# Magnetic properties of Nd-Fe-B/ $\alpha$ -Fe multi-layered thick film magnets

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**Abstract.** Nd-Fe-B/$\alpha$-Fe multi-layered nanocomposite film-magnets were prepared from a rotating $Nd_{2.6}Fe_{14}B$/$\alpha$-Fe composite target by the pulse laser deposition method with a Nd-YAG laser ($\lambda$=355 nm). The prepared film-magnets were composed of approximately 800 layers with the thickness of about several tens nanometre, and exhibited hard magnetic properties after annealing. The laser power for the ablation of the target was varied between 3 and 7 W, and it was found that the composition, magnetic properties, and the surface roughness of the prepared films depend on the laser power. A reduction in the laser power caused increases in remanence and the maximum energy product $(BH)_{max}$ as well as decreases in Nd content and coercivity of the films. All the samples had Nd-poor compositions compared with that of the $Nd_{2.6}Fe_{14}B$ target and the Nd-contents of the films deposited with laser power of 3 W were less than that of the stoichiometric composition of $Nd_2Fe_{14}B$. The highest $(BH)_{max}$ and remanence values were obtained for a film deposited with laser power of 3 W, and were 90 kJ/m$^3$ and 1.0 T, respectively. The obtained high remanence value is discussed from the viewpoint of the remanence enhancement as well as the large saturation magnetization of $\alpha$-Fe .

## 1. Introduction

Recently, magnets with several tens of microns are needed for applications to small machines such as milli-size motors [1]. As thinning of a bulk magnet has a tendency of degrading its magnetic properties [2], thick film-magnets are hopeful candidates for magnets used in such applications [3]. Although excellent magnetic properties have been reported for anisotropic thick film-magnets [4]-[6], 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 isotropic film-magnets which has output torque larger than that of a motor consisting of an anisotropic magnet [7].

Nanocomposite magnets are expected to exhibit isotropic and superior magnetic properties by controlling their nanostructures [8], [9]. One of promising methods of controlling nanostructures is

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synthesis of an artificial layered structure, because the layered structure prevents the growth of grains and achieves a fine nanostructure [10], [11].

In order to synthesis multi-layered film-magnets with several tens microns, we need to prepare $10^3$-$10^4$ layers. The PLD method with a composite target is suitable to prepare the layered structure for this purpose, and we reported the results of evaluation of magnetic properties of Nd-Fe-B/$Fe_3B$ multi-layered films prepared by the PLD (Pulsed Laser Deposition) method [11]. In this contribution, we prepared Nd-Fe-B/$\alpha$-Fe nanocomposite film-magnets using a Nd-YAG laser with different laser powers, and evaluated their magnetic properties and morphology. Subsequently, the obtained magnetic properties were discussed in terms of the remanence enhancement.

## 2. Experimental Procedure

### *2.1 Preparation of multi-layered films*

A composite target, which were composed of $Nd_{2.6}Fe_{14}B$ and $\alpha$-Fe segments, were prepared and the area fraction of the $\alpha$-Fe segment was set to 1/4 in this investigation. Subsequently, the target was ablated by a Nd-YAG laser beam ($\lambda$=355 nm) in a vacuum chamber for 1 hour under the vacuum of $5\times10^{-7}$-$2\times10^{-6}$ Torr. As shown schematically in Fig.1, the target was rotated with the speed of 6.5-7.0 rpm during the deposition, which enabled us to deposit 13-14 layers/min. Thus, the deposited films were composed of approximately 800 layers. The repetition frequency of the laser and the distance between a target and a substrate were set to 30 Hz and 10 mm, respectively. The laser power was varied between 3 and 7 W.

We used a Ta substrate and composite films with the thickness of 8.1–28.4 $\mu$m were obtained. The Nd-Fe-B layers in an as-deposited film were amorphous after deposition, and the as-deposited films were crystallized and hardened magnetically by pulse-annealing for 1.8-2.0 s with an infrared furnace.

### *2.2 Evaluation of magnetic properties and microstructures*

The prepared films were isotropic magnetically, and in-plane magnetic properties were measured with a vibrating sample magnetometer after magnetization under a pulse field of 6.4 MA/m. The thickness of a film was determined from the in-plane and perpendicular magnetization curves measured for the corresponding film in the as-deposited [12].

