Enhancement in coercivity of pulse-laser-deposition-made Nd–Fe–B thick film magnets by high-speed crystallization

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In order to improve the magnetic properties of Nd–Fe–B thick film magnets prepared by pulsed laser deposition, high-speed crystallization method of a pulse annealing was adopted as a postannealing process. In the case of using a $Nd_{2.4}Fe_{14}B$ target, the pulse annealing under an optimum condition for the period of 1.8 s enabled us to enhance the coercivity by approximately 300 kA/m compared with the average value of samples annealed by a conventional method. The obtained sample whose composition was almost as same as the target's one had the large coercivity of approximately 1300 kA/m, and values of remanence and $(BH)_{max}$ were 0.62 T and 64 kJ/m^3, respectively. It was also clarified that use of a $Nd_{2.2}Fe_{14}B$ target together with the pulse annealing method is effective to increase remanence and $(BH)_{max}$ although the coercivity value decreased. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2712818]

I. INTRODUCTION

We have already reported the fabrication of isotropic Nd–Fe–B thick film magnets, which have coercivity, remanence, and $(BH)_{max}$ values of approximately 1000 kA/m, 0.6 T, and 60 kJ/m^3, respectively, prepared by the pulse laser deposition (PLD) method with a high deposition rate larger than 20 μm/h.[1,2] Further, application of a 200-μm-thick Nd–Fe–B film on a Fe substrate to a millisize motor was carried out as a practical usage, and it was clarified that the rotational speed and torque constant under no-load test are 15 160 rpm and 0.0236 mNm/A, respectively, at the gap of 0.1 mm between a rotor and a stator. Advancement in micromachines such as small motors strongly depends on further improvement in magnetic properties of film magnets.

According to the observation on microstructure in the previously reported films,[3] the size of $Nd_2Fe_{14}B$ grains varied widely from 5 to 440 nm, and the average grain size was estimated at 150 nm, which suggests that refinement in grain size is a key technology to improve magnetic properties of films. It has been reported that high-speed crystallization is effective to obtain homogeneous microstructure, and resultantly to increase remanence and coercivity for rare earth hard magnetic ribbons and sputtered films.[4,5]

In this article, we adopted a pulse annealing as a high-speed crystallization method and systematically investigated the relationship between magnetic properties and the pulse annealing conditions.

II. EXPERIMENTAL PROCEDURE

In order to compensate loss of metallic Nd due to oxidization, the nominal compositions of the targets were set to $Nd_{2.2}Fe_{14}B$ and $Nd_{2.4}Fe_{14}B$. They were ablated with a Nd:YAG (yttrium aluminum garnet) pulse laser (λ = 355 nm) at the repetition rate of 30 Hz, and the films were deposited on Ta substrates. The distance between a target and a substrate was fixed at 10 mm. Before the ablation, the chamber was evacuated down to approximately 10^{-5} Pa with two molecular turbo pumps. In addition, a Ti sublimation pump was used as an auxiliary pump during the deposition. As-deposited films were amorphous, and, therefore, they were crystallized by a pulse annealing for 1.3–4.0 s with an infrared furnace at output power of 8 kW, and then they were cooled down to room temperature.

After a sample was magnetized up to 7 T using a pulsed magnetic field by a magnetization meter, magnetic properties were measured with a vibrating sample magnetometer (VSM), which could apply a magnetic field up to approximately 1800 kA/m reversibly. All postannealed films were isotropic; therefore, in-plane magnetic properties were shown in this article. The analysis of crystal structure was carried out with an x-ray diffractometer (XRD), and the average thickness was estimated from hysteresis loops of as-deposited films as described in the previous paper.[1] In this report, the thickness range of samples was from 20 to 40 μm.

III. RESULTS AND DISCUSSION

After the pulse annealing during the annealing period T_p, the magnetic properties and the x-ray diffraction patterns of samples were evaluated, and the results were shown in Figs. 1 and 2. Here, the samples were prepared by using a $Nd_{2.4}Fe_{14}B$ target. The dotted lines in each figure show the average values of samples fabricated by a conventional annealing method whose annealing temperature, heating rate, and holding time were 923 K, 400 K/min, and 0 min, respectively.[1] In the annealing conditions of 1.4 s $\leqq T_p$ and 3 s $\leqq T_p$, the films did not exhibit hard magnetic properties, and resultantly it was found that T_p = 1.8 s is the optimum

