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Phase Formation and Magnetic Properties of $Nd_2Fe_{14}B$ Film Magnets by Using the Laser Ablation Technique

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Abstract

$Nd_2Fe_{14}B$ film magnets were synthesized by using the laser ablation technique. For the purpose of development of superior magnetic thin films, the phase formation and magnetic properties according to the variation of the film thickness, the Ta substrate, and the Ti sublimation pump were investigated. The experimental result indicated one method to prepare $Nd_2Fe_{14}B$ films with superior magnetic properties was to synthesize a NdFeB film with a thickness of 5μm on a Ta substrate in a chamber with a Ti sublimation pump.

1. INTRODUCTION

Nowadays, thin film magnets have attracted much attention because they are expected to improve the characteristics and/or to reduce the size of electronic devices, such as circulators, isolators, magnetostrictive heads, and micromachines. Lately most of the thin-film magnets, rare-earth thin-film magnets, and rare-earth nanocomposite thin-film magnets have been synthesized using the sputtering technique or the laser ablation technique. [1-3]

We expect to get superior thin film magnets by using the laser ablation technique because that technique has some advantages over the sputtering technique, such as good reflection of the target composition on the prepared film, high preparation speed, freedom of gas pressure in the chamber during preparation, and simple apparatuses in the chamber. In spite of the advantages, thin films synthesized using the laser ablation technique do not show magnetic properties as good as those of thin films synthesized using the sputtering technique because the detailed effects of the preparation conditions of thin-film magnets by using the laser ablation technique on their magnetic properties have not yet been clarified. In Table 1, the coercivities of thin-film magnets synthesized using the sputtering and the laser ablation techniques are compared [4-7].

In addition, the application of this technique to synthesize NdFeB thin-film magnets has been reported little. Therefore, it is worthwhile to synthesize NdFeB film magnets using the laser ablation technique and to study the relation of their magnetic properties to their preparation conditions.

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Table 1. Comparison of magnetic properties in thin-film magnets synthesized using the sputtering and laser ablation techniques.

Film magnets	Coercivity [kOe]		Ref.
	Sputtering technique	Laser ablation technique	
$Nd_2Fe_{14}B$	20	1	(1)
Sm_2Co_{17}	20	11	(2)
$Sm_2Fe_{17}N_3$	23	6.5	(3, 4)

In this paper, we focus on the investigation of the phase formation and magnetic properties of $Nd_2Fe_{14}B$ film magnets synthesized using the laser ablation technique. The magnetic properties of the $Nd_2Fe_{14}B$ films can be improved by synthesizing films with a thickness of 5μm on a Ta substrate in a chamber with a Ti sublimation pump.

2. EXPERIMENT

We prepared targets with the composition of $Nd_2Fe_{14}B$ for synthesis of Nd-Fe-B film magnets, and the Nd-Fe-B films were deposited on SiO_2 and Ta substrates by ablating the target with a XeCl excimer laser. The target was rotated so that the laser beam did not ablate only one point.

The incident laser beam made an angle of 45° to the surface of the target. The laser had a wavelength of 308 nm, an average pulse energy of 3.5 J/cm^2, a frequency of 30 Hz, and an average power of 11 Watt. A Ti sublimation pump was used as an auxiliary pump during the ablation. In general, Ti has properties to combine with oxygens and Table 2 shows evacuation speed per unit area of the pump used in this research.

The deposition time, the substrate temperature, and the pressure in the chamber were 60 min, room temperature, and less than 1×10^{-6} Torr during deposition, respectively. The thickness of the films was controlled by varying the distance between the target and the substrate under a constant deposition time and energy density of the laser beam. The deposited films were annealed in a vacuum of 1×10^{-5} Torr at 650 ℃ for 60 min.

The thickness of the synthesized films was determined with a surface roughness meter, and magnetic properties were evaluated with a vibrating sample magnetometer. The temperature dependence of the magnetization was measured under a vacuum of 1×10^{-5} Torr at a heating rate of 5℃/min. The depth profile of the X-ray photoelecton spectrum was measured by etching the films with Ar ions, and the X-ray diffraction patterns were investigated.

