

Enhancement in $(BH)_{\max}$ of PLD-made isotropic Nd-Fe-B thick film magnets deposited on Si substrates

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Increase in Nd contents of a PLD-made isotropic Nd-Fe-B thick-film magnet enabled us to enhance the thickness of the film magnet deposited on a Si substrate because the linear expansion coefficient of Nd is an intermediate value between Nd₂Fe₁₄B and Si. The large amount of Nd, however, degraded the residual magnetic polarization and $(BH)_{\max}$. In the study, we reduced the Nd contents of each Nd-Fe-B film by inserting a Nd or a Nd-rich Nd-Fe-B buffer layer between a Nd-Fe-B film and a Si substrate in order to suppress the mechanical destruction together with the improvement in magnetic properties. It was found that the mechanical property of a Nd-Fe-B film comprising the Nd-Fe-B buffer layer in the thickness range from 10 to 60 μm was superior than that of a sample with the Nd buffer layer. Resultantly, an average $(BH)_{\max}$ value of Nd-Fe-B films with each Nd-Fe-B buffer layer deposited on Si substrates could be enhanced by approximately 15 kJ/m³ compared to that of non-buffer-layered films. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4979688>]

I. INTRODUCTION

Preparation of a rare-earth permanent film magnet on a metal substrate has been reported using several methods such as a sputtering, a vacuum arc deposition and an aerosol deposition.¹⁻⁵ Our group has focused on a PLD method to obtain isotropic Nd-Fe-B thick-film magnets on metal substrates (Ta, Fe, W) under the relatively high deposition rate of several-ten-microns per hour and applied the films to several miniaturized electronic devices.^{6,7} Moreover, a sputtering-fabricated Nd-Fe-B thick-film magnet on a Si wafer with a Ta buffer layer has been reported in order to develop the device using a micromachining technology.⁸⁻¹⁰ We, however, confirmed that a Nd-Fe-B thick-film magnet deposited on a Si substrate with an approximately 1 μm -thick Ta buffer layer was broken through a dicing process. It is also generally known that the use of the thick Ta layer is not suitable for a chemical etching process. Although a Nd-Fe-B film thicker than 5 μm on a Si substrate without a buffer layer was demonstrated using a sputtering method,¹¹ the film was peeled out from the Si substrate through the preparation process. In order to overcome the difficulty, we increased the Nd content of a PLD-made Nd-Fe-B thick-film by taking account of the linear expansion coefficient for each material (see Table I) and succeeded in enhancing the thickness up to approximately 160 μm on a Si substrate without a Ta buffer layer.¹² The average $(BH)_{\max}$ value of the thick-films, however, was extremely low of approximately 35 kJ/m³. In the previous report,¹² we concluded that the existence of Nd around a grain boundary together with a triple junction was indispensable to suppress the mechanical destruction through a post-annealing process.¹² A recent observation of the microstructure for the above-mentioned film also indicated that an approximately 500 nm-thick Nd layer precipitated between a Nd-Fe-B film and a SiO₂/Si substrate as shown in Fig. 1.

This contribution reports an increase in $(BH)_{\max}$ of a PLD-made isotropic Nd-Fe-B film magnet deposited on a Si substrate. Although the value of Nd/(Nd+Fe) in previously reported films exceeded

TABLE I. Linear expansion coefficient of each material.

materials	linear expansion coefficient ($\times 10^{-6} \text{ K}^{-1}$)
$\text{Nd}_2\text{Fe}_{14}\text{B}$	14.7
Nd	9.6
Ta	6.3
Si	2.6

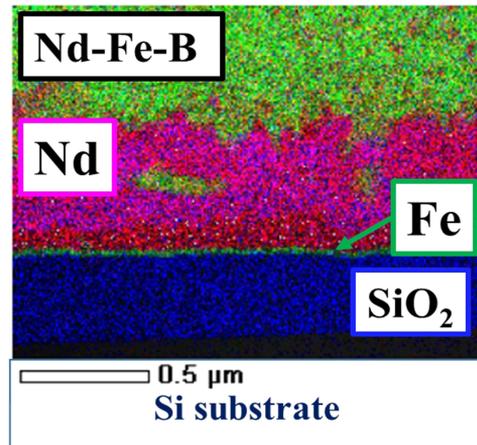


FIG. 1. TEM observations of a PLD-made isotropic Nd-Fe-B film magnet deposited on a Si substrate without a buffer layer. Nd/(Nd+Fe) of a Nd-Fe-B film was approximately 26 at.%. The precipitation of Nd together with Fe layer could be observed.