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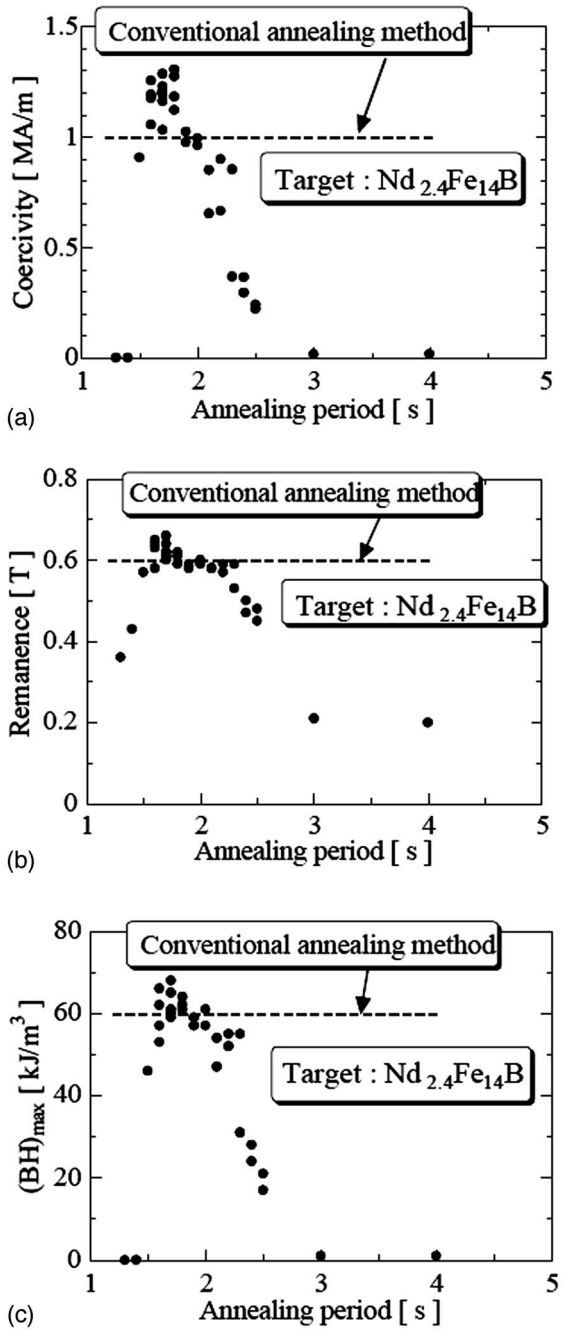

FIG. 1. Coercivity, remanence, and $(BH)_{max}$ of PLD-made Nd–Fe–B thick film magnets prepared by pulse annealing as a function of annealing period range from 1.3 to 4.0 s. These samples were prepared by using a $Nd_{2.4}Fe_{14}B$ target. Annealing period of 1.8 s led to the maximum coercivity value of approximately 1300 kA/m.

annealing period in our experiment. As seen in Fig. 2, a film prepared by T_p=1.4 s had no diffraction peaks originating from crystal structure and a film prepared by T_p=4 s had a lot of unknown peaks. The structures of the samples are consistent with the magnetic properties displayed in Fig. 1.

Figures 3 and 4 show two *M-H* loops and x-ray diffraction patterns of films crystallized by a pulse annealing and a conventional annealing process. The samples in each figure were prepared by using a $Nd_{2.4}Fe_{14}B$ target. Crystallization by pulse annealing for 1.8 s resulted in enhancement of coercivity by 300 kA/m compared with average one of samples annealed by a conventional method. It was also found that a slight increase in remanence and $(BH)_{max}$ at T_p of 1.6–1.8 s agrees with that previously reported.[4,5] The *M-H* loop of the pulse-annealed sample shown in Fig. 3 was not symmetries. This is attributed to the fact that the coercivity value of the sample is approximately 1300 kA/m and it is, therefore, difficult to magnetize the sample sufficiently during the measurement of the loop, which means that the magnetization processes are different between (1) from +1800 kA/m (a starting point) to −1800 kA/m and (2) from −1800 kA/m to +1800 kA/m because a sample is only fully magnetized at the starting point by using a pulse magnetic field of 7 T. In addition, the diffraction pattern of a pulse-annealed film indicated the reduction in the peaks originating from Nd_2O_3 and α-Fe phases compared with those of diffraction pattern of a sample annealed by a conventional method. That is, the pulse annealing is effective to suppress surface oxidization during an annealing process because of the extremely short annealing time.

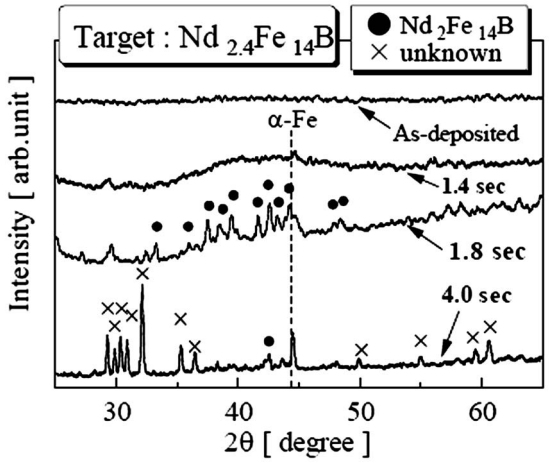

FIG. 2. X-ray diffraction patterns of PLD-made Nd–Fe–B thick film magnets prepared by a pulse annealing process under the various annealing period. These samples were prepared by using a $Nd_{2.4}Fe_{14}B$ target.