3. RESULTS AND DISCUSSION

3. 1 Preparation of $Nd_2Fe_{14}B$ Films

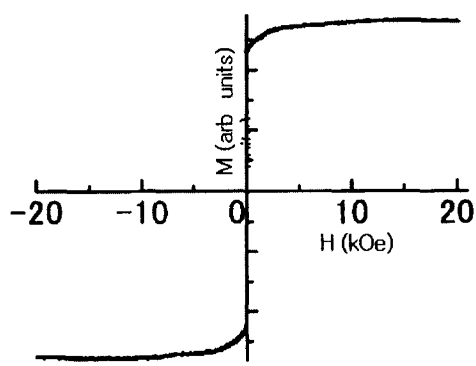

Fig. 1. Hysteresis loop for a film with a thickness of 1 μm deposited on a SiO_2 substrate at room temperature.

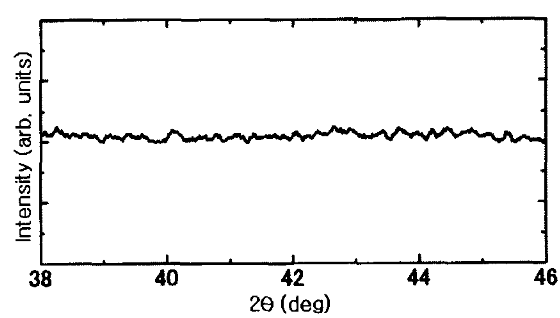

Fig. 2. X-ray diffraction pattern for a film with a thickness of 1 μm deposited on a SiO_2 substrate at room temperature.

The magnetization curve of a Nd-Fe-B film with a thickness of 1 μm deposited on a SiO_2 substrate at room temperature is shown in Figure 1. As shown in Fig. 1, the film had a very small coercivity of 17.9 Oe. In order to study the reason for the small coercivity, we investigated the crystal structure of the film from an X-ray diffraction pattern, and the result is shown in Fig. 2. This figure shows no clear peak in the range of 2θ from 38 to 46°. Thus, we conclude that the as-deposited Nd-Fe-B film synthesized on the SiO_2 substrate at room temperature with the laser ablation technique did not crystallize.

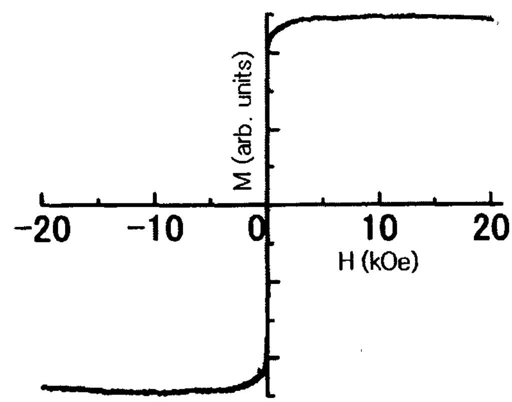

Fig. 3. Hysteresis loop for a film with a thickness of 1 μm deposited on a SiO_2 substrate and annealed in a vacuum of 1×10^{-5} Torr at 650℃ for 60 min .

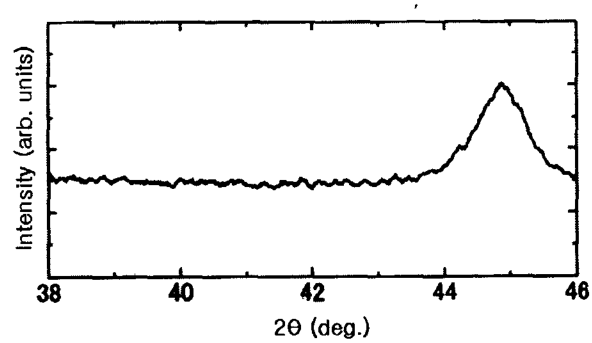

Fig. 4. X-ray diffraction pattern for a film with a thickness of 1 μm deposited on a SiO_2 substrate and annealed in a vacuum of 1×10^{-5} Torr at 650℃ for 60 min.

We annealed the as-deposited film in order to crystallize the film, and Figs. 3 and 4 show the magnetization curve and the X-ray diffraction pattern of the annealed film, respectively. The thermally treated film had a very small coercivity of 38.9 Oe, as shown in Fig. 3 and a clear peak at near 2θ=44.7° which corresponded to the α-Fe phase, as shown in Fig. 4. To confirm the composition of the thermally treated Nd-Fe-B film, we measured the X-ray photoelectron spectra, and the results showed that the amounts of Nd, Fe, and B are almost consistent with corresponding target.

