

Review of Fabrication and Characterization of Nd-Fe-B Thick Films for Magnetic Micromachines

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Isotropic Nd-Fe-B thick film magnets were prepared by a high-speed pulsed laser deposition method followed by a post annealing. The deposition rate of 90 $\mu\text{m}/\text{h}$ could be successfully achieved, and a pulse annealing was adopted as the post annealing process in order to enhance coercivity. Use of a substrate heating system under the high deposition rate enabled us to obtain anisotropic thick films with $(BH)_{\text{max}}$ of approximately 120 kJ/m^3 , which show the potential for an improvement in the properties of the micromachines. Novel micromachines comprising the isotropic films were introduced.

Index Terms—Laser ablation, micromachines, micromotors, thick film devices.

I. INTRODUCTION

NUMEROUS studies [1], [2] on microactuators and micromachines comprising small permanent magnets indicate that reduction in thickness for bulk magnets without deterioration of magnetic properties [3]–[5] and preparation of thick film magnets [6]–[9] thicker than 10 μm are key points to developing magnetic microactuators and systems (MAGMAS). For example, Nienhaus *et al.* have prepared disk-rotors made from Nd-Fe-B powders for planer micromotors [2]. Nakamura [4] *et al.* prepared a 100- μm -thick sintered bulk magnet with $(BH)_{\text{max}}$ of 245 kJ/m^3 by using refinement of grain size and surface treatment after a mechanical etching process. Since Cadieu [6] has reported a 40- μm -thick Sm-Co film magnet in 1989, many researchers have reported sputtering-made anisotropic thick film magnets prepared under the high deposition rate of 20–40 μm . Recently, Uehara [9] also reported high $(BH)_{\text{max}}$ value of 279 kJ/m^3 . In addition, other fabrication methods for thick film magnets such as the aerosol deposition method [10], plasma spraying [11], and tape casting [12] have been reported.

In our study, the pulsed laser deposition (PLD) method was used for preparing thick film magnets applied to various applications. Some researchers have reported PLD-prepared Nd-Fe-B [13], [14], Sm-Co [15], and Sm-Fe-N films [16], and the thickness of almost all the samples was less than 1 μm because the aims in fabricating the films were based on application for recording media and physical interest such as the epitaxial method. We prepared Nd-Fe-B film magnets thicker

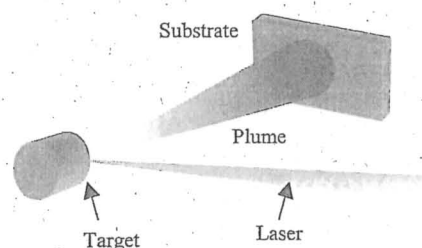


Fig. 1. Diagram of PLD method. The distance between a target and a substrate was fixed at 10 mm. The laser power was controlled from 3 to 10 W at an irradiation exit of a laser system.

than 20 μm under the deposition rate of 20–40 $\mu\text{m}/\text{h}$ by the PLD method [17]. In addition, the thick films were applied to a dc brush-less motor [18], a micromachine swimming in liquid [19], and an electromagnetic friction-drive motor.

This contribution reports the magnetic properties and structure for the PLD-fabricated isotropic Nd-Fe-B films under the high deposition rate up to 90 $\mu\text{m}/\text{h}$. Improvement in magnetic properties by preparing anisotropic films using a substrate heating system [18] was introduced. We also reviewed three applications comprising the isotropic films [19], [20].

II. PREPARATION OF THICK FILM MAGNETS

We used a high-speed PLD method to fabricate Nd-Fe-B thick film magnets. In order to compensate loss of metallic Nd due to oxidation, the nominal composition of targets was set to $\text{Nd}_{2.4}\text{Fe}_{14}\text{B}$, which include a larger amount of Nd than the stoichiometric composition. These targets were ablated with a Nd-YAG pulse laser ($\lambda = 355 \text{ nm}$) at the repetition rate of 30 Hz, and the distance between a target and various substrates such as Ta and Fe was fixed at 10 mm, as shown in Fig. 1. The deposition rate was varied from 20 to 90 $\mu\text{m}/\text{h}$ by controlling the laser power from 3 to 10 W at the irradiation exit of a Nd-YAG laser system. Before the ablation, the chamber was evacuated down to about 10^{-4} Pa with a molecular turbo pump.

