A method of preparing anisotropic Nd–Fe–B film magnets by pulsed laser deposition

M. Nakano,[a] S. Sato, H. Fukunaga, and F. Yamashita[b]
Department of Electrical and Electronic Engineering, Nagasaki University, Nagasaki 852-8521, Japan

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The interceptive deposition method (IDM) was applied to the high-speed pulsed laser deposition (PLD) method with a substrate heating system, and it was clarified that anisotropic Nd-Fe-B thick-film magnets can be obtained under a high deposition rate larger than 20–50 μm/h. The obtained coercivity, remanence, and $(BH)_{max}$ values of a film were 577 kA/m, 0.90 T, and 118 kJ/m^3, respectively. In addition, we succeeded in increasing the thickness up to 90 μm for an anisotropic Nd-Fe-B film magnet by using IDM.
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I. INTRODUCTION

Rare earth magnets with thickness of 100–300 μm are required for advancing a size reduction in electronic devices such as millisize motors and microactuators.[1] In the sputtering method, Nd-Fe-B film magnets thicker than 10 μm have been reported.[2–6] It has also confirmed that a substrate heating method is effective in obtaining anisotropic sputtered Nd-Fe-B samples.[2–6] For example, Uehara reported superior magnetic properties of a Nd-Fe-B/Ta multilayered thick film. (H_{CJ}: 1210 kA/m, J_r: 1.24 T, $(BH)_{max}$: 279 kJ/m^3).[6] However, it is generally accepted that the sputtering method has a difficulty in realizing a high deposition rate. Although Kapitanov *et al.* reported anisotropic Nd-Fe-B film magnets with thickness of 30–300 μm under a high deposition rate of 20–40 μm/h,[3] the detailed sputter conditions, the relationship between magnetic properties and film thickness, and the evaluation method of magnetization were not shown clearly.

On the other hand, we reported Nd-Fe-B thick films prepared by the PLD method with a deposition rate of 20–80 μm/h at the substrate area of 10×10 mm^2,[7] and succeeded in applying a 200 μm thick isotropic Nd-Fe-B film on a Fe substrate to a millisize motor.[8] Recently, we also confirmed that the PLD method with a substrate heating system enables us to obtain anisotropic Nd-Fe-B thick-film magnets under a high deposition rate. In this study, we adopted the continuous deposition method (CDM) shown in Fig. 1(a), which means continuous deposition on a heated substrate. Although the obtained remanence and $(BH)_{max}$ values of the anisotropic films were higher than those of isotropic ones reported previously, the value of coercivity decreased by 40% compared to that of the isotropic ones.[7–9] 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 method, in which the deposition process was intercepted repeatedly during the substrate heating [see Fig. 1(b)]. We designated this method as the interceptive deposition method (IDM).

In this report, a preparation method, IDM, was applied to preparation of Nd-Fe-B film magnets, and it was clarified that IDM is effective in preparing anisotropic Nd-Fe-B thick-film magnets under the deposition rate of 20–50 μm/h. The values of coercivity, remanence, and $(BH)_{max}$ of the films prepared by IDM were superior to those of the CDM-made films reported previously.

II. EXPERIMENTAL PROCEDURE

In order to compensate loss of metallic Nd due to oxidation during a deposition, the nominal composition of targets was set to $Nd_{2.4}Fe_{14}B$. The targets include a larger amount of Nd compared with the stoichiometric composition. Prior to deposition, the chamber was evacuated down to approximately 10^{-4} Pa with a molecular turbo pump, and a Ti sublimation pump was used as an auxiliary pump during the deposition. The targets were ablated with a Nd-YAG pulse laser ($\lambda=355$ nm) at the repetition rate of 30 Hz, and the distance between a target and a Ta substrate was fixed at 10 mm. The area of each film was 5×5 mm^2. The substrate heating system was made of a Ta foil. Electric current flowed through the Ta foil, and a sample on the substrate was heated

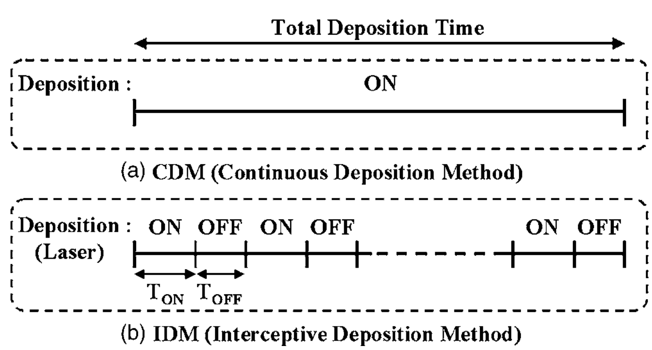

FIG. 1. Two types of methods of preparing thick-film magnets adopted in this study.