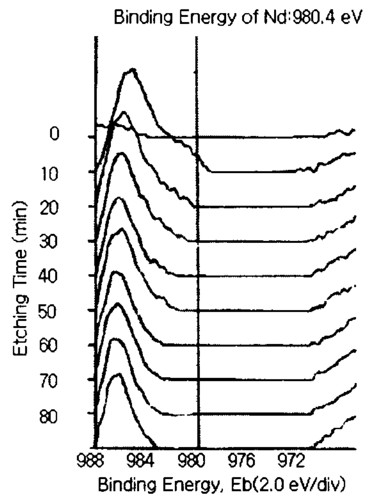

Fig. 5. Binding energy curves for a film with a thickness of 1 μm deposited on a SiO_2 substrate and annealed in a vacuum of 1×10^{-5} Torr at 650℃ for 60 min.

However, Fig. 5 shows that the X-ray photoelectron spectrum measured by etching the films with Ar ions was shifted to the left compared with 980.4 eV corresponding to the binding of metallic Nd [8], which suggests that the Nd in the film was oxidized.

3. 2. Effects of Film Thickness, Ta Substrate, and Ti Sublimation Pump

From the above result, the prevention of oxidation of Nd is thought to be very important to synthesize Nd-Fe-B films with superior magnetic properties. Therefore, we investigated the effects of the film thickness, the Ta substrate, and the Ti sublimation pump on the oxidation in Nd-Fe-B films. We have already reported that the $Sm_2Fe_{17}N_3$ film with a thickness of 5 μm had magnetic properties superior to those of the film with a thickness of 0.2 μm and that a $Sm_2Fe_{17}N_3$ film deposited on a Ta substrate had superior magnetic properties compared with films deposited on the other substrates studied, such as SiO_2, Si, Mo, and W [8].

There was some oxygen in the chamber although a high vacuum was kept during the deposition, and the oxygen became the origin of oxidation of metallic Nd. In order to reduce the amount of oxygen, we introduced a Ti sublimation pump into the chamber. Ti evaporated by heating the Ti sublimation pump is expected to interact with the oxygen, and, as the result, the oxidation of Nd will be suppressed. Accordingly, we undertook to prepare films with thicknesses of 5 μm on Ta substrates in a chamber with a Ti sublimation pump to synthesize Nd-Fe-B films with superior magnetic properties.

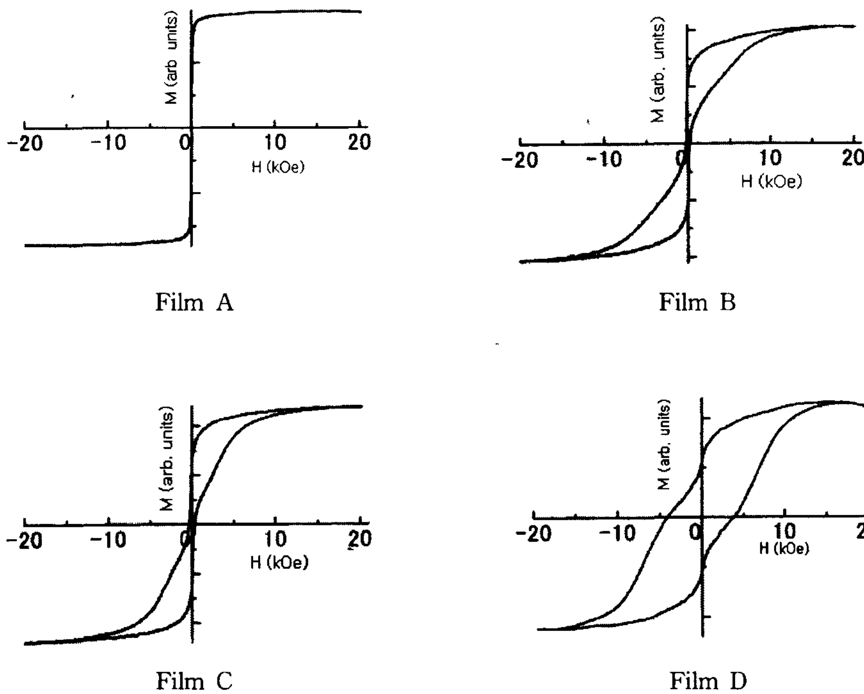

Fig. 6. Magnetization curves for films with a thickness of 5μm heated at 650℃ for 60 min. Film A was deposited on a SiO_2 substrate without a Ti sublimation pump, film B was deposited on a Ta substrate without a Ti sublimation pump, film C was deposited on a SiO_2 substrate with a Ti sublimation pump, and film D was deposited on a Ta substrate with a Ti sublimation pump.