Digital Object Identifier 10.1109/TMAG.2007.893778

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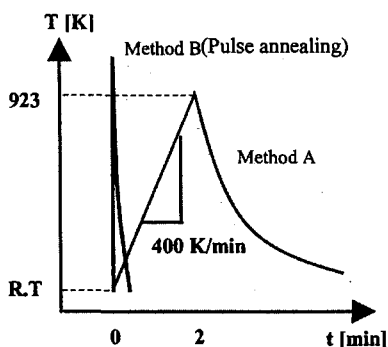


Fig. 2. Diagram of postannealing processes after deposition. In Method A, the samples were heated up to the designated temperature (923 K) under the heating rates of 400 K/min with an infrared furnace, and then they were cooled down to room temperature. In Method B, a pulse annealing with a high heating rate beyond 850 K/min was done.

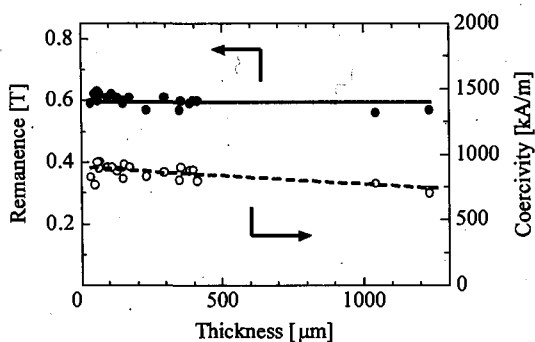


Fig. 3. Magnetic properties of remanence and coercivity in PLD-made Nd-Fe-B films as a function of film thickness. Although the coercivity slightly decreased as the thickness increased, the remanence was almost constant in the thickness range of 20–1200 μm .

The structure of all the as-deposited films prepared without using a substrate heating system was amorphous. In order to form the $\text{Nd}_2\text{Fe}_{14}\text{B}$ crystallized structure, the as-deposited films were postannealed under the vacuum of 10^{-3} Pa by using two methods, Methods A and B. In Method A, the samples were heated up to the designated temperature of 923 K under the heating rate of 400 K/min with an infrared furnace, and then they were cooled down to room temperature. In Method B, a pulse annealing with a high heating rate beyond 850 K/min²¹ was carried out as displayed in Fig. 2.

In-plane and perpendicular $M-H$ loops were measured with a vibrating sample magnetometer under an applied field up to 2.5 T. Before the measurement, a pulsed field exceeding 8 T was applied to all the samples in order to magnetize. The analysis of crystal structure was carried out with an X-ray diffractometer. Thickness was measured with a digital micrometer. Nd-Fe-B films with the thickness range up to 1200 μm could be successfully obtained without deterioration of mechanical properties although the coercivity slightly decreased as the thickness increased (see Fig. 3).

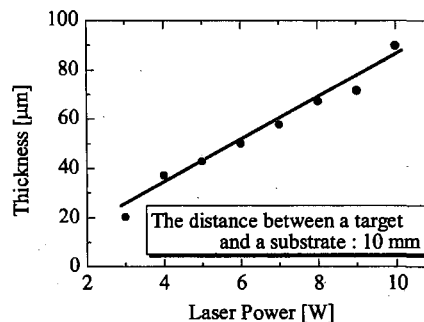


Fig. 4. Dependence of thickness on laser power in PLD-made Nd-Fe-B thick film magnets. Deposition time was fixed at 1 h. Increase in the power enabled us to increase the deposition rate.

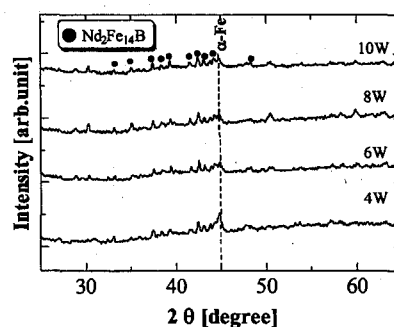


Fig. 5. X-ray diffraction patterns of annealed Nd-Fe-B thick film magnets. The films were prepared under various laser powers. The peaks corresponding to $\text{Nd}_2\text{Fe}_{14}\text{B}$ phases were observed in all the samples.

