

# Effect of Preparation Condition on Magnetic Properties of Nd-Fe-B/ $\alpha$ -Fe Multi-layered Thick Film-Magnets Prepared by PLD Method

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Nd-Fe-B/ $\alpha$ -Fe multi-layered thick film-magnets were synthesized by PLD method from a Nd<sub>2.4</sub>Fe<sub>14</sub>B/ $\alpha$ -Fe composite target. In order to improve magnetic properties and their reproducibility, effects of the stacking period of Nd-Fe-B/ $\alpha$ -Fe layers,  $t_p$ , the target-substrate distance,  $D_{TS}$ , and the laser beam energy on a target,  $E_L$ , on them were studied. The reproducibility of magnetic properties was improved significantly by decreasing  $L_E$ . In particular, the standard deviation of the Nd content was suppressed within 3%, when  $L_E$  was reduced from a value higher than 200 mJ/mm<sup>2</sup> to 25 mJ/mm<sup>2</sup>. An increase in  $D_{TS}$  and selection of a suitable  $t_p$  value improved the maximum energy product  $(BH)_{max}$ . The largest  $(BH)_{max}$  value obtained in this investigation was 112 kJ/m<sup>3</sup>, which is higher than the values reported previously for isotropic film-magnets thicker than 10  $\mu$ m. Origins of superior magnetic properties were discussed from the viewpoints of suitable interlayer exchange interaction between Nd-Fe-B and  $\alpha$ -Fe layers and morphology of prepared film-magnets.

**Index Terms**—Layered structure, nanocomposite magnet, Nd-Fe-B, PLD method, thick film-magnet.

## I. INTRODUCTION

Thick film-magnets are hopeful candidates for magnets used in small motors [1]. Although excellent magnetic properties have been reported for anisotropic thick film-magnets [2]-[4], isotropic ones are also attractive, because they are easily magnetized multi-polarly, and the multi-polar magnetization increases output torque of a motor. We have already reported a design example of a small motor consisting of isotropic film-magnets which has output torque larger than that of a motor consisting of an anisotropic magnet [5]. An increase in the remanence  $I_r$  is expected to improve characteristics of small motors using a magnet-rotor with multi-polar pairs. As the remanence  $I_r$  of isotropic thick film-magnets is lower than that of anisotropic ones [6]-[9], we prepared Nd-Fe-B/ $\alpha$ -Fe multi-layered nanocomposite film-magnets by the pulse laser deposition (PLD) method. The prepared film-magnets were composed of approximately 800 Nd-Fe-B/ $\alpha$ -Fe layers, and exhibited  $I_r$  of 1 T and the coercivity  $H_c$  of 430 kA/m [10]. However, the obtained properties scattered from a sample to a sample, and the improvement in reproducibility of magnetic properties is strongly required for applying them to a small motor.

On the other hand, we have already reported that the control of the laser beam conditions affects the reproducibility of magnetic properties of Nd-Fe-B thick film-magnets, and have indicated that the repeatability of the magnetic properties can be improved by controlling the energy density of laser beam on a target [11]. Therefore, we applied the method developed in the previous investigation [11] to Nd-Fe-B/ $\alpha$ -Fe multi-layered nanocomposite film-magnets, and investigated effects of the stacking period of Nd-Fe-B/ $\alpha$ -F layers,  $t_p$ , the target-

substrate distance,  $D_{TS}$ , and the laser beam energy on a target,  $E_L$ , on magnetic properties. Consequently, the magnetic properties as well as their reproducibility were improved by increasing  $D_{TS}$  and decreasing  $L_E$  as well as selecting a suitable  $t_p$ .