20 at. % to suppress the mechanical destruction, we could reduce the value range 10 to 14 at. % by inserting a Nd or a Nd-rich Nd-Fe-B buffer layer. It was found that the adoption of the Nd-Fe-B buffer layer is effective to suppress a mechanical destruction and to achieve the reproducibility. Moreover, we could enhance the average $(BH)_{\text{max}}$ value of PLD-made Nd-Fe-B thick-film magnets in the thickness up to 60 μm .

II. EXPERIMENTAL PROCEDURE

A Nd-Fe-B target ($\text{Nd}_{2.0}\text{Fe}_{14}\text{B}$ or $\text{Nd}_{2.4}\text{Fe}_{14}\text{B}$) was ablated with a Nd-YAG pulse laser (wave length: 355 nm) at the repetition rate of 30 Hz in vacuum atmosphere. In order to prepare two kinds of buffer layer in situ, a Nd or a $\text{Nd}_{4.0}\text{Fe}_{14}\text{B}$ target, respectively, was also ablated in a chamber comprising a multi target holder (see Figs. 2 and 3). The Nd/(Nd+Fe) value of a Nd-rich buffer layer

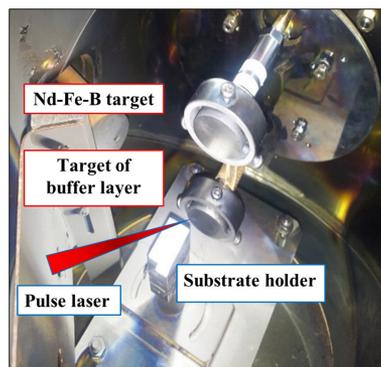


FIG. 2. Photo of a multi-target holder in a PLD equipment. The deposition of a buffer layer followed by the preparation of a Nd-Fe-B film magnet in-situ was carried out using the system.

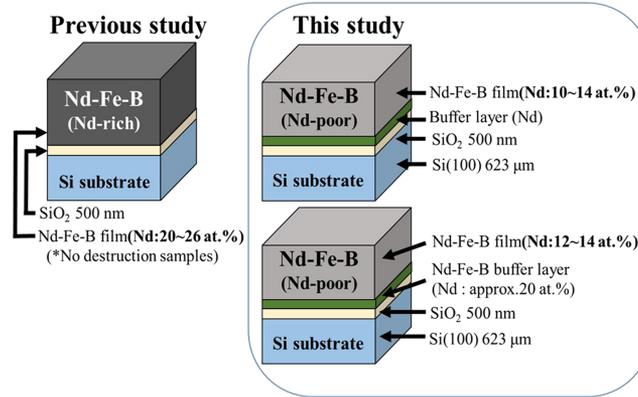


FIG. 3. Diagrams of the films prepared in the study. Nd or Nd-rich Nd-Fe-B buffer layer was deposited on a SiO₂/Si substrate.

was approximately 20 at. %. The laser energy density on each surface of a target was estimated as approximately 4 J/m² using the laser power of 4 W which 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 2.0~8.0×10⁻⁵ Pa using a rotary pump together with a molecular turbo one. The distance between a target and a SiO₂ (500 nm)/Si substrate was fixed at 10 mm, and the area of all the obtained films were 5×5 mm². Average deposition rate of each film exceeded 20 μm/h. All the as-deposited Nd-Fe-B film had amorphous structure. We, therefore, used a pulse annealing (PA) to crystallize the films.¹³ After an annealed sample was magnetized up to 9 T with a pulse magnetizer, J-H loops were measured with a vibrating sample magnetometer (VSM) which could apply a magnetic field up to approximately 1800 kA/m reversibly. The film composition was evaluated with a SEM-EDX, and the surface observation was also carried out using a SEM. 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 or estimated by measuring each weight.

