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Novel Properties of Boron Added Amorphous Rare Earth-transition Metal Alloys for Giant Magnetostrictive and Magneto-optical Recording Materials

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Large magneto crystalline anisotropy energy and demagnetization energy of rare earth-transition metal (RE-TM) alloys play roles of bottlenecks towards their commercial applications for giant magnetostrictive and blue wavelength magneto-optical recording materials, respectively. To solve the above problems, boron is added into amorphous RE-TM alloys to produce its electron transferring. The boron added amorphous RE-TM alloys show novel magnetic and magneto-optical properties as follows; 1) an amorphous $(Sm_{33}Fe_{67})_{97}B_3$ alloy obtains the magnetostriction of -550×10^{-6} at 400 Oe compared with saturation magnetostriction of -60×10^{-6} in conventional Ni based alloys, 2) an amorphous $(Nd_{33}Fe_{67})_{95}B_5$ alloy increases effective magnetic anisotropy to -0.5×10^{6} ergs/cm^3 from -3.5×10^{6} ergs/cm^3 without boron, which correspond to the polar Kerr rotation angles of 0.52° and 0.33°, respectively. These results attribute to selective 2p-3d electron orbits exchange coupling (SEC).

1. Introduction

Despite the unique magnetic properties of rare earth (RE) metals, their low Curie temperatures (T_C) below the room temperature prevented from practical applications. In rare earth-transition metal (RE-TM) alloys, the low T_C problem was solved by the magnetic exchange coupling between RE and TM magnetic moments. However, bottlenecks towards their commercial applications still remains in magnetic and magneto-optic (MO) properties, such as small giant magnetostriction (λ_S) of RE-Fe_2 Laves phase intermetallic compounds in low external magnetic fields and low polar Kerr rotation angles (θ_K) of amorphous light RE-TM alloys in short blue wavelengths, which result from large magneto crystalline anisotropy energy and large demagnetization energy, respectively. Previous researches reported that amorphization of RE-Fe_2 compounds [1] and partial substitution of light RE to heavy RE element in amorphous light RE-TM alloys [2, 3, 4] could reduce magneto crystalline anisotropy energy and demagnetization energy, respectively. But these methods resulted in the reductions of λ_S and θ_K which are not desirable for their commercial applications.

In this research, boron (B) is added into amorphous RE-TM alloys to make breakthroughs in giant magnetostrictive materials for obtaining large λ_S with low magnetic fields and in MO recording media for producing large θ_K in the wavelengths of blue wavelengths.

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2. Experimental Procedures

$(SmFe)_{100-X}B_X$ alloys were deposited on Cu substrates from SmFeB alloy targets by a DC sputtering apparatus. The Cu substrates were then dissolved by a mixture of chromium trioxide and sulphuric acid solution. In the case of $(NdFe)_{100-X}B_X$ alloys, they were deposited on quartz substrates by the RF co-sputtering method of a NdFe target and boron pallets on a Fe target with the rotation of 9 RPM.

Crystallographic states of $(SmFe)_{100-X}B_X$ and $(NdFe)_{100-X}B_X$ alloys were confirmed by an X-ray diffractometer (XRD). The composition of the alloys was analysed by X-ray florescent (XRF) depth profiling and inductively coupled plasma-atomic emission spectrometry (ICP-AES). A vibration sample magnetometer (VSM) was used to measure magnetic hysteresis loops. Magnetostriction and polar Kerr rotation angle were obtained by a strain gage method and a Kerr spectrometer, respectively.

3. Results and Discussion

The compositional ratio between RE (Sm, Nd) and Fe is (33 ± 2) and (67 ± 2) at. %. Only boron concentration is systematically changed within the above ratio. In X-ray diffraction curves, no crystalline diffraction peaks of $(SmFe)_{100-X}B_X$ and $(NdFe)_{100-X}B_X$ alloys were observed, except for those of the substrates. Therefore, the addition

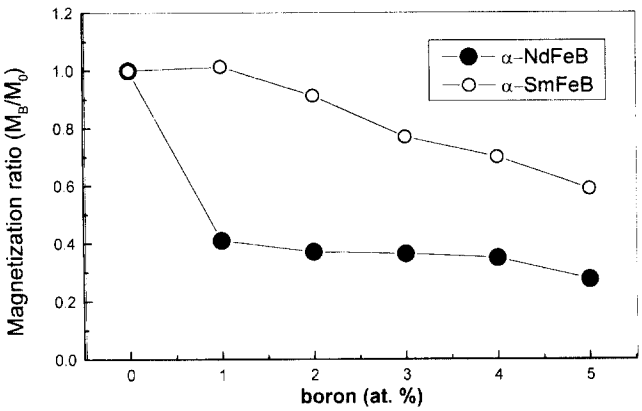

Fig. 1. Changes in magnetization ratio at 15 kOe (M_B/M_0; magnetization of with and without boron, respectively) of amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ and $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys with respect to boron concentration.

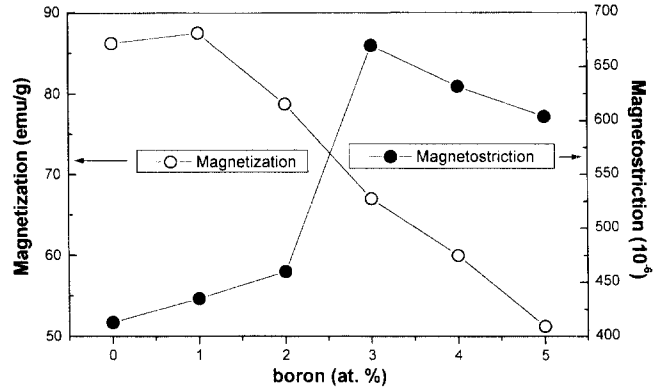

Fig. 2. Relationship between magnetization