## II. EXPERIMENTAL PROCEDURE

A composite target, which was composed of Nd<sub>2.4</sub>Fe<sub>14</sub>B and  $\alpha$ -Fe segments, was prepared and the area fraction of the  $\alpha$ -Fe segment was set to 1/4 in this investigation. Subsequently, the target was ablated with a Nd-YAG pulse laser ( $\lambda = 355$  nm) with the laser power of 5 W at the repetition rate of 30 Hz in a vacuum chamber with the back pressure of approximately 10<sup>-5</sup> Pa. During the ablation, the target was rotated at the speed of 3.3 – 13 rpm and a multi-layered film was deposited on Ta substrate. The deposited films are 12–30  $\mu$ m in thickness. The layer numbers synthesized in one hour are deduced from the above rotation speeds as 396 and 1560, respectively. The  $D_{TS}$  values were set at 10 and 20 mm. The energy density of laser,  $L_E$ , was varied by changing the defocusing rate,  $DFR$ , defined by  $(TD - FD)/FD$ , where  $TD$  is the distance between the condensing lens and the target, and  $FD$  is the focal length.  $DFR$  values were set at 0 and 0.3 in this investigation, which correspond to  $L_E > 200$  and  $L_E \cong 25$  mJ/mm<sup>2</sup>.

As-deposited films were amorphous, and were crystallized by a pulse annealing for 1–2 s in an infrared furnace with output power of 8 kW. Magnetic properties of films were measured with a vibrating sample magnetometer after magnetization under a pulsed magnetic field of 6.4 MA/m. As all post-annealed films were isotropic, in-plane magnetic properties are shown in this article. The thickness of a film was determined by a micrometer. Morphology of film-magnets was analyzed with a scanning electron microscope (SEM), and their compositions were analyzed with an energy dispersive X-ray spectrometer (EDS). In the analysis, the B

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TABLE I

Standard deviations of Nd content and  $(BH)_{\max}$  for film-magnets synthesized at  $L_E = 25 \text{ mJ/m}^2$ , together with those for  $L_E > 200 \text{ mJ/m}^2$ . The standard deviation values are reduced by average value of the corresponding quantities.  $D_{TS}$  was set at 10 mm

Properties	$E_L$ ( $\text{mJ/m}^2$ )	Target rotation speed [rpm]			
		3.3	6.5	9.7	13
Nd Content	> 200	—	13.7%	—	—
	25	3.0%	2.3%	1.8%	7.7%
$(BH)_{\max}$	> 200	—	19.2%	—	—
	25	12.8%	10.9%	7.5%	14.3%

content was omitted. The roughness of the surface of a film was evaluated with a surface roughness meter with a contact probe. Five scans were carried out for one sample at different places and the average of five samples was shown in this manuscript.

### III. RESULTS

#### A. Effect of Energy Density of Laser Beam

As mentioned in ‘‘Introduction’’, a reduction in the energy density of laser beam is expected to improve the reproducibility of magnetic properties of prepared films. Thus, we evaluated the standard deviation of the Nd content and  $(BH)_{\max}$  for samples deposited with  $L_E$  values of 25 and  $>200 \text{ mJ/m}^2$ .  $L_E$  was varied by changing  $DFR$  as explained previously, and the  $L_E$  values of 25 and  $>200 \text{ mJ/m}^2$  correspond to the  $DFR$  values of 0.3 and 0, respectively. It should be noted that a decrease in  $L_E$  does not decrease the deposition rate, because total energy of the laser beam does not vary by changing  $DFR$  [11],

The obtained results are shown in TABLE I for the samples with  $D_{TS}=10 \text{ mm}$ . The standard deviation values shown in the table are reduced by the average values of the corresponding quantities. The both standard deviations of the Nd content and  $(BH)_{\max}$  synthesized at  $L_E=25 \text{ mJ/m}^2$  is less than those for  $L_E > 200 \text{ mJ/m}^2$ , which suggests that the reproducibility of magnetic properties of film-magnets was improved by controlling  $L_E$ . In particular, the scattering of the Nd content was suppressed by adopting the ablation condition of  $L_E=25 \text{ mJ/m}^2$ .

#### B. Effect of Target-Substrate Distance and Stacking Period of Nd-Fe-B/ $\alpha$ -Fe layers

Based on the results shown in TABLE I, we set  $L_E$  at  $25 \text{ mJ/m}^2$ , and the target-substrate distance,  $D_{TS}$ , and the thickness of the stacking period of Nd-Fe-B/ $\alpha$ -Fe layers,  $t_p$ , were varied.