III. MAGNETIC PROPERTIES OF ISOTROPIC THICK FILM MAGNETS

A. Isotropic PLD-Made Nd-Fe-B Films Prepared Under the High Deposition Rate up to 90 $\mu\text{m}/\text{h}$

Fig. 4 shows the thickness of PLD-made Nd-Fe-B film magnets as a function of the laser power. As the power increased, the obtained thickness increased, and resultantly, the deposition rate was proportional to the laser power, approximately $9 \mu\text{m}/\text{h} \cdot \text{W}$. The wide deposition rate range between 20 and 90 $\mu\text{m}/\text{h}$ could be achieved in our PLD system, and the maximum value of 90 $\mu\text{m}/\text{h}$ was approximately two times as large as that reported by Kapitanov *et al.* [8].

According to observations in X-ray diffraction patterns for all the as-deposited films, the structure of the films was amorphous. Method A as a postannealing was executed to form $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase, and then magnetic properties for the annealed samples were measured. As shown in Fig. 5, Nd-Fe-B phase was observed in the samples under various laser powers. However, precipitation of $\alpha\text{-Fe}$ phase, which was attributed to the surface oxidation, was also observed. Remanence, coercivity, and $(BH)_{\text{max}}$ showed the constant values of 1050 kA/m, 0.6 T, and 60 kJ/m³, respectively, which were comparable to those previously reported [19] (see Figs. 6 and 7).

Although the surface roughness increased with increasing laser power and deposition rate, the peeling phenomenon was not observed in all the as-deposited films. We also confirmed that the postannealed films did not peel from its substrate. It is generally said that a peeling phenomenon occurs as the

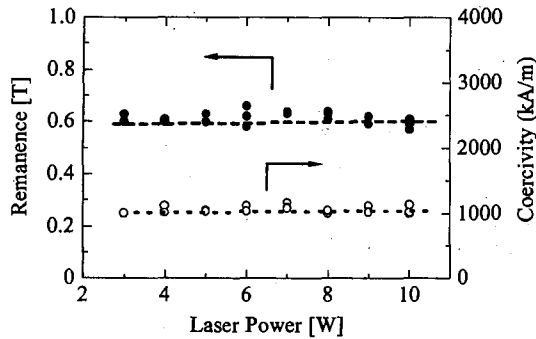


Fig. 6. Magnetic properties as a function of laser power. In the wide range of laser power between 3 and 10 W, the remanence and coercivity showed the constant values of approximately 0.6 T and 1000 kA/m, respectively.

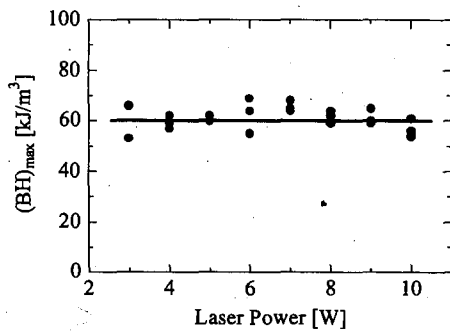


Fig. 7. $(BH)_{\max}$ as a function of laser power. In the wide range of laser power between 3 and 10 W, the $(BH)_{\max}$ showed the constant value of approximately 60 kJ/m³.

thickness of films increases because of the increase in an inner stress; however, the above-mentioned PLD-made samples did not show the peeling phenomenon. From these results, the increase in the laser power is effective in the increase in deposition rate without deterioration of magnetic and mechanical properties.

B. Microstructure of Isotropic PLD-Made Nd-Fe-B Films and Adoption in a Pulse Annealing

Investigation on microstructure of PLD-prepared Nd-Fe-B films is effective for the improvement in magnetic properties. Fig. 8 shows a transmission electron microscopy (TEM) image of a PLD-prepared isotropic film with the coercivity and remanence of 1080 kA/m and 0.59 T, respectively. The sample shown in Fig. 8(b) was prepared by etching from the surface by an FIB. TEM observations for the inside [Fig. 8(a)] of the film and boundary on a Ta substrate [Fig. 8(c)] were executed.