Microstructures of film-magnets were analyzed after crystallization with SEM, TEM, and XRD. The composition of a film was determined with an energy dispersive X-ray spectrometer (EDX). The roughness of the surface of a film was evaluated with a surface roughness meter with a contact probe. The average roughness $R_a$ was calculated from the results of 3-5 scans (2.4 mm in length) with the probe for as-deposited films.

## 3. Results and discussion

Multi-layered films were deposited with the laser powers of 3, 5, and 7 W, and their microstructures and magnetic properties were evaluated after magnetic hardening. Results of X-ray diffraction analysis and TEM observation are shown n Fig.2 for a film deposited with the laser power of 5W. The multi-layer structure, approximately 20 nm in thickness, was clearly seen in Fig.2 (a). This thickness

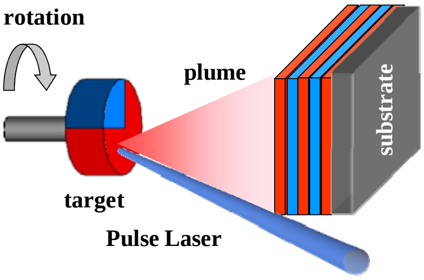
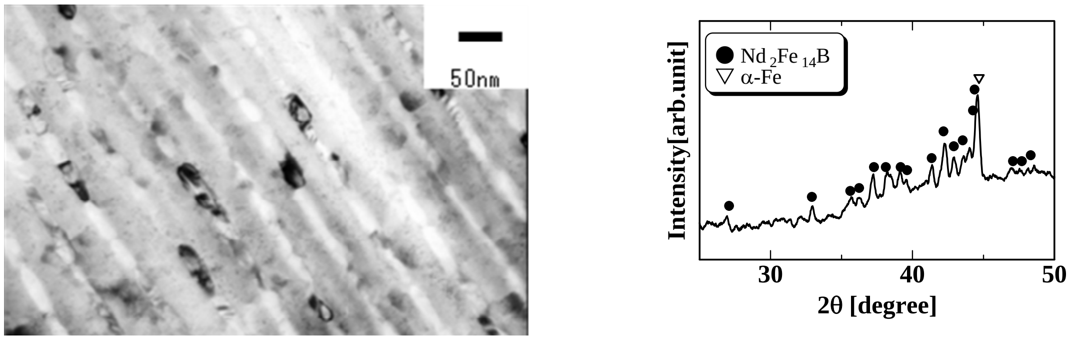


**Figure 1.** Schematic representation of PLD method for preparing multi-layered nano-composite film.



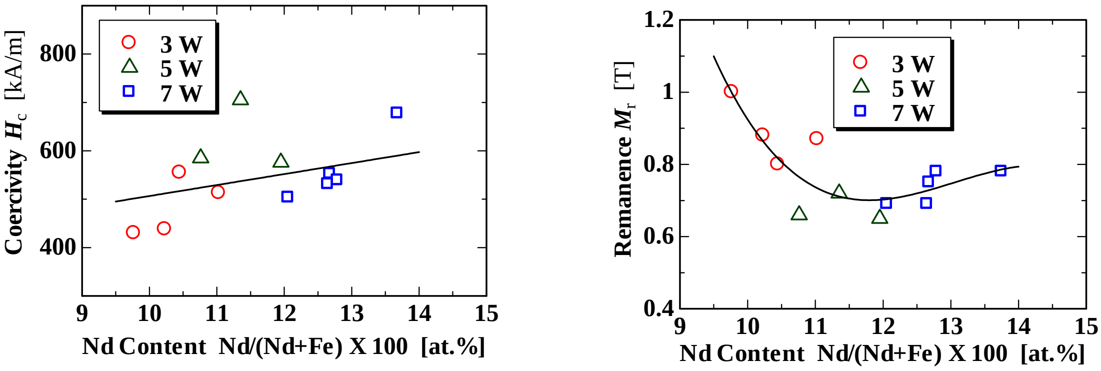
(a) (b)

**Figure 2.** TEM micrograph (a) and X-ray diffraction pattern (b) of Nd-Fe-B/α-Fe multi-layered film-magnet deposited with laser power of 5W.

