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# Magnetic properties of pulsed laser deposition-fabricated isotropic Pr-Fe-B thick-films magnets for magnetic micro-machines

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A preparation of Pr-Fe-B thick-film magnets by using a PLD (Pulsed Laser Deposition) method with the energy density at approximately 50 mJ/m$^2$ using the laser power of 2 W was effective to obtain the required magnetic properties for the application to a multi-polarly magnetized rotor. The isotropic films mainly consisted of $Pr_2Fe_{14}B$ magnetic phase whose saturation magnetization is approximately 1.56 T, however the remanence ($B_r$) showed higher than 0.9 T. The obtained value of $B_r$ was considered to be attributed to a remanence enhancement due to the interaction of $Pr_2Fe_{14}B$ hard magnetic phases. Resultantly, the coercivity and $(BH)_{max}$ of the samples exceeded 600 kA/m and 115 kJ/m$^3$, respectively, which were larger than that of previously reported PLD-fabricated isotropic rare-earth based thick-films. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4867130]

## I. INTRODUCTION

In order to advance a microelectro–mechanical systems (MEMS), small rare-earth magnets have been prepared.[1–3] A lot of researchers worked on pulsed laser deposition (PLD) rare-earth films such as Nd-Fe-B, Sm-Co, and Sm-Fe-N ones.[4–10] Recently, we found that the fabrication of a multi-polarly magnetized rotor using isotropic Nd-Fe-B thick-film magnets is effective to develop a miniaturized cylindrical motor.[11] The required values of coercivity, remanence, and $(BH)_{max}$ for the thick-films applied to the rotor are 0.9 T, 400 kA/m, and 90 kJ/m$^3$, respectively, and we demonstrated that isotropic Nd-Fe-B/α-Fe nano-composite thick-films had sufficient values of remanence and $(BH)_{max}$ by using the PLD method with the laser energy density at 100 mJ/m$^2$.[12] Furthermore, the usage of a small spot size of a laser beam, which means that a laser beam was focused on the surface of a target according to the focus distance, in the experiment showed a grain refinement without the adoption of additives in the microstructure of PLD-fabricated thick films. On the other hand, the coercivity value of the nano-composite samples was less than 400 kA/m, namely it was difficult to achieve the reproducibility of required coercivity value. It is generally known that the magnetic crystalline anisotropy constant of a $Pr_2Fe_{14}B$ phase ($K_u = 6.8$ MJ/m$^3$) is larger by approximately 2.3 MJ/m$^3$ than that of a $Nd_2Fe_{14}B$ phase ($K_u = 4.5$ MJ/m$^3$).[13]

This contribution reports the ablation of Pr-Fe-B targets with various compositions by using the small spot size and the evaluation on the properties of the obtained samples. Resultantly, the laser energy density at approximately 50 mJ/mm$^2$ using the laser power of 2 W enabled us to obtain the required properties. It was also considered that the origin of superior magnetic properties is attributed to a remanence enhancement due to the magnetic interaction between $Pr_2Fe_{14}B$ grains.

## II. EXPRIMENTAL PROCEDURE

The Pr-Fe-B targets with various compositions were ablated by an Nd-YAG pulse laser (wave length = 355 nm, frequency of the wave = 30 Hz). Each film was deposited on a Ta substrate in the vacuum atmosphere of approximately $10^{-5}$ Pa. The laser beam was focused on the surface of each rotating target, and the laser energy density varied by controlling the laser power range from 1 to 4 W, which was measured with a power meter in front of the entrance lens of the chamber. A deposition rate higher than 10 μm/h could be obtained for a distance of 10 mm between a target and a substrate. All the as-deposited films thicker than 10 μm were crystallized by a pulse annealing (PA) method in the vacuum atmosphere of $2–5 \times 10^{-5}$ Pa. The pulse-annealing time was approximately 1.7 s with an infrared furnace at output power of 8 kW, and then they were cooled down to room temperature.[14]

The magnetic properties of the samples were measured with a vibrating sample magnetometer (VSM) under the maximum applied magnetic field of 2.5 T after a magnetization with a pulsed magnetic field of 7 T. All the films had isotropic magnetic properties, therefore in-plane ones were shown in the paper. The film thickness was measured with a micrometer, and the composition of each film was analyzed with an energy dispersive X-ray spectrometry (EDX). Furthermore, the crystalline structure of each film was observed by using a X-ray diffraction.

