Journal of Physics: Conference Series

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To cite this article: R Tokumaru *et al* 2009 *J. Phys.: Conf. Ser.* **191** 012021

Effect of annealing on magnetic properties of Nd-Fe-B thin films prepared by ECR ion beam sputtering method

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Abstract. Nd-Fe-B thin films were prepared by electron cyclotron resonance (ECR) ion beam sputtering and subsequent annealing. The influence of annealing on the magnetic properties and X-ray diffraction patterns of the product films was investigated. Amorphous films deposited at room temperature were annealed at temperatures between 600 and 800 °C. The *c*-axis oriented crystallization of the $Nd_2Fe_{14}B$ phase did not appear by annealing of the buffer layer and magnetic Nd-Fe-B layer deposited at room temperature, and the hysteresis loops of the films indicated magnetic isotropy.

1. Introduction

Recently, Nd-Fe-B permanent magnets have attracted much attention due to their excellent intrinsic properties, such as high saturation magnetization, crystalline magnetic anisotropy, and high coercivity. Thin films with *c*-axis orientation have been studied for their potential applications in microelectronic mechanical systems (MEMS) and magnetic recording media. Preparation of thin films with excellent magnetic properties requires the formation of a well-textured $Nd_2Fe_{14}B$ phase with *c*-axis orientation that is preferentially aligned normal to the film plane, and the grain size of the $Nd_2Fe_{14}B$ phase in the film should be maintained to a single domain size of around 30 nm [1,2]. Two methods are considered for the crystallization of thin films; direct crystallization during film deposition, and crystallization of the as-deposited amorphous films by annealing. The first method has been widely studied, because such films easily acquire magnetic anisotropy perpendicular to the film plane; however, this method may cause the over-growth of grains exceeding a single domain size, which results in degradation of the magnetic properties. On the other hand, grains of single domain size may be obtained using the latter method by control of the film composition and annealing conditions [3]. In this study, the effect of the annealing process on the magnetic properties of Nd-Fe-B thin films was investigated.

2. Experimental procedures

Nd-Fe-B films were prepared using an originally assembled electron cyclotron resonance (ECR) ion beam sputtering system with a 5-cm diameter $Nd_{36.5}Fe_{51.1}B_{12.4}$ alloy target sintered by spark plasma sintering. The background pressure used was below 4.0×10^{-4} Pa and the Ar pressure for sputtering was maintained at 6.0×10^{-3} Pa during film deposition. The following ECR ion beam sputtering conditions were used, acceleration voltage of 1 kV, microwave power output of 100 W, and plasma flux density of 1.2–1.5 mA/cm^2. Amorphous Nd-Fe-B films (100 nm thick) were deposited on Si(100) substrates. W layers were used as buffer (50 nm) and passivation (30 nm) layers. All layers were deposited at room temperature. The as-deposited films were annealed at temperatures between 600 and 800 °C in a tube furnace with Ar gas flow. The O_2 partial pressure was controlled in the range between 10^{-17} and 10^{-19} Pa-O_2 during film annealing using an O_2 partial pressure controller to avoid oxidation of the magnetic layer. The crystal structure was identified using X-ray diffraction (XRD) with Cu $K\alpha$ radiation. The magnetic properties were measured using a vibrating sample magnetometer (VSM) with a maximum field of 20 kOe.

3. Results and Discussion

XRD patterns of the (a) as-deposited film, and films annealed at (b) 600, (c) 700, and (d) 750 °C for 30 min after deposition are presented in Fig. 1. The as-deposited film pattern did not display diffraction peaks assignable to the magnetic layer; only a broad beta-W peak was observed, which implies the magnetic layer is amorphous after deposition at room temperature. The patterns of the post-annealed films did not indicate peaks of the $Nd_2Fe_{14}B$ phase, although diffraction peaks from the W(110), (220) and (211) planes and impurity phases such as Nd-O and a Nd-rich phase, possibly a binary Fe–Nd compound [4], were observed.

Magnified (2θ = 36–44°) XRD patterns of the films annealed at (a) 600 and (b) 700 °C for 30 min are shown in Fig. 2. Weak diffraction lines were confirmed for the film annealed at 700 °C, in contrast to that annealed at 600 °C. However, *c*-axis orientation crystallization of the $Nd_2Fe_{14}B$ phase was not expected, because peaks assignable to the $Nd_2Fe_{14}B(00l)$ phase were not observed.

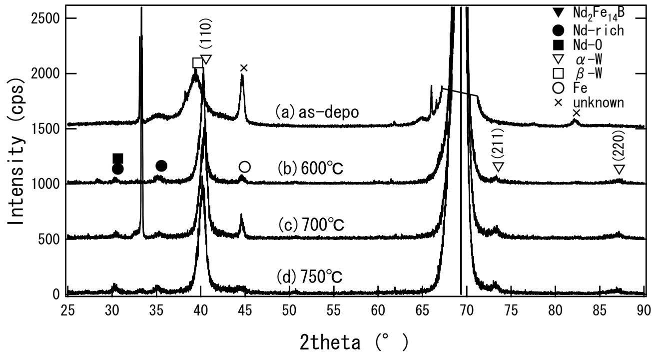

Figure 1. XRD patterns of the (a) as-deposited film and films annealed at (b) 600, (c) 700, and (d) 750 °C for 30 min after deposition at room temperature.