[a]Electronic mail: mnakano@net.nagasaki-u.ac.jp
[b]Present address: Motor Technology Center, Matsushita Electric Industrial Co., Osaka 574-0044, Japan.

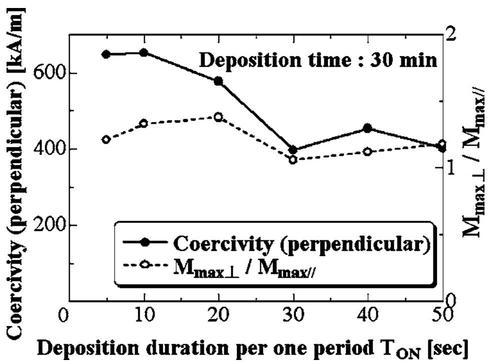

FIG. 2. Coercivity and $M_{\max\perp}/M_{\max\parallel}$ of film magnets prepared by IDM as a function of deposition duration per one period.

by Joule heat during deposition. The substrate temperature was fixed at 873 K. We used two deposition methods, CDM and IDM, shown in Fig. 1.

In-plane and perpendicular *M-H* loops were measured with a vibrating sample magnetometer (VSM) under an applied field up to 2.5 T after magnetization under a pulsed field exceeding 8 T. The analysis of crystal structure was carried out with an x-ray diffractometer (XRD). Thickness was measured with a micrometer.

III. RESULTS AND DISCUSSION

Figure 2 shows coercivity and $M_{\max\perp}/M_{\max\parallel}$ of film magnets prepared by IDM as a function of deposition duration T_{on} per one period. The total deposition time and the interception duration T_{off} per one period were fixed at 30 min and 10 s, respectively. The values of $M_{\max\perp}/M_{\max\parallel}$ were evaluated without the correction of the demagnetizing effect. The film thickness was approximately 3 μm at $T_{\mathrm{on}}=5$ s, approximately 10 μm at $T_{\mathrm{on}}=10$ s, and 20–25 μm at $T_{\mathrm{on}}=20-50$ s. The deposition rate did not depend on T_{on} for T_{on} between 20 and 50 s, and further investigations on this mechanism are required. The values of $M_{\max\perp}/M_{\max\parallel}$ were larger than 1.0 for all the T_{on} values investigated, and show a tendency of increasing with decreasing T_{on}, which suggests that IDM is effective for obtaining films with perpendicular anisotropy under the high deposition rate of 20–50 μm/h. Although a decrease in T_{on} leads to increases not only in the values of coercivity but also $M_{\max\perp}/M_{\max\parallel}$, it also results in

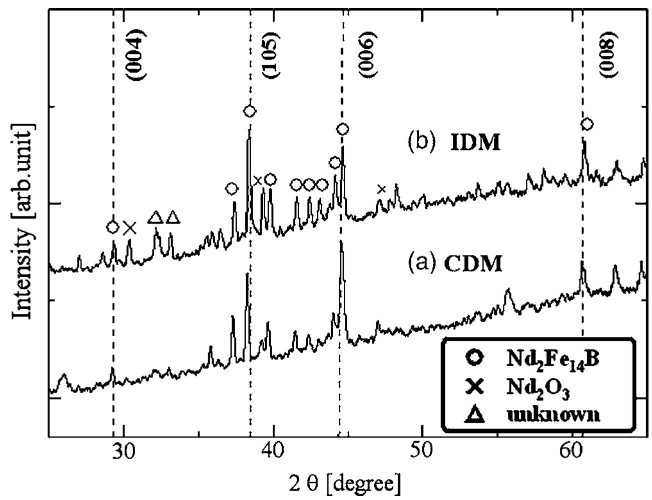

FIG. 4. X-ray diffraction patterns of Nd-Fe-B films prepared by CDM and IDM.

a reduction in thickness and a deposition rate. We therefore selected $T_{\mathrm{on}}=20$ s as an optimum deposition condition.

Figure 3 shows *M-H* loops of an IDM-made Nd-Fe-B film with thickness of approximately 22 μm prepared at $T_{\mathrm{on}}=20$ s. Although the correction of the demagnetization effect was not carried out, the values of magnetization and coercivity of the perpendicular loop were higher than those of the in-plane one. The coercivity value of the perpendicular one increased by 100 kA/m compared with those of CDM-made films.

In order to investigate the mechanism for obtaining anisotropic Nd–Fe–B thick-film magnets with good magnetic properties, x-ray diffraction patterns of the above-mentioned IDM-made film were observed as displayed in Fig. 4. In the figure, the pattern of a CDM-made film with thickness of approximately 24 μm was also shown. The strong peaks corresponding to the *c* plane such as (004), (006), (008), and (105) were observed in the films prepared by the both methods. On the other hand, unknown peaks at $2\theta=32,33$, were observed only for the IDM-made film. As Uehara reported that anisotropic Nd-Fe-B films with superior magnetic properties had the same unknown phases,[6] our result agrees with his experimental one.