## III. RESULTS AND DISCUSSION

### A. Magnetic properties and crystalline structure of Pr-Fe-B/α-Fe together with Nd-Fe-B/α-Fe nano-composite thick-film magnets prepared by using the laser power of 4 W

Figure 1 shows the values of remanence and coercivity of Pr-Fe-B/α-Fe together with Nd-Fe-B/α-Fe thick-film magnets prepared by using the laser energy density at approximately 100 mJ/mm$^2$ using the laser power of 4 W. Each target composition was $Pr_{2.4}Fe_{14}B$ and $Nd_{2.4}Fe_{14}B$, respectively, and the

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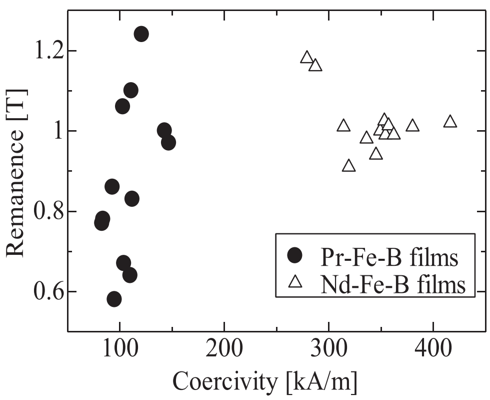


FIG. 1. Remanence and coercivity values of Pr-Fe-B/α-Fe and Nd-Fe-B/α-Fe nano-composite thick-films prepared by using the laser energy density at approximately 100 mJ/m$^2$ using the laser power of 4 W. Each target composition was $Pr_{2.4}Fe_{14}B$ and $Nd_{2.4}Fe_{14}B$, respectively, and the magnetic properties of post-annealed films were indicated. The same deposition and post-annealing processes were carried out in all the samples.

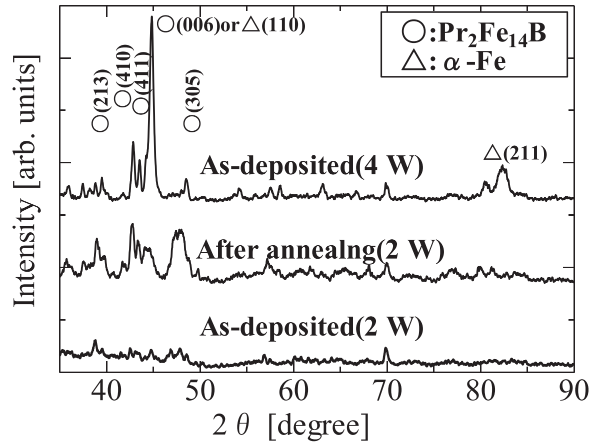


FIG. 2. X-ray diffraction patterns of as-deposited and annealed Pr-Fe-B films prepared by using a $Pr_{2.4}Fe_{14}B$ target together with the laser power of 2 W. XRD pattern of an as-deposited film prepared at 4 W was also shown.