III. RESULTS AND DISCUSSION

As a first step, the deposition of a Nd buffer layer followed by the preparation of a Nd-Fe-B film magnet in-situ was carried out. The thickness range of the Nd layer was set from 1 to 5 μm. Figure 4 shows the relationship between the composition excluding the Nd buffer layer and the total thickness in each sample. The samples displayed in the figure did not show a mechanical destruction due to the annealing process. As shown in Fig. 5, increase in the thickness of the Nd layer enabled us to enhance the maximum thickness of the films. It was confirmed that as the values of $(BH)_{\max}$ became higher than 50 kJ/m³, Nd/(Nd+Fe) tended to become less than 12 at. %. The 60 μm-thick Nd-Fe-B film magnet

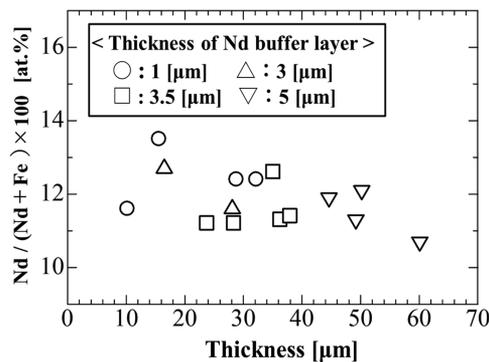


FIG. 4. Relationship between the composition of a Nd-Fe-B layer and the total thickness of each sample including a Nd buffer layer. Various Nd-Fe-B films with each Nd buffer layer.

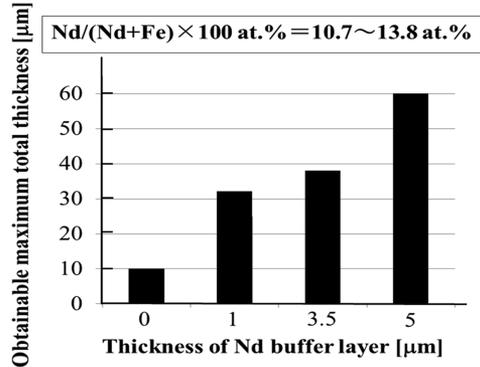


FIG. 5. Relationship between the composition of a Nd-Fe-B layer and the total thickness of each sample including a Nd buffer layer. Obtained maximum total thickness of Nd-Fe-B film as a function of thickness of a Nd buffer layer.

had the $(BH)_{\max}$ value of approximately 64 kJ/m^3 on a SiO_2/Si substrate because $\text{Nd}/(\text{Fe}+\text{Nd})$ of the Nd-Fe-B film could be reduced to 10.7 at. %. We, however, confirmed that several films comprising a Nd buffer layer showed a mechanical destruction such as a peeling phenomenon as seen in Fig. 6. In particular, the peeling phenomenon occurred as the thickness of the Nd layer exceeded approximately $5 \mu\text{m}$. Namely, it was difficult to achieve the reproducibility in the films using a Nd buffer layer. As seen in the previous report,¹² all the PLD-made Nd-Fe-B films deposited on SiO_2/Si substrates excluding a buffer layer didn't show the peeling phenomenon. Moreover, we could observe the existence of very thin Fe layer at the boundary area in the films (see Fig. 1). These results suggest that the existence of Fe is required in order to obtain the strong adhesion between a Nd-Fe-B film and a SiO_2/Si substrate.

