suggest that the magnetic properties of Fe-Pt thick film magnets strongly depend on the numbers of droplets. Resultantly,

a Fe-Pt thick film magnet with the Ra of approximately $0.16 \mu\text{m}$ had the values of coercivity, remanence and $(BH)_{\text{max}}$ of 424 kA/m, 1.0 T, and 115 kJ/m^3 , respectively, which were comparable to ones of sputtering-made Fe-Pt thick film magnets^{6,7} (see Fig. 6).

IV. CONCLUSION

In this study, the relationship between the droplets and magnetic properties was investigated in PLD-fabricated Fe-Pt thick film magnets. It was found that deterioration of coercivity due to the increase in the numbers of droplets occurs, which is considered to be attributed to the existence of Fe-Rich or Pt-rich droplets compared with the $\text{Fe}_{50}\text{Pt}_{50}$ film composition. By taking advantage of the reduction in the numbers of droplets, a Fe-Pt thick film magnet with the $(BH)_{\text{max}}$ of 115 kJ/m^3 could be obtained under the high deposition rate of $12 \mu\text{m/h}$.

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