magnetic properties of post-annealed films were indicated. The conditions of deposition and post-annealing processes were same in the both samples, and the results of Nd-Fe-B films shown in the figure have already been reported in Ref. 12. In addition, the rare-earth (Pr or Nd) composition range from 9 to 11 at. % was almost the same in the both of nano-composite films, however, the magnetic properties were much different. Although the values of coercivity and remanence were higher than 300 kA/m and 0.9 T in the Nd-Fe-B/α-Fe thick-film magnets, the coercivity and remanence values of Pr-Fe-B/α-Fe films were lower and more fluctuating, respectively. In order to investigate the different properties for the both films, the observation of crystalline structure together with the measurement on M-H loops of the as-deposited films were carried out. As previously reported in Ref. 12, an α-Fe crystalline phase was mainly observed in an as-deposited Nd-Fe-B/α-Fe film. On the other hand, an as-deposited Pr-Fe-B/α-Fe film had the diffraction peaks of a $Pr_2Fe_{14}B$ crystalline phase together with an α-Fe phase. In addition, the M-H loop of as-deposited Pr-Fe-B/α-Fe film widened compared with that of Nd-Fe-B/α-Fe one. It is generally known that the amorphous structure of the hard magnetic phase before an annealing process is indispensable to achieve good magnetic properties in nano-composite magnets.[15] It, therefore, was considered that we had difficulty in obtaining large coercivity values of Pr-Fe-B/α-Fe films as shown in Fig. 1. Furthermore, the substrate temperatures of the both films during the deposition with the laser power of 4 W were confirmed to be approximately 673 K which suggests the requirement of reduction in the substrate temperature to suppress the crystallization of $Pr_2Fe_{14}B$ phase.

## B. Magnetic properties of thick-film magnets prepared by ablating a Pr-Fe-B target with a small spot size of a laser beam under the laser power range between 1 and 3 W

We have already reported that the rise of substrate temperature is attributed to the heat radiation from a target during the fabrication of Fe-Pt films by using the PLD,[16] therefore the reduction in the laser power from 3 to 1 W was carried out in order to suppress the crystallization of $Pr_2Fe_{14}B$ phase. Resultantly, it was clarified that the use of 2 W (Laser energy density: Higher than 50 mJ/mm$^2$) is an optimum condition to prepare a thick film with good magnetic properties under the high deposition rate. Namely, in the case of 3 W, the magnetic properties did not change compared with those of films prepared at 4 W, and we had difficulty in obtaining relatively high deposition rate larger than 10 $\mu$m/h at the power of 1 W.

Figure 2 shows the X-ray diffraction patterns of as-deposited and annealed Pr-Fe-B films prepared by using a $Pr_{2.4}Fe_{14}B$ target together with the laser power of 2 W. Here, an XRD pattern of an as-deposited film prepared at 4 W was also shown in the figure. The film compositions for the 2 W case were almost the same as the stoichiometric one. The diffraction peaks corresponding to $Pr_2Fe_{14}B$ phases in the samples prepared at 2 W were not strong compared with those for the 4 W case. Furthermore, the average grain size of $Pr_2Fe_{14}B$ phase was approximately 18 nm according to the estimation of Scherrer's equation.[17] The magnetic properties of the annealed films were shown in Fig. 3. Although the

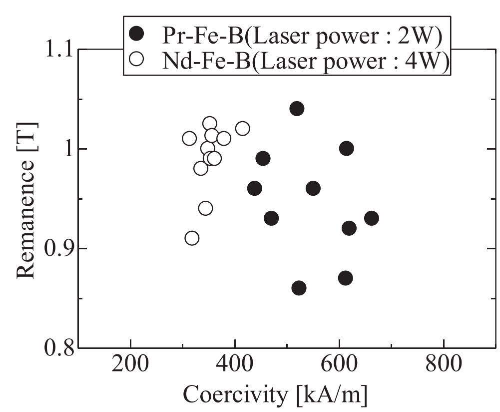


FIG. 3. Coercivity and remanence values of annealed Pr-Fe-B and Nd-Fe-B/α-Fe thick-films. The Pr-Fe-B films were prepared by using the laser energy density at approximately 50 mJ/m$^2$ using the laser power of 2 W. The plots of Nd-Fe-B ones were also shown in Fig. 1. The remanence values of Pr-Fe-B thick-films fluctuated between 0.85 and 1.05 T, however the values of coercivity were higher compared with those of Nd-Fe-B/α-Fe thick-films.