Figure 5 shows demagnetization curves of the Nd-Fe–B films shown in Fig. 4. Both curves were measured in the

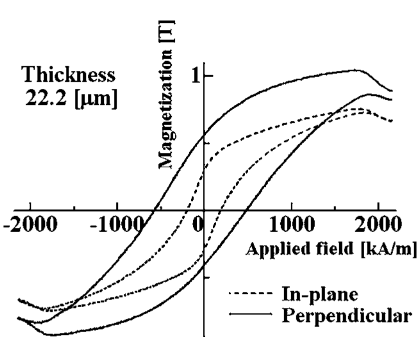

FIG. 3. In-plane and perpendicular *M-H* loops of Nd–Fe–B film prepared by IDM. In the preparation, T_{on} was set at 20 s. The correction of the demagnetization effect is not carried out.

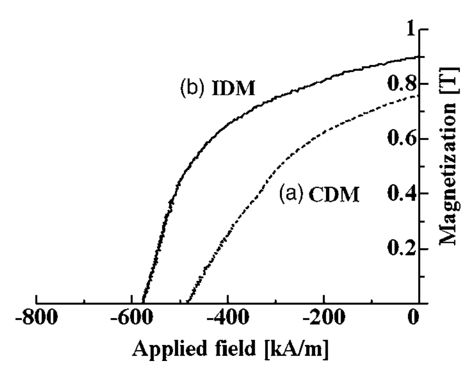

FIG. 5. Demagnetization curves of Nd-Fe-B films prepared by CDM and IDM. The curves were measured in the perpendicular direction and were corrected by using the demagnetization factor of 1.0.

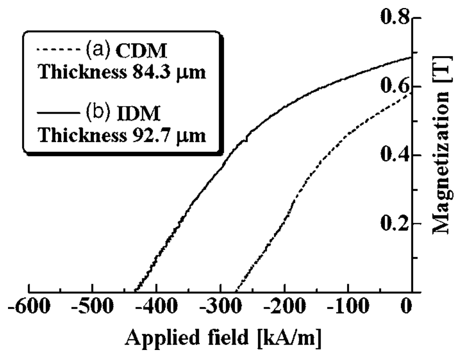

FIG. 6. Demagnetization curves of IDM-made film with thickness of 92.7 μm and CDM-made film with thickness of 84.3 μm. The curves were measured in the perpendicular direction, and were corrected by using demagnetization factor of 1.0.

perpendicular direction and corrected by using the demagnetization factor of 1.0.[10] Magnetic properties of the IDM-made film were superior compared to those of the CDM-made film. The coercivity, remanence, and $(BH)_{max}$ of the IDM-made film were 577 kA/m, 0.90 T, and 118 kJ/m^3, respectively. As an addition of interceptive duration in IDM is expected to suppress heterogeneous crystal growth or bloated crystal grains, an observation of microstructure of the films is needed for further investigations.

In some applications, it is hoped to increase the thickness up to approximately 100 μm.[1] Figure 6 shows a demagnetization curve of IDM-made Nd-Fe–B film with thickness of approximately 93 μm. The total deposition time, T_{on} and T_{off} were 360 min, 10 s, and 10 s, respectively. A demagnetization curve of a CDM-made film with thickness of approximately 84 μm was also shown in the figure. Although magnetic properties were deteriorated by increasing the thickness as shown in Figs. 5 and 6, magnetic properties of the IDM-made 93 μm thick film were superior compared to those of the CDM-made 84 μm thick film as shown in Fig. 6. In addition, the coercivity value of 437 kA/m was approximately twice as large as that of previously reported anisotropic 80 μm thick Nd-Fe-B films prepared by sputtering.[4] These results suggest that IDM is effective in improving the magnetic properties of anisotropic magnetic films with thickness up to approximately 90 μm.

IV. CONCLUSION

A new preparation method IDM (interceptive deposition method) was applied to preparation of anisotropic Nd-Fe-B thick-film magnets. Usage of IDM boosted the values of coercivity, remanence, and $(BH)_{max}$ of anisotropic Nd-Fe-B thick films compared with those of CDM-made anisotropic ones reported previously. It was also clarified that IDM is effective in improving the magnetic properties of anisotropic magnetic films with thickness up to approximately 90 μm. Consequently, we can conclude that IDM is a hopeful method for obtaining anisotropic Nd-Fe-B thick-film magnets under a high deposition rate.

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