The Nd content,  $\text{Nd}/(\text{Nd}+\text{Fe})$ , is plotted in Fig.1 as a function of  $t_p$  deduced from the film thickness and the rotation speed of the target. The Nd content does not depend on  $t_p$  except  $t_p < 10 \text{ nm}$  for  $D_{TS}$  of 20 mm where the Nd content is slightly low compared with those of  $t_p > 10 \text{ nm}$ . The Nd contents for the  $D_{TS}$  values of 10 and 20 mm were approximately 11.6 and 10.5 at.%, respectively. These values suggest that the prepared films are Nd-Fe-B/ $\alpha$ -Fe

composites, because the Nd contents of the films are lower than that of the  $\text{Nd}_{2.4}\text{Fe}_{14}\text{B}$  segment (14.6 at.%). It should be also noted that the Nd contents of the films is lower than the stoichiometric Nd content of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (12.5 at.%),

The coercivity  $H_c$  and the remanence  $I_r$  after crystallization are shown in Fig.2 as a function of  $t_p$ .  $H_c$  increased with decreasing  $t_p$  except  $t_p < 10 \text{ nm}$ , although dependence of  $I_r$  is not so significant. This result is consistent with the results by computer simulation for nanocomposite magnets [12], because a reduction in thickness of  $\alpha$ -Fe prevents magnetization reversal of  $\alpha$ -Fe. In addition,  $H_c$  and  $I_r$  were affected by  $D_{TS}$ . The films synthesized at  $D_{TS}$  of 20 mm tended to have small  $H_c$  and large  $I_r$  values compared to those synthesized at  $D_{TS}$  of 10 mm. This tendency can be partially explained by small Nd contents of the films synthesized at  $D_{TS}$  of 20 mm. Details are discussed in Chapter III.

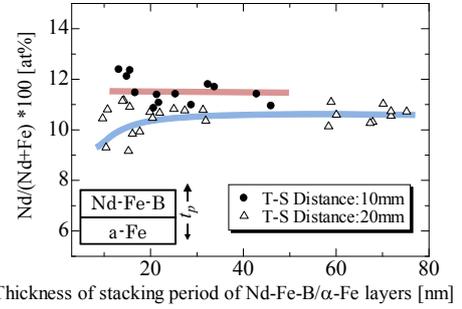
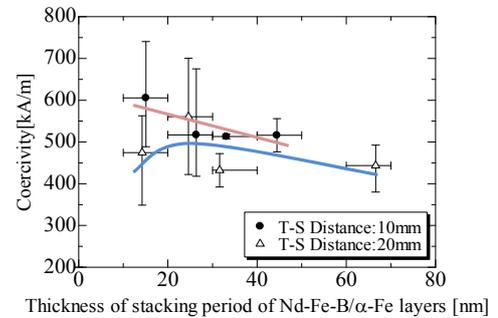
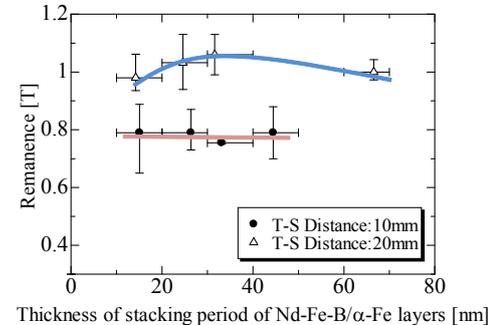


Fig.1 Nd contents as a function of thickness,  $t_p$ , of stacking period of Nd-Fe-B/ $\alpha$ -Fe layers deduced from film thickness and rotation speed of target. The B content was omitted from the analysis.



(a) Coercivity



(b) Remanence

Fig.2 Coercivity and remanence after crystallization as a function of stacking period of Nd-Fe-B/ $\alpha$ -Fe layers,  $t_p$ , deduced from film thickness and rotation speed of target. The closed circles and open triangles indicate the values averaged in every 10 nm, and the error bars shows the maximum and minimum values in the corresponding region.