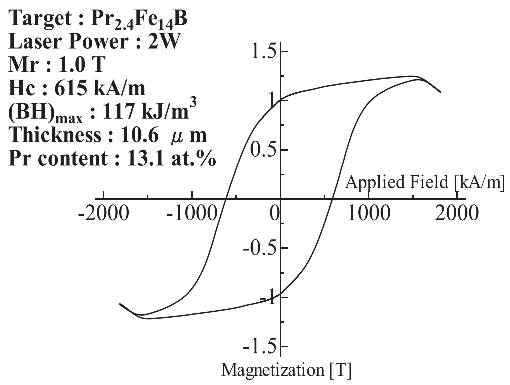


FIG. 4. In-plane M-H loop of an annealed Pr-Fe-B film which had the largest $(BH)_{max}$ value in the experiment. The film was prepared by using the laser energy density at approximately 50 mJ/mm$^2$ using the laser power of 2 W. ($Pr_{2.4}Fe_{14}B$ target).

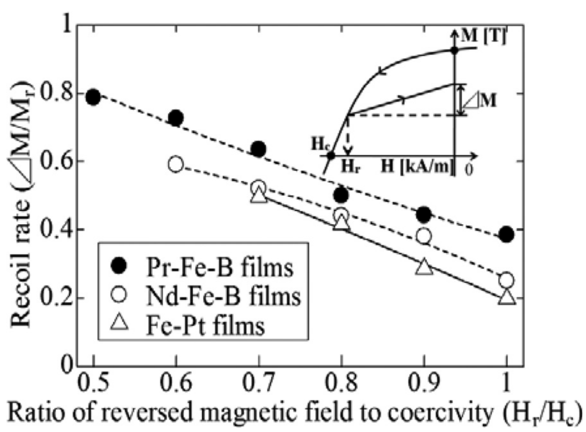


FIG. 5. Recoil rates of Pr-Fe-B, Nd-Fe-B/α-Fe nano-composite, and Fe-Pt thick-films prepared by using the PLD method. As shown in the inset figure, the horizontal and vertical axes showed a ratio of reversed magnetic field to coercivity and a recoil rate, respectively.

values of remanence fluctuated from 0.85 to 1.05 T, almost all the coercivity values of the thick-films exceeded 400 kA/m. We also confirmed $(BH)_{max}$ values became higher than 90 kJ/m$^3$. Figure 4 shows a M-H loop of a sample which had the largest $(BH)_{max}$ through the experiments. In order to evaluate the exchange interaction, a recoil rate of the above-mentioned Pr-Fe-B thick-film magnet was measured. Here, the rates of previously reported PLD-fabricated Nd-Fe-B/α-Fe[12] and Fe-Pt films[16] were also plotted in Fig. 5. It was found that the recoil behavior of the Pr-Fe-B thick film is superior to those of other PLD-made films together with that of a nano-composite bulk magnet with $Nd_2Fe_{14}B$ and α-Fe phases.[15]

From the above results, it is considered that the grain refinement of approximately 20 nm together with the good recoil rate in a Pr-Fe-B film with similar stoichiometric composition enables us to obtain the exchange interaction between the hard magnetic phase of $Pr_2Fe_{14}B$. Resultantly, we succeeded in achieving the large coercivity and the high $(BH)_{max}$ value.

## IV. CONCLUSION

Use of a Pr-Fe-B target together with an optimum laser condition enabled us to obtain isotropic thick-film magnets with superior magnetic properties compared with those of previously reported rare-earth based thick-films by using the PLD method with the deposition rate higher than 10 $\mu$m/h. In particular, the reduction in the laser power from 4 to 2 W leaded to suppress the heating of a substrate temperature and to obtain amorphous structure of an as-deposited Pr-Fe-B film. Resultantly, the coercivity and $(BH)_{max}$ of the samples exceeded 600 kA/m and 115 kJ/m$^3$, respectively, after a post-annealing. The origin of the good properties of a post-annealed film is considered to be attributed to the exchange interaction between $Pr_2Fe_{14}B$ hard grains.

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