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Preparation and characterization of (Tb,Dy)Fe_2 giant magnetostrictive thin films for surface acoustic wave devices

H.H. Uchida
Department of Resources and Environmental Science, Tokai University, Hiratsuka, Kanagawa 259-12 (Japan)

V. Koeninger
Kanagawa Academy of Science and Technology (KAST), Kawasaki, Kanagawa 213 (Japan)

H. Uchida, M. Wada and H. Funakura
Department of Applied Physics, Tokai University, Hiratsuka, Kanagawa 259-12 (Japan)

Y. Matsumura
Kanagawa Academy of Science and Technology (KAST), Kawasaki, Kanagawa 213 (Japan)

T. Kurino and H. Kaneko
Society of Non-Traditional Technology (SNTT), Toranomon, Minato-ku, Tokyo 105 (Japan)

Abstract

Thin films of giant magnetostrictive (Tb,Dy)Fe_2 alloys were prepared by three different deposition processes: ion beam sputtering, ion plating and flash evaporation. The flash evaporation process readily yields films with little deviation in composition from that of the source materials, while the ion-beam-sputtering process results in a composition deviation which depends on the substrate location. The ion-plating process yields a high rate of deposition but leads to a marked composition variation between source and film materials. The deposition conditions, mainly the background pressure, also have an important influence on the film characteristics.

1. Introduction

Since a very large magnetostriction was first found in rare earth–transition metal alloys with an AB_2 cubic Laves phase [1], many applications of these giant magnetostrictive bulk materials have been proposed and tested, *e.g.* new types of ultrasonic sound generators, actuators, etc. [2].

One of the most specific characteristics of giant magnetostrictive materials (GMMs) is their elastic modulus change under an ambient magnetic field intensity, which results in changes in the acoustic sound velocity both in the bulk and on the surface. One possible useful application of this feature is the utilization of GMM thin films for constructing a surface acoustic wave (SAW) device [3–5] which enables the control of surface wave velocity by an external magnetic field, since conventional SAW devices using piezoelectric materials are not tunable.

In the actual development of such applications as a tunable GMM-SAW device, the preparation of a film with an optimum composition and structure is essential. For example, Hietala and Robbins [3] reported that a $Tb_{0.4}Fe_{0.6}$ film shows the largest striction and velocity change within the film composition range Tb_xFe_{1-x}, where $0.2<x<0.6$. However, film characteristics such as structure, composition and concentration of contamination, which may lead to drastic changes in physical properties, have been less investigated. Films prepared by different processes may have different characteristics.

In this paper giant magnetostrictive thin films were prepared by three different processes and characterized from the viewpoint of film structure, film composition and contamination. The magnetic properties of the films were also investigated.

2. Experimental details

GMM films (1–2 μm thick) were prepared by ion beam sputtering (IBS) [6], ion plating (IP) [7] and flash evaporation (FE). X-Ray diffraction analyses were carried out to determine the film crystallization. The film composition was investigated by inductively coupled plasma (ICP) emission spectroscopy. Auger electron

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spectroscopy (AES) was also used to measure the depth profile of each element and the film contamination.

2.1. IBS process

Films were prepared by Ar ion beam sputtering with an acceleration of 10 kV in an ultrahigh vacuum apparatus. The ion beam current density was 0.5–1 mA cm^{-2} as measured by the current through the target to ground. The target was set against the ion beam and the substrate at an angle of 45°. The substrate was located 120 mm above the target. The films were deposited on Si crystals and/or Kapton foils (75 μm thick) without cooling or heating. A sintered $Tb_{0.7}Dy_{0.3}Fe_{1.9}$ and cast $TbFe_2$ and $Tb_{0.3}Dy_{0.7}Fe_2$ targets were used. Because the sputter yields and distribution direction of each element (Tb, Dy and Fe) are different, the concentrations and deposition rates in different positions over the target were determined.