The microstructure indicated that the film was mainly composed of Nd₂Fe₁₄B grains whose size varied widely from 5 to 440 nm, and the average grain size was estimated as 150 nm. In addition, the grain growth seemed to occur directly on the substrate, because the boundary phase between the sample and the substrate did not affect the grain growth behavior in PLD-prepared thick films.

High-speed crystallization has been reported as one of hopeful methods for reducing the grain size and its distribution, and resultantly it enables us to improve the coercivity values of sputtered Nd-Fe-B films and rapid-quenched Pr-Fe-B ribbons

[21], [22]. We, therefore, adopted a pulse annealing method with a high heating rate beyond 850 K/min (see Fig. 2). As shown in Fig. 9, coercivity strongly depended on the pulse annealing period. Resultantly, crystallization by pulse annealing for 1.8 s led to enhanced coercivity of the films by 300 kA/m compared with previously reported ones [17], [19]. The microstructure of pulse-annealed sample is under investigation.

IV. ANISOTROPIC Nd-Fe-B FILM MAGNETS PREPARED BY THE HIGH-SPEED PLD METHOD WITH A SUBSTRATE HEATING SYSTEM

We further attempted to develop the above-mentioned PLD-made Nd-Fe-B thick films, and preparation of anisotropic thick film magnets was carried out by adoption of a substrate heating system between a target and a substrate. Use of the system enabled us to obtain anisotropic Nd-Fe-B thick film magnets under a high deposition rate.

We adopted a continuous deposition method, which means that a film is continuously deposited during heating substrate. Although the obtained remanence and $(BH)_{\max}$ values of the anisotropic films were higher than those of decreased by 40% compared to that of the isotropic ones [17], [19]. The deterioration of coercivity can be attributed to a heterogeneous grain growth due to heating during the long deposition time.

In order to overcome this difficulty, we adopted a new method, in which the deposition process was intercepted repeatedly during the substrate heating. We designated this method as the interceptive deposition method (IDM). The new preparation method, IDM, was applied to the preparation of Nd-Fe-B film magnets, and it was clarified that IDM is effective in fabricating anisotropic Nd-Fe-B thick film magnets under the deposition rate of 20–50 $\mu\text{m/h}$. Although the mechanism for the improvement in magnetic properties is under investigation, a magnetic micromotor comprising the IDM-made films is a hopeful candidate as a developed micromachine.

V. MICROMACHINES COMPRISING ISOTROPIC PLD-MADE Nd-Fe-B FILMS

Fig. 10 shows the structure of a small dc brush-less motor comprising a 200- μm -thick isotropic PLD-prepared Nd-Fe-B film magnet, which has 971 kA/m in coercivity and 0.57 T in remanence, respectively. Here, the film was deposited on a Fe substrate and the thickness and diameter of the motor were 0.8 and 5 mm, respectively. We confirmed that it rotates at 15160 rpm under no-load test, and has torque constant of 0.0236 mNm/A at the gap of 0.1 mm between a rotor and a stator. The motor is applicable to a small hard disk and a vibration motor inside a mobile phone.

We have further reported a spiral type micromachine with 0.14 mm in outer diameter and 1.0 mm in length prepared by a tungsten wire deposited on Nd-Fe-B film magnet. As the film magnet was magnetized in the circumferential direction, the machine rotated in synch with the rotating external magnetic field and the spiral structure generated propellant force. In our experiment, three types of liquids with kinematic viscosity of 1, 10, and 100 mm²/s, respectively, were used, and the external magnetic field of 8 kA/m was applied under the frequency range between 2 and 10 Hz. It was found that the wireless micromachine

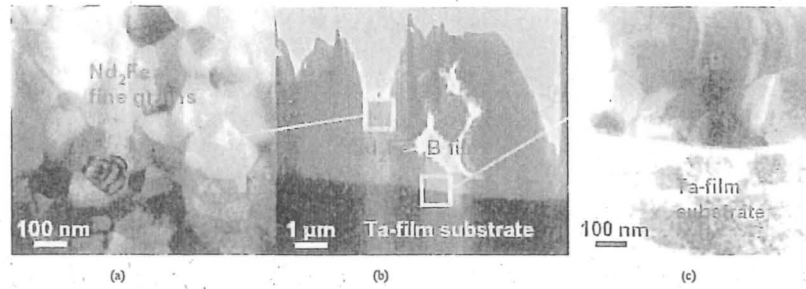


Fig. 8. Photos of TEM observations and cross sectional view for an isotropic Nd-Fe-B film magnet. (a) inside a sample, (b) TEM image in low magnification, (c) boundary on a Ta substrate.