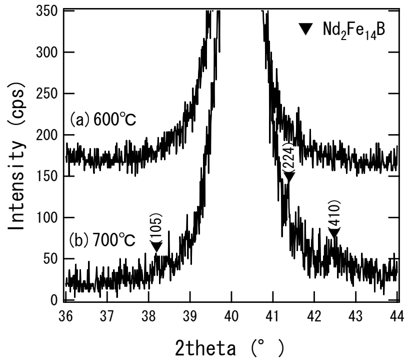

Figure 2. Magnified XRD patterns of the films annealed at (a) 600 and (b) 700 °C.

Magnetic hysteresis loops of the (a) as-deposited film and the films annealed at (b) 600, (c) 700, and (d) 750 °C for 30 min are shown in Fig. 3. Measurements were carried out at room temperature with applied fields perpendicular (⊥) to and parallel (||) to the film plane. No demagnetization correction was carried out for the measurements. The hysteresis loops of the as-deposited film and the film annealed at 600 °C indicate soft magnetic character. The loop of the film annealed at 600 °C indicates higher coercivity than that of the as-deposited film (almost zero). Although no obvious peaks of the Nd-Fe-B phase were observed in the XRD patterns, the coercivity seems to be due to the partial formation of the Nd-Fe-B phase in the film annealed at 600 °C. With increase in the annealing temperature, the hysteresis loops of the films exhibit hard magnetic behavior for the samples annealed between 700–775 °C. Considering the XRD pattern in Fig. 1, this seems to be due to crystallization of the $Nd_2Fe_{14}B$ phase. However, for the films annealed at 700 and 750 °C (Figs. 3(c) and (d), respectively) no obvious difference was observed in the hysteresis loops recorded with magnetic fields parallel and perpendicular to the film plane. This is consistent with the lack of XRD peaks that would indicate the $Nd_2Fe_{14}B$ (00*l*) plane meaning the degree of c-axis orientation. The magnetic properties of coercivity and remanent magnetization were decreased with increase of the annealing temperature over 700 °C. Additionally, large steps are present in the hysteresis loop around the applied field of H=0, measured with both directions of applied field. These steps are due to the magnetic reversal of the soft magnetic phase. It is possible that both the soft and hard magnetic phases are present in the magnetic layer, and that exchange coupling between hard and soft grains is poor.

The respective dependency of coercivity and remanent magnetization on the annealing temperature is shown in Figs. 4 and 5. The sample annealed at 600 °C shows low coercivity, which is reflected by poor crystallization of the hard magnetic phase in the film. The increase of coercivity with further increase of the annealing temperature (650–775 °C) indicates crystalline growth of the magnetic (Nd-Fe-B) phase. The highest coercivity of 8.2 kOe (653 kA/m) was obtained by annealing at 700 °C, with remanent magnetization of 3.4 kG (0.34 T). Annealing of samples above 800 °C resulted in the deterioration of hard magnetic grains, and consequently a non-magnetic phase. The same trend was observed for the remanent magnetization with variation in the annealing temperature, except for the sample annealed at 600 °C, which reveals the presence of the soft magnetic (α-Fe) phase, as observed by XRD.

The results indicate no significant difference in coercivity and remanent magnetization between the parallel and perpendicular directions, even in the samples annealed at 675 and 700 °C, the XRD patterns of which indicated the presence of the Nd-Fe-B phase. This indicates the difficulty of obtaining the *c*-axis orientated $Nd_2Fe_{14}B$ films perpendicular to the film plane using the annealing method. In order to obtain anisotropic *c*-axis textured Nd-Fe-B films, some research groups have applied the film growth technique, which involves deposition at an intermediate temperature between the crystallization between the crystallization temperature and sputtering temperature and sputtering temperature[3,5,6]. Even with a deposition temperature below the temperature required to crystallize Nd-Fe-B phase, thermal assistance during the deposition process may play an important role for the alignment of Nd-Fe-B magnetic grains.