The impingement rates of residual gases on the deposited film surface were assumed to be proportional to the background pressure. The film contamination assessment can therefore be expressed for each process by the ratio $r = P_r/d_f$, where P_r is the background pressure and d_f is the film deposition rate. A lower background pressure and a higher deposition rate thus yield a less contaminated film. In the sputtering process deposition rates of 0.01–0.3 nm s^{-1} were obtained. The partial pressure of contaminating gas in Ar (99.999% purity) and the background pressure of the system were lower than 8×10^{-7} Pa. Therefore the ratio of the impingement of residual gas molecules to the deposition rate was in the approximate range 2.3×10^{-6}–7×10^{-5} Pa s nm^{-1}.

2.2. IP process

For the ion-plating process an electron beam evaporation system was used with a d.c. glow discharge generated by a positively biased electrode (40 V, 5 mA) located between the vapour source and the substrate. Block samples of $TbFe_x$ ($x = 0$–3) were used. The deposition rate and background pressure were 10–20 nm s^{-1} and lower than 2×10^{-4} Pa respectively. The ratio of the impingement of residual gas molecules to the deposition rate was calculated as about 10^{-5} Pa s nm^{-1}.

2.3. FE process

The flash evaporation process was also used for GMM film preparation. $TbFe_2$ and $DyFe_2$ powders, both pulverized by cyclic hydriding–dehydriding reactions [8], were dispersed on to a heated W plate (2800 K). The deposition rate was in the range 0.13–2.4 nm s^{-1}. The obtained value of r was between 2.9×10^{-4} and 1×10^{-2} Pa s nm^{-1}. Because the substrate was heated to as much as 650 K by th W plate, Kapton foil could not be used in this process. Foils of W, Cu and Si crystals were used as substrates.

2.4. Magnetic measurements

Magnetization curves were measured by a vibrating sample magnetometer (VSM). The sample films were set parallel and perpendicular to the magnetic field.

3. Results and discussion

3.1. Film structure

The crystallization of deposited films usually has a large effect on the magnetostriction and magnetic hardness. Generally the substrate temperature is a more influential factor in the crystallization than the type of deposition apparatus. Figure 1 shows the X-ray diffraction patterns of films prepared by IBS, IP and FE. It is seen that the films prepared by IBS and IP are amorphous like. In both processes the substrate temperature was near room temperature. On the other hand, when the substrate was refractory heated by the W plate, the crystallization of Laves and/or oxide phases was observed in the deposited films if the substrate was heated above 600 K. Substrates at a higher temperature, however, usually accelerate the reaction of residual gas molecules with the film surface.

Because the deposition rate of IBS is much lower than for the other processes, low stress in the film or between film and substrate can be expected. In fact, a film 1 μm thick on thin Kapton showed almost no deformation, while films produced by the IP process showed a large warp of the substrates.

3.2. Film composition

The advantage of film deposition is the possibility to form the intermetallic phase directly on the substrate, such as the $TbFe_2$ Laves phase which can normally only be formed through two-step peritectic reactions from the liquidus.

Film deposition using a sputtering process yields usually different deposition rates in different directions for each element. This results in films with different compositions at different substrate locations.

In Fig. 2 the most preferred position for RT_2 (R, rare earth; T, transition metal) film formation by the IBS process is seen to be with the substrate located directly above the target. In this position films with almost the same composition as the source materials were obtained. In any other position either Fe-rich films or very low deposition rates were obtained. In the IP process, because of the large difference in vapour pressure between rare earth metals and iron, which are both melted together by the electron beam, there is a difference in composition between source and film.