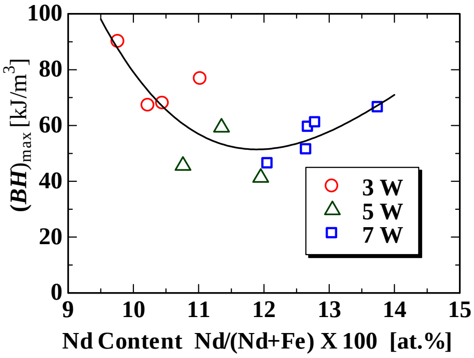
is suitable for obtaining a nanocomposite magnet [8], [9]. The X-ray diffraction pattern indicates that the film is composed of $Nd_2Fe_{14}B$ and α-Fe mainly and that the $Nd_2Fe_{14}B$ grains have no preferred orientations. The diffraction pattern obtained by TEM were also supported these results.

The compositions of the prepared films were scattered from a film to a film, and the obtained coercivity $H_c$, remanence $M_r$, and maximum energy product $(BH)_{max}$ after crystallization are shown in Fig.3 as a function of the content of Nd, Nd/(Nd+Fe). As the Nd contents of $Nd_{2.6}Fe_{14}B$ and $Nd_2Fe_{14}B$ are 0.156 and 0.125, respectively, all the samples shown in the figure have Nd-poor compositions compared with that of $Nd_{2.6}Fe_{14}B$. It is also seen that a small laser power results in a poor Nd composition, and the samples deposited with the laser power of 3 W were Nd-poor compared with the stoichiometric composition of $Nd_2Fe_{14}B$. Origins of the scattering and shift of Nd-content are not clear at the present stage.

The coercivity $H_c$ decreased with decreasing the Nd content as expected from magnetically soft



(a) Coercivity $H_c$ (b) Remanence $M_r$



(c) Maximum energy product $(BH)_{max}$

**Figure 3.** Coercivity $H_c$, remanence $M_r$, and maximum energy product $(BH)_{max}$ after crystallization as a function of the content of Nd, Nd/(Nd+Fe). The samples were deposited with the laser powers of 3, 5, and 7 W.

Table 1 Magnetic properties of isotropic thick RE-TM thick film-magnets.

| Alloy | Method | Film Thickness (μm) | $H_c$ (kA/m) | $M_r$ (T) | $(BH)_{max}$ (kJ/m$^3$) | Ref. |
|---|---|---|---|---|---|---|
| Nd-Fe-B | PLD | 14.9 | 430 | <u>1.0</u> | <u>90</u> | <u>This Work</u> |
| Nd-Fe-B | Plasma Spray | 500 | 1380 | 0.59 | 61 | 13 |
| Nd-Fe-B | Tape Casting | 300 | 760 | 0.45 |  | 14 |
| Nd-Fe-B | Screen Printing | 10-50 | 860 | 0.5 |  | 15 |
| Sm-Co | Sputtering | 30 | 1200 | 0.75 | 90 | 16 |
| Sm-Fe-N | Aerosol Deposition | ~45 | 1440 | 0.55 |  | 17 |

properties of α-Fe. The remanence has a tendency of increasing with a decrease in the Nd content as expected from the small saturation magnetization of $Nd_2Fe_{14}B$, although a broad minimum was seen at around N/(Nd+Fe) = 0.12, which suggests that the squareness of a demagnetization curve is not good at this content. The maximum energy product $(BH)_{max}$ also showed a broad minimum at around N/(Nd+Fe) = 0.12, reflecting the behaviour of the remanence. The largest $(BH)_{max}$ value was obtained for the film with N/(Nd+Fe) = 0.098, and the hysteresis loop of this film is shown in Fig.4. The average layer-thickness calculated from its thickness was 17.7 nm. The obtained $(BH)_{max}$ and $M_r$, 90 kJ/m$^3$ and 1.0 T respectively, are larger than those of the Nd-Fe-B/$Fe_3B$ multi-layered film reported previously [11]. Magnetic properties for other RE-TM isotropic film-magnets are shown in Table 1. The film obtained in this study has the largest $M_r$ and $(BH)_{max}$ values in those shown in the table.