TABLE II  
Magnetic properties of isotropic thick RE-TM thick film-magnets

Alloy	Thickness ( $\mu\text{m}$ )	$H_c$ (kA/m)	$M_r$ (T)	$(BH)_{\text{max}}$ ( $\text{kJ/m}^3$ )	Ref.
Nd-Fe-B	11.8	504	1.1	112	This Work
Nd-Fe-B	14.9	430	1.0	90	10
Nd-Fe-B	500	1380	0.59	61	9
Nd-Fe-B	300	760	0.45		8
Nd-Fe-B	10-50	860	0.5		7
Sm-Co	30	1200	0.75	90	14
Sm-Fe-N	$\sim 45$	1440	0.55		15

The maximum energy product  $(BH)_{\text{max}}$  is shown in Fig.3 as a function of  $t_p$ .  $(BH)_{\text{max}}$  of the film-magnets synthesized at  $D_{\text{TS}}$  of 20 mm is higher than those at  $D_{\text{TS}}$  of 10mm. The large  $(BH)_{\text{max}}$  values were obtained for  $t_p$  of around 20-30 nm, and exceeded  $90 \text{ kJ/m}^3$ . The above stacking period for obtaining a large  $(BH)_{\text{max}}$  value is reasonable from the viewpoint of a suitable nanostructure predicted by computer simulation [13]. The large  $I_r$  values of the film-magnets synthesized at  $D_{\text{TS}}$  of 20 mm are responsible to the obtained large  $(BH)_{\text{max}}$  values. The values of  $I_r$ ,  $H_c$  and  $(BH)_{\text{max}}$  obtained were 1.1 T, 504 kA/m and  $112 \text{ kJ/m}^3$ , respectively.

Typical magnetic properties of isotropic film-magnets thicker than  $10 \mu\text{m}$  are shown in TABLE II. As far as we know, the  $(BH)_{\text{max}}$  value obtained in this investigation is the largest one in isotropic film-magnets thicker than  $10 \mu\text{m}$ .

#### IV. DISCUSSION

The large  $(BH)_{\text{max}}$  values for the films synthesized at  $D_{\text{TS}}$  of 20 mm can be partially attributed to their large  $I_r$  values due to their large Fe content (small Nd content) as shown in Fig.1 and Fig.2 (b), because an increase in Fe content has a tendency of increasing  $I_r$  and decreasing  $H_c$ . However, another origin would be also responsible to their large  $I_r$  values, because a reduction in  $H_c$  is not so significant as shown in Fig.2 (b). As shown in Fig.4,  $I_r$  of film-magnets synthesized at  $D_{\text{TS}}$  of 20 mm is larger than that at  $D_{\text{TS}}$  of 10 mm with the same  $H_c$  value. This tendency would be explained by the remanence

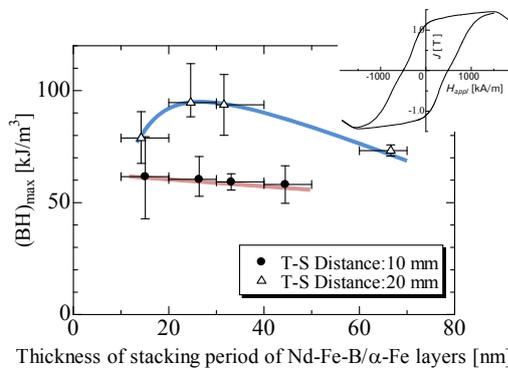


Fig.3 Maximum energy product,  $(BH)_{\text{max}}$ , as a function of stacking period,  $t_p$ , of Nd-Fe-B/ $\alpha$ -Fe layers. The closed circles and open triangles indicate the values averaged in every 10 nm, and the error bars shows to the maximum and minimum values in the corresponding region. The inset show the hysteresis curve of the film with the largest  $(BH)_{\text{max}}$  value.