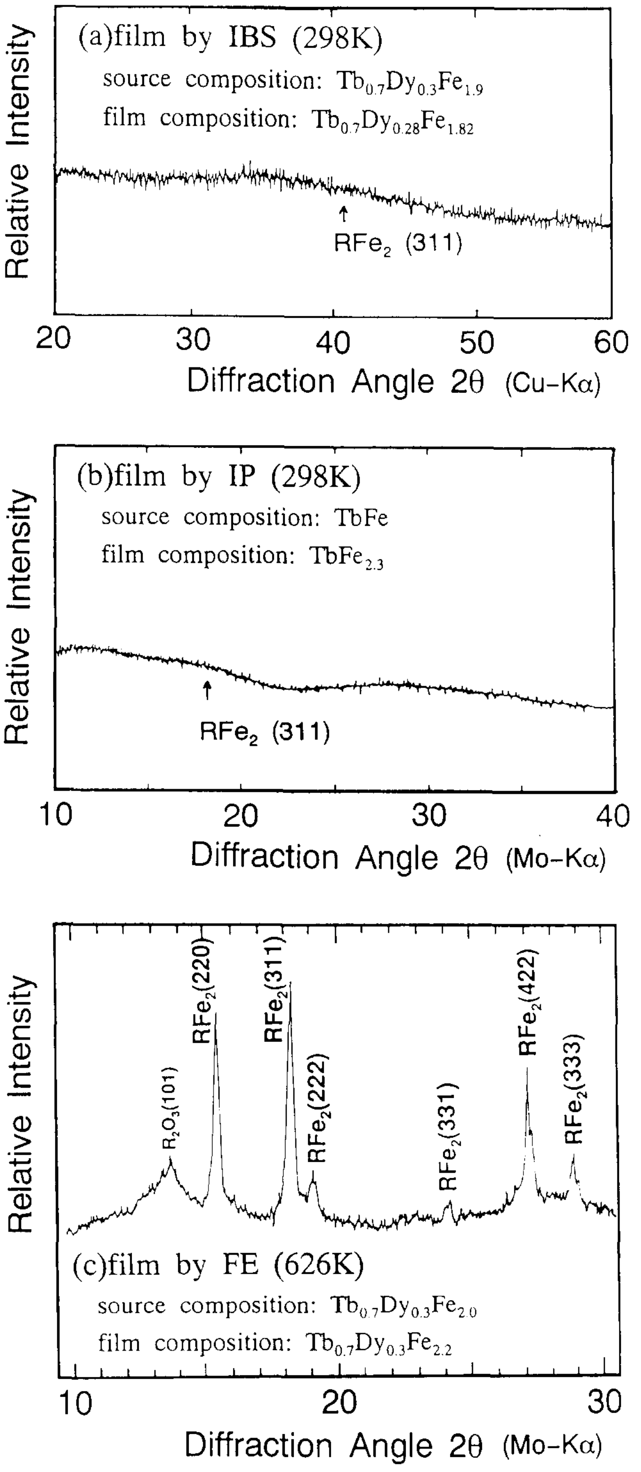

Fig. 1. Diffraction patterns of films: (a) by IBS process; (b) by IP process and (c) by FE process.

However, a linear relationship was found between source and film compositions (Fig. 3). A film near $TbFe_2$ was formed with a TbFe source alloy.

In the FE process no marked composition change was found between film and source materials.

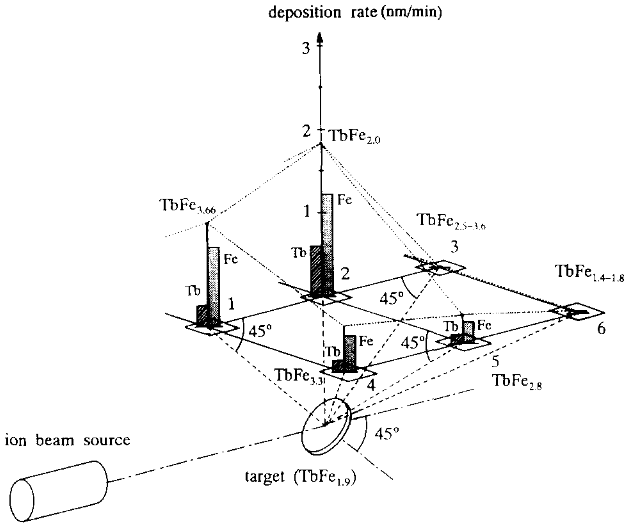

Fig. 2. Total and each elemental deposition rates on differently located substrates in IBS process.

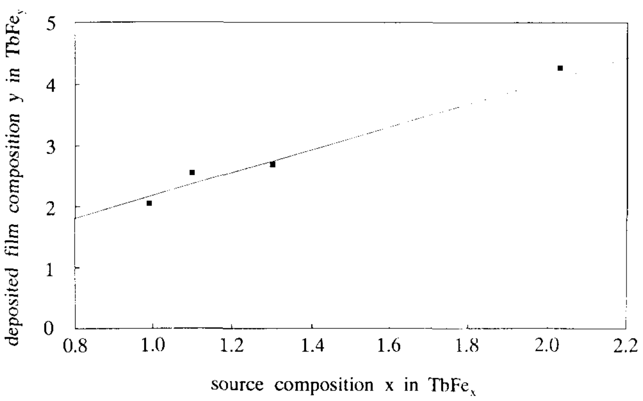

Fig. 3. Dependence of film composition on source material composition in IP process.