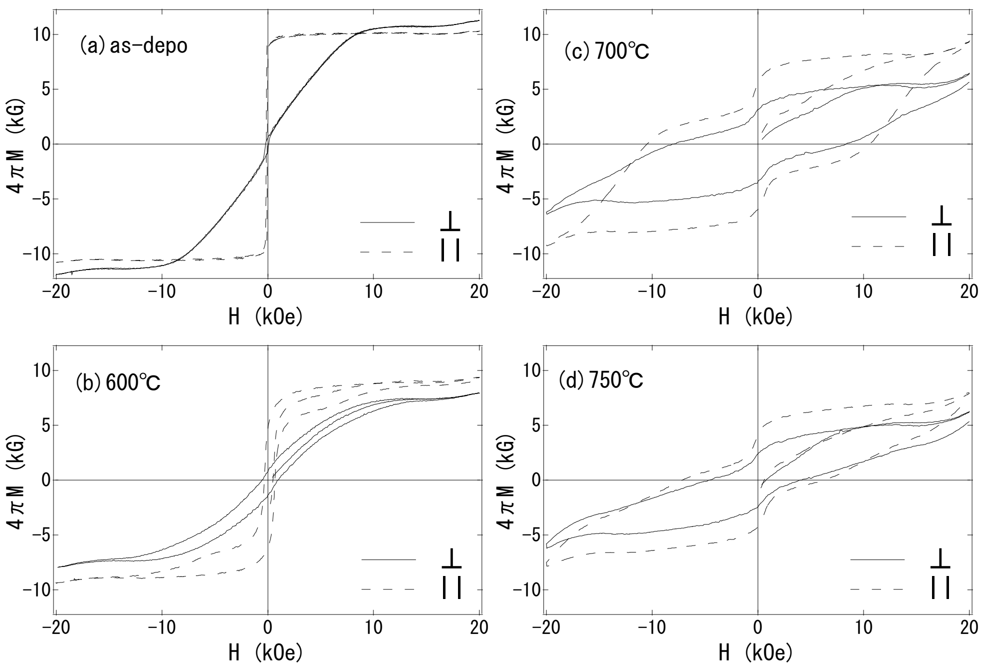

Figure 3. Magnetic hysteresis loops of the (a) as-deposited film, and films annealed at (b) 600, (c) 700, and (d) 750 °C for 30 min after deposition at room temperature. No demagnetization correction was carried out for these measurements.

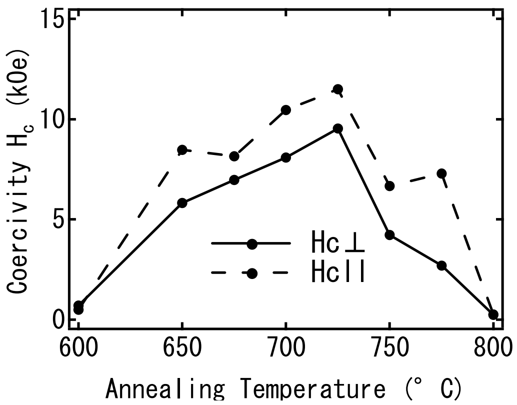

Figure 4. Dependence of the coercivity of Nd-Fe-B thin films on the annealing temperature.

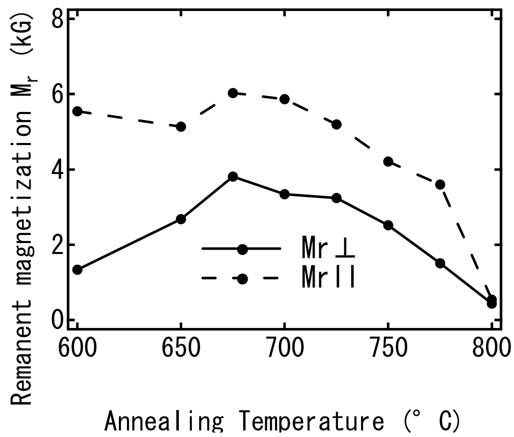

Figure 5. Dependence of the remanent magnetization of Nd-Fe-B thin films on the annealing temperature.

4. Conclusion

Nd-Fe-B thin films were prepared by ECR ion beam sputtering and crystallized by annealing. The influence of annealing temperature on the magnetic properties of the films was investigated. Hard magnetic behavior resulted from annealing temperatures of 650–775 °C. However, the *c*-axis texture of the $Nd_2Fe_{14}B$ phase perpendicular to the film plane was not achieved by crystallization of the buffer layer and magnetic layer deposited at room temperature and subsequent annealing. Hysteresis loops of the prepared films did not indicate perpendicular anisotropic behavior along the *c*-axis. A magnetic Nd-Fe-B film with coercivity of 8.2 kOe (653 kA/m) and remanent magnetization of 3.4 kG (0.34 T) was obtained by annealing at 700 °C.

Acknowledgement

This study was supported by the "Academic Frontier" Project for Private Universities and a matching fund subsidy from the Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2006–2010.

References

[1] Helbest J F 1991 *Rev. Mod. Phys.* **63** 819

[2] Okuda T 2003 *Magn. Soc. Jpn.* **27** 1007

[3] Okuda T, Sugimura A, Eryu O, Serrona L K E B, Adachi N, Sakamoto and Nakanishi A 2003 *Jpn. J. Appl. Phys.* **42** 6859

[4] Chen S.L., W. Liu, C.L. Chen, and Zhang Z.D 2005 *J.Appl.Phys.***98** 033907

[5] Serrona L K E B, Sugimura A, Adachi N, Okuda T, Nakanishi, Motokawa M, Ping D H and Hono K, 2003Appl. Phys. Lett. **82** 1751

[6] Tang S L, Gibbs M R J, Davies H A, Liu Z W, Lane S C, Mateen N E, Du Y W, 2007 *J. Appl. Phys*. **101** 09K501