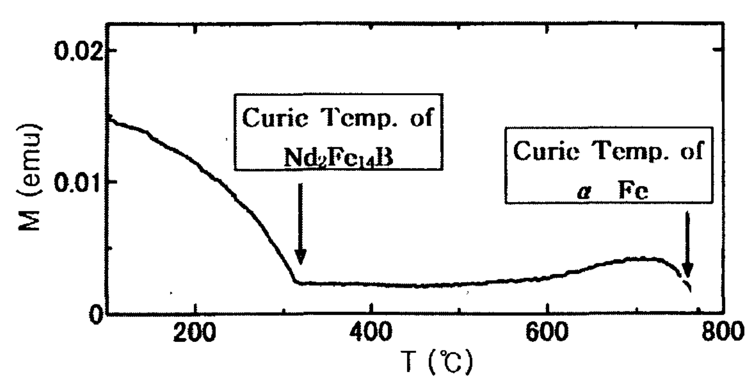

Fig. 7. Temperature dependence of the magnetization for a film heated at 650℃ for 60 min with a thickness of 5μm deposited on a Ta substrate in a chamber with a Ti sublimation pump.

Figure 6 shows magnetization curves for films deposited under various conditions. Film A was deposited on a SiO_2 substrate without a Ti sublimation pump, film B was deposited on a Ta substrate without a Ti sublimation pump, film C was deposited on a SiO_2 substrate with a Ti sublimation pump, and film D was deposited on a Ta substrate with a Ti sublimation pump. As seen in fig. 6, Film D had superior magnetic properties compared with the other films, which suggests the Ta substrate and the Ti sublimation pump affect the suppression of the oxidation in Nd-Fe-B films. To confirm the suppression of the oxidation in Nd-Fe-B films due to the usage of the Ta substrate and the Ti sublimation pump, we compared X-ray photoelectron spectra by etching the surface with Ar ions for the film corresponding to Fig. 1 and for film D. The result showed that the spectral peaks of film D shifted to the left compared with 980.4 eV which corresponds to the binding energy of metallic Nd [10], as mentioned in section III-1; however, the peaks shifted less than the peaks of the film corresponding to Fig. 1. These facts suggest that the Ta substrate and the Ti sublimation pump suppressed the oxidation of metallic Nd, and, as a result, film D deposited on a Ta substrate in the chamber with a Ti sublimation pump was less oxidized than the film deposited on a SiO_2 substrate without a Ti sublimation pump.

The detailed role of Ta in preventing oxidation is not clear at present. Considering that Ta has an affinity for oxygen, however, a Ta substrate may absorb oxygen, and thus, prevent the oxidation of metallic Nd in the Nd-Fe-B film.

Also, we measured temperature dependence of the magnetization to confirm the phase of film D. The result is shown in Fig. 7. This figure shows two sharp decreases; one at 315℃ which corresponds to the Curie temperature of the $Nd_2Fe_{14}B$ phase and the other at 770℃ which corresponds to the Curie temperature of the α-Fe phase. From these facts, we conclude that the prepared film had both $Nd_2Fe_{14}B$ and α-Fe phases and that the presence of $Nd_2Fe_{14}B$ phase was responsible for the increased coercivity. Thus, we conclude that using the laser ablation technique to prepare films with a thicknesses of $5\mu m$ on Ta substrates in a chamber with a Ti sublimation pump is one method to improve the magnetic properties of NdFeB films.

4. CONCLUSIONS

The $Nd_2Fe_{14}B$ film magnets were successfully synthesized by the laser ablation technique, and, the effects of the film thickness, the Ta substrate, and the Ti sublimation pump on oxidation in Nd-Fe-B films were investigated. Though the $Nd_2Fe_{14}B$ film with a thickness of $5\mu m$ synthesized on a Ta substrate in a chamber with a Ti sublimation pump contained an α-Fe phase, that film contained $Nd_2Fe_{14}B$ as the main phase, and the presence of the $Nd_2Fe_{14}B$ phase was responsible for the observed coercivity of 4.6 kOe. Thus, one method of preparing $Nd_2Fe_{14}B$ films with superior magnetic properties is to synthesize a Nd-Fe-B film with a thickness of $5\ \mu m$ on a Ta substrate in a chamber with a Ti sublimation pump.

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