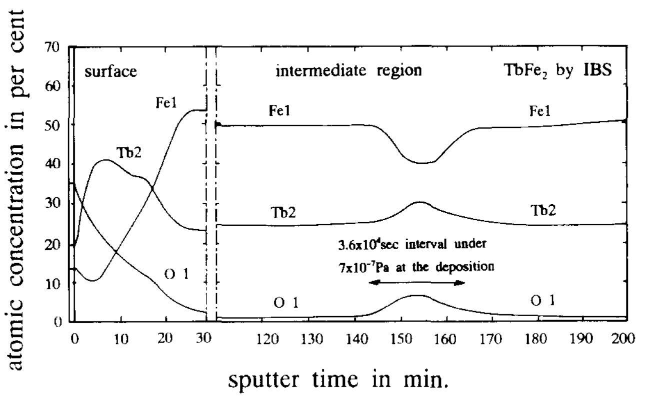

Fig. 4. Elemental depth profiling by AES on a film deposited by IBS process for film surface and intermediate region.

3.3. Contamination

Figure 4 shows an AES depth profile of a film surface and intermediate region deposited by the IBS process. The film was deposited in two periods with an interval of 3.6×10^4 s between them under a vacuum of 7×10^{-7}

TABLE 1. Comparison of film deposition processes

	IBS	IP	FE
Substrate temperature	Room temperature	Room temperature	400–650 K
Structure	Amorphous like	Amorphous like	Amorphous–crystalline
Source composition	$TbFe_2$	$TbFe_2$	$Tb_{0.3}Dy_{0.7}Fe_2$
Film composition	$TbFe_2(TbFe_{1.4-3.6})$	$TbFe_4$	$Tb_{0.3}Dy_{0.7}Fe_{2.2}$
Deposition rate (nm s^{-1})	0.01–0.3	10–20	0.13–2.4
$r=P_r/d_f$ (Pa s nm^{-1})	$(0.3–8)\times10^{-5}$	$(1–2)\times10^{-5}$	$2.9\times10^{-4}–1\times10^{-2}$

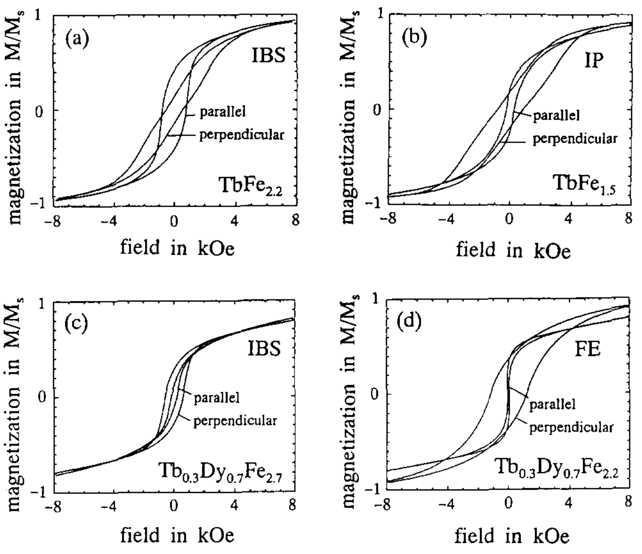

Fig. 5.

Pa. The film surface shows a large concentration change because of exposure to air before the analysis. Rare earth metal segregation caused by oxygen yields a large concentration change. This effect is also seen in the intermediate region where the film formed in the first period was oxidized under the vacuum (7×10^{-7} Pa).

It can be calculated from gas molecule kinetics that within the interval time several tens of monolayers of residual gas molecules (oxygen-containing molecules) were impinging on the surface. This also yielded rare earth and oxygen enrichment on the film surface. Even if little change in composition between film and source materials in a deposition process occurs, oxidation by the residual gas impinging both on the film and on the source material may emphasize the composition change in the film.