In order to study origins of a large $M_r$ value of the films deposited with the laser power of 3 W, we evaluated the spring back ratio *SBR* which is the ratio of the recoiled magnetization from the coercive field to the remance, and found that the *SBR* values (0.46-0.60) of the films deposited with laser power of 3 W were larger than those (0.38-0.46) deposited with the laser power of 5 W. This result suggests strong intergrain exchange interactions in the former films. As a decrease in laser power results in reduction of film-thickness, the average layer-thickness is also reduced with decreasing the laser power. Therefore the intergrain exchange interaction and, resultantly, the remanence would be enhanced [8], [9].

It is well known that small particles called “droplet” are emitted from a target during deposition, and they would break the layered structure of deposited films. From this point of view, the surface roughness was evaluated for films deposited from $Nd_{2.6}Fe_{14}B$ target.. The results are shown in Fig.5 as a function of the film-thickness. It is clearly seen that the reduction in the laser power decreases not

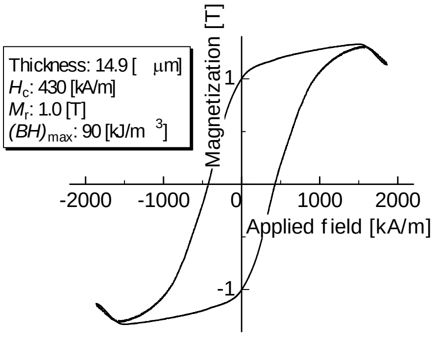


**Figure 4.** Hysteresis loop of Nd-Fe-B/α-Fe multi-layered film-magnet deposited with laser power of 3W.

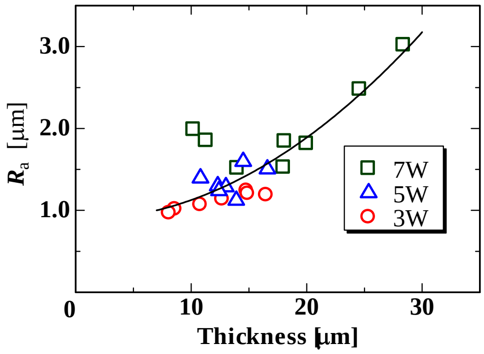


**Figure 5.** Average surface roughness of films of films deposited from $Nd_{2.6}Fe_{14}B$ target. The samples were deposited with laser powers of 3, 5, and 7 W.

only the film-thickness but also the average roughness $R_a$, which suggests that the reduction in the laser power decreases the number/or size of droplets and improves the layered structure. Thus, an improvement in the surface roughness seems to enhance the remanence through the intergrain exchange interaction.

## 4. Conclusions

Nd-Fe-B/α-Fe multi-layered nanocomposite film-magnets were prepared from a rotating $Nd_{2.6}Fe_{14}B$/α-Fe composite target by the pulse laser deposition method with a Nd-YAG laser (λ=355 nm). The laser power for ablation was varied between 3 and 7 W, and the rotating speed of a target was set so as to deposit approximately 800 layers with the thickness of about several tens nanometre. The synthesis of the layered structure was confirmed by TEM observation.

It was found that the composition, magnetic properties, and the surface roughness of the prepared films depend on the laser power. A reduction in the laser power caused increases in remanence and the maximum energy product $(BH)_{\max}$ as well as decreases in Nd content and coercivity. All the films had Nd-poor compositions compared with that of the $Nd_{2.6}Fe_{14}B$ target and the Nd-contents of the films deposited with laser power of 3 W were less than that of the stoichiometric composition of $Nd_2Fe_{14}B$. The highest $(BH)_{\max}$ and remanence values were obtained for a film deposited with laser power of 3 W, and were 90 kJ/$m^3$ and 1.0 T, respectively.

The spring back ratio of the films deposited with laser power of 3 W was larger than that with laser power of 5 W, which suggests strong intergrain exchange interaction. As a reduction in the laser power causes the decrease in the layer-thickness and the number/size of droplets, the enhancement of exchange interaction due to the reduction in laser power seems to be reasonable. Therefore, the large remanence and resultant large $(BH)_{\max}$ obtained in this investigation can be attributed to the large saturation magnetization of α-Fe and the remanence enhancement due to intergrain exchange interaction.

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