enhancement in film-magnets synthesized at  $D_{\text{TS}}$  of 20 mm, because a micromagnetic simulation predicts that an increase in intergain exchange interaction can increase  $I_r$  without a significant decrease in  $H_c$  for nanocomposite magnets composed of 75%  $\text{Nd}_2\text{Fe}_{14}\text{B}$  and 25%  $\alpha$ -Fe [12]. Actually, as shown by SEM images in Fig.5, the increase in  $D_{\text{TS}}$  reduces the size of droplets which disturb a periodic layer structure and resultantly suppress effective interlayer exchange interaction between Nd-Fe-B and  $\alpha$ -Fe layers. In order to evaluate the number and size of droplets, we measured the arithmetic mean deviation of the profile,  $R_a$ , of surfaces of film-magnets.  $R_a$  was reduced from 1.8 to  $1.4 \mu\text{m}$  by increasing  $D_{\text{TS}}$  from 10 to 20 mm, which is consistent with the result of SEM observation.

#### V. CONCLUSION

Nd-Fe-B/ $\alpha$ -Fe multi-layered thick film-magnets were synthesized by PLD method from a  $\text{Nd}_{2.4}\text{Fe}_{14}\text{B}/\alpha$ -Fe composite target with varying the energy density of laser beam,  $L_E$ , the target-substrate distance,  $D_{\text{TS}}$ , and the stacking period of Nd-Fe-B/ $\alpha$ -Fe layers (the rotation speed of the target),  $t_p$ . Main results are summarized as follow;

The reproducibility of magnetic properties is improved by changing  $L_E$  from a value higher than  $200 \text{ mJ/m}^2$  to  $25 \text{ mJ/m}^2$ . In particular, the standard deviation of the Nd content was suppressed within 3% for all the rotation speed of the target. An increase in  $D_{\text{TS}}$  improves magnetic properties and this tendency can be attributed to an increase in remanence due to increase in Fe content and decrease in the number and/or size of droplets. Superior magnetic properties are obtained, when the thickness of the stacking period of Nd-Fe-B/ $\alpha$ -Fe layers is controlled to 20 – 30 nm. This stacking period for obtaining

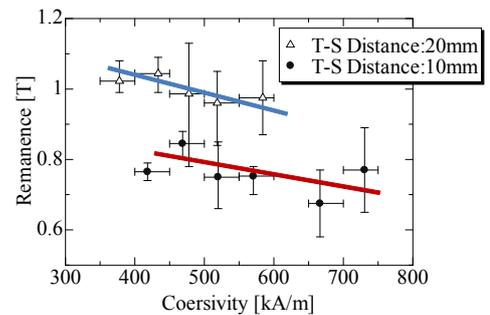


Fig.4 Relationship between coercivity and remanence. The closed circles and open triangles indicate the values averaged in every 10 nm, and the error bars shows to the maximum and minimum values in the corresponding region.

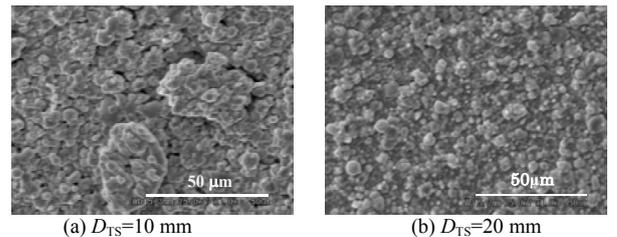


Fig.5 Typical SEM images of surface of a film-magnet. The observed films were synthesized at  $D_{\text{TS}}$  of (a) 10 mm and (b) 20 mm.

superior magnetic properties is consistent with the previous prediction by computer simulation. The values of  $I_r$ ,  $H_c$  and  $(BH)_{\max}$  obtained in this investigation were 1.1 T, 504 kA/m and 112 kJ/m<sup>3</sup>, respectively. The obtained  $(BH)_{\max}$  value is higher than the values reported previously for isotropic film-magnets thicker than 10  $\mu\text{m}$ .

#### ACKNOWLEDGMENT

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