A similar depth profile in the surface region was obtained for films produced by the FE process. The concentration of oxygen in the film was in good agreement with the r value. A film of $Tb_{0.3}Dy_{0.63}Fe_{1.92}$ with 1.2 at.% O was obtained with $r=1.4\times10^{-4}$ Pa s nm^{-1} and another film of $Tb_{0.3}Dy_{0.56}Fe_{0.75}$ with 32.6 at.% O was obtained with $r=2.41\times10^{-3}$ Pa s nm^{-1} under the same deposition conditions in the FE process.

3.4. Magnetic properties

The magnetic properties of films produced by the three processes are shown in Fig. 5. The magnetization M/M_s, where M_s is the saturation magnetization, is plotted as a function of the field. Films of $TbFe_{2.2}$ (Fig. 5(a)) and $TbFe_{1.5}$ (Fig. 5(b)) were deposited by IBS and IP respectively. The IP film shows a smaller hysteresis if the magnetization is measured parallel to the film. This difference may be caused by the large internal stress and composition deviation in the IP film, which showed a large warp of the substrate, indicating a negative stress existing in the film.

Because the substitution of Dy for Tb decreases the anisotropy of magnetization [1], much smaller hystereses were measured (Fig. 5(c) and 5(d)). The larger hysteresis of the magnetization perpendicular to the FE film may be due to crystallization and another morphology of the film.

4. Concluding remarks

In Table 1 the characteristics of the three deposition processes are summarized. By the IBS and FE processes films with little deviation in composition from that of the source materials were formed. The IP process yields a much higher deposition rate than the other processes, but control of the composition of the source alloy is necessary prior to evaporation. Also, the high deposition rate leads to larger stress in the film. The characteristics of the deposition process and conditions must be considered for proper film utilization.

The contamination can be assessed by the ratio of residual gas partial pressure to film deposition rate for each process. In particular, the content of oxygen drastically changes the film composition of rare earth and transition metals.

Results of measuring the magnetostriction and the sound velocity of surface acoustic waves on GMM films [9] show that film thickness and composition are the most important factors. From this viewpoint the IBS and FE processes are the preferred processes for the formation of magnetostrictive thin films. The dependence of the residual stress of films on the magnetic characteristics is less understood. In bulk GMMs external stress can increase the magnetostriction [1]. Assuming the same behaviour in GMM films, the prestress given by the IP process could be helpful.

References

1 A.E. Clark, *Ferromagnetic Materials,* Vol. 1, North-Holland, Amsterdam, 1980, Chap. 7, p. 531.

2 K. Lenz, *Proc. Int. Symp. on Giant Magnetostrictive Materials and Other Applications, Tokyo, November 1992,* Advanced Machining Technology and Development Association, Tokyo, 1992, pp. 69 and 137.

3 A. Hietala and W.P. Robbins, *Proc. IEEE Ultrasonics Symp., 1982,* p. 354–358.

4 D.C. Webb, D.W. Forester, A.K. Ganguly and C. Vittoria, *IEEE Trans. Magn., MaG-15*(6) (1979) 1410.

5 M. Yamaguchi, K.Y. Hashimoto, H. Kogo and M. Naoe, *IEEE Trans. Magn., MAG-16*(5) (1980) 916.

6 H.H. Uchida, H. Uchida, Y. Matsumura, V. Koeninger, H. Funakura, T. Noguchi, T. Kurino and H. Kaneko, *Proc. Int. Symp. on Giant Magnetostrictive Materials and Other Applications, Tokyo, November 1992,* Advanced Machining Technology and Development Association, Tokyo, 1992, p. 145.

7 R.F. Bunshah and A.C. Ranghuram, *J. Vac. Sci. Technol., 9* (1972) 1385.

8 H. Uchida, H.H. Uchida and Y.C. Huang, *J. Less-Common Met., 101* (1984) 459.

9 V. Koeninger, H.H. Uchida, H. Uchida and Y. Matsumura, *J. Alloys Comp., 211/212* (1994) 581.