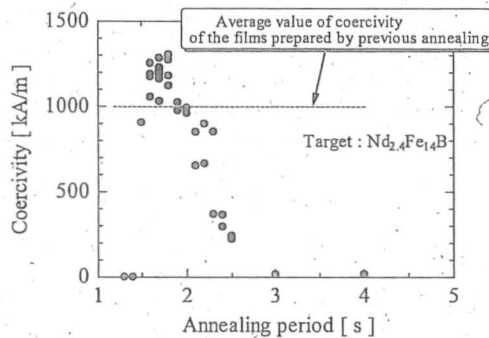


Fig. 9. After obtaining deposited Nd-Fe-B films, the samples were post-annealed by a pulse annealing method. Coercivity as a function of the annealing time. Samples prepared by the pulse annealing for 1.8 s led to enhance coercivity of the films by 300 kA/m compared with previously reported ones.

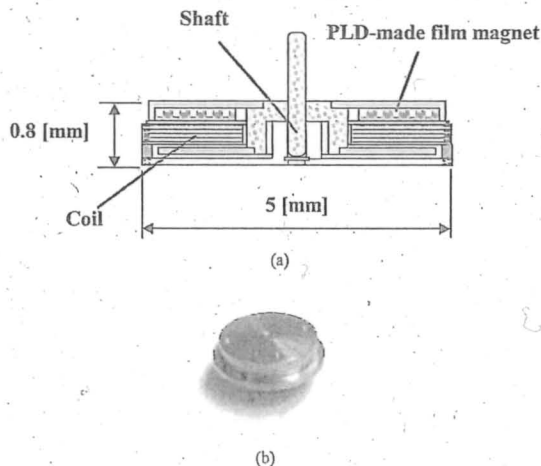


Fig. 10. Structure of a dc brush-less motor comprising a 200- μm -thick PLD-made Nd-Fe-B film magnet. It rotates at 15160 rpm under no-load test, and has torque constant of 0.0236 mNm/A. (a) Schematic diagram of a milli-size motor. (b) Photograph of a milli-size motor.

swam at the speed of 0.2–1.6 mm/s under the various conditions. The actuator is a hopeful candidate for a drug delivery machine in human blood vessel.

Recently, a new type micromotor comprising a PLD-made 384 μm -thick Nd-Fe-B film magnet has been reported. The electromagnetic friction-drive micromotor had a large torque and a low rotational speed. The stator is a coil with a ferrite core in the center and a ferrite disc on the bottom. Alternating voltage was applied to the coil. Between the film magnet and the ferrite core, pulling and reacting forces worked alternatively,

and resultantly the magnet vibrated. The rotor is magnetically mounted onto the stator without mechanical attachments. The motor rotated at approximately 300 rpm with starting torque of approximately 2 μNm . It was confirmed that the new class of micromotor using a thick film magnet is one of hopeful candidates as a new micromachine.

VI. CONCLUSION

Isotropic Nd-Fe-B thick film magnets prepared by the high-speed PLD method could be applied to a small dc brush-less motor, a machine swimming in liquid, and an electromagnetic friction-drive micromotor. Anisotropic PLD-prepared Nd-Fe-B thick film magnets are promising materials to advance various small magnetic micromachines.

ACKNOWLEDGMENT

This work was supported in part by the Ministry of Education, Science, Sports and Culture of Japan under a Grand-in-Aid (16686022, 10136533) and in part by the Energy Use Rationalization Technology and Strategy Developments Program in 2005 from the New Energy and Industrial Technology Development Organization (NEDO, Project P03033) of Japan.

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Manuscript received October 31, 2006 (e-mail: mnakano@nagasaki-u.ac.jp).