From these results, it was found that an optimum pulse annealing enables us to obtain films with coercivity larger than 1300 kA/m and to suppress the oxidization. In Refs.

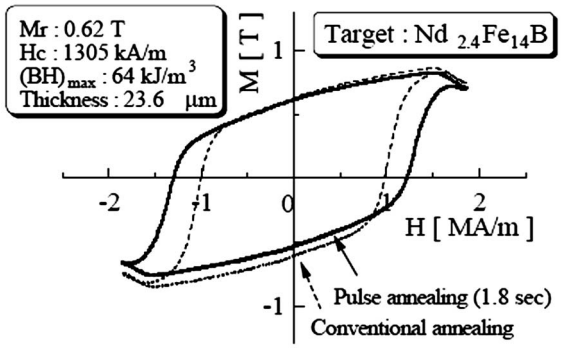

FIG. 3. *M-H* loops of PLD-made Nd–Fe–B thick film magnets crystallized by two methods of the pulse annealing and a conventional annealing process. These samples were prepared by using a $Nd_{2.4}Fe_{14}B$ target. Coercivity of the film pulse annealed for 1.8 s was lager by approximately 300 kA/m compared with that annealed by a conventional method.

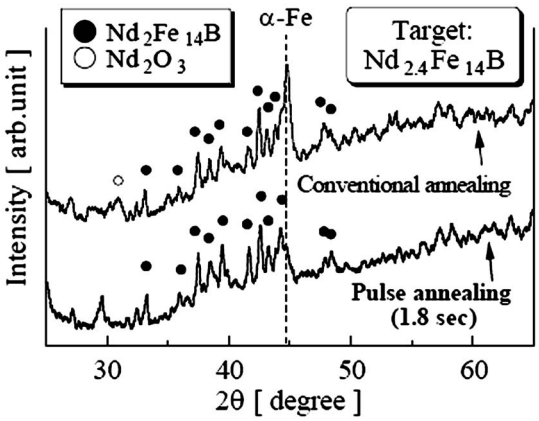

FIG. 4. X-ray diffraction patterns of PLD-made Nd–Fe–B thick film magnets after crystallization by two methods of the pulse annealing and a conventional annealing process. These samples were prepared by using a $Nd_{2.4}Fe_{14}B$ target.

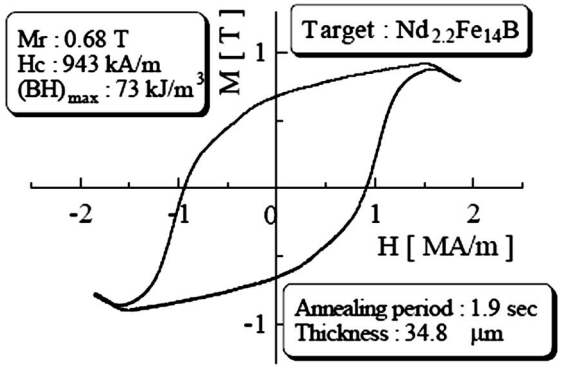

FIG. 5. An *M-H* loop of PLD-made Nd–Fe–B thick film magnet by using a $Nd_{2.2}Fe_{14}B$ target followed by pulse annealing for 1.9 s. The obtained remanence and $(BH)_{max}$ values were lager than those prepared by using a $Nd_{2.4}Fe_{14}B$ target.

1–3, the nominal compositions of the targets were set to $Nd_{2.4}Fe_{14}B$ in order to compensate the loss of metallic Nd due to oxidization. Here, we attempted to decrease the amount of Nd in a target by taking advantage of the increasing effect of coercivity and the reduction of surface oxidization in the pulse annealing. An *M-H* loop of a PLD-made film prepared by a $Nd_{2.2}Fe_{14}B$ target followed by pulse annealing is shown in Fig. 5. A film with coercivity, remanence, and $(BH)_{max}$ of 943 kA/m, 0.68 T, and 73 kJ/m^3, respectively, could be obtained at T_p of 1.9 s. Although coercivity decreased compared with that of the films fabricated by ablating a $Nd_{2.4}Fe_{14}B$ target, enhancement in remanence and $(BH)_{max}$ was successfully achieved. Further investigation is required for an optimum relationship between a target composition and pulse annealing conditions.

IV. CONCLUSION

A pulse annealing process was adopted to obtain PLD-prepared Nd–Fe–B thick film magnets with high coercivity larger than 1300 kA/m. In PLD method followed by the pulse annealing process, reduction in amount of Nd in a target was effective to enhance remanence and $(BH)_{max}$. Further investigations on the relationship between the pulse annealing conditions and target compositions are required to obtain PLD-prepared Nd–Fe–B films with superior magnetic properties.

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