We, therefore, prepared a Nd-rich Nd-Fe-B buffer layer with the thickness range from 3.8 to $7.0 \mu\text{m}$ using a $\text{Nd}_{4.0}\text{Fe}_{14}\text{B}$ target instead of the above-mentioned Nd buffer layer. As shown in Fig. 7, the total thickness of each film could be enhanced up to approximately $60 \mu\text{m}$ without mechanical destruction. Here, the $\text{Nd}/(\text{Nd}+\text{Fe})$ values were measured in each Nd-Fe-B film excluding a Nd-Fe-B buffer layer. The result indicated that a Nd-rich Nd-Fe-B buffer layer is effective to suppress the stress between a Si substrate and a Nd-Fe-B film and the reproducibility without the mechanical destruction could be improved. On the other hand, we had difficulty in enhancing the thickness above $60 \mu\text{m}$. An optimization of composition and thickness of a Nd-rich Nd-Fe-B layer, therefore, is required to increase the total thickness of a sample. Figure 8 shows the comparison of magnetic properties between Nd-Fe-B films with and without a Nd-rich Nd-Fe-B buffer layer. Reduction in the total amount of Nd in each thick-film by inserting the buffer layer enabled us to increase the average $(BH)_{\max}$ value by approximately 15 kJ/m^3 . The average value, however, was more inferior than those of previously reported ones of nano-composite Nd-Fe-B/ α -Fe film magnets deposited on

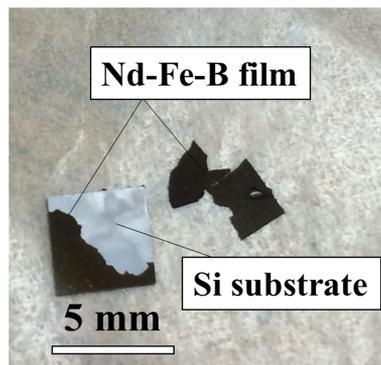


FIG. 6. Photo of a broken film through a post annealing process. Peeling phenomenon occurred in a Nd-Fe-B film with a Nd buffer layer deposited on a Si substrate.

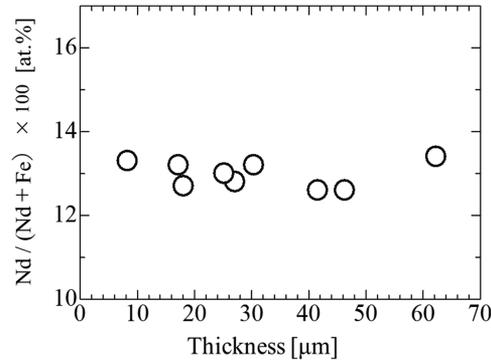


FIG. 7. Relationship between Nd/(Nd+Fe) of each Nd-Fe-B film and the thickness of a Nd-Fe-B film with a Nd-rich Nd-Fe-B buffer layer deposited on a Si substrate. Mechanical destruction didn't occur in the films through a post annealing.

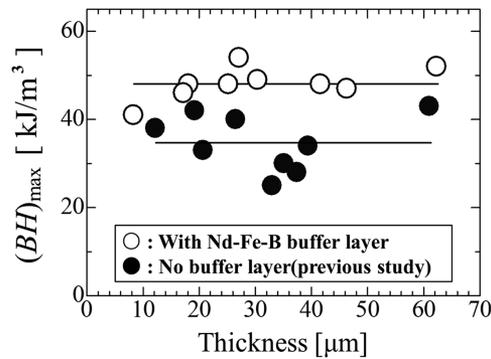


FIG. 8. $(BH)_{\text{max}}$ values in each Nd-Fe-B film with a Nd-rich Nd-Fe-B buffer layer. Here, $(BH)_{\text{max}}$ values of Nd-Fe-B films without a buffer layer were also shown. Use of a Nd-Fe-B buffer layer enabled us to increase $(BH)_{\text{max}}$ by approximately $15 \text{ kJ}/\text{m}^3$.

Ta substrates.^{14,15} Further improvement in $(BH)_{\text{max}}$ of a Nd-Fe-B film on a Si substrate is required as a future work.

IV. CONCLUSION

In order to improve the magnetic properties, an adoption of a Nd or a Nd-rich buffer layer was carried out. Although the both buffer layers were effective to enhance $(BH)_{\text{max}}$ value in PLD-made isotropic Nd-Fe-B thick-films with the thickness range up to approximately $60 \mu\text{m}$ deposited on a Si substrate, the Nd-rich Nd-Fe-B buffer layer had a superior reproducibility in the mechanical property. In order to achieve a further increase in the thickness and $(BH)_{\text{max}}$, an optimization of composition together with thickness of a Nd-rich Nd-Fe-B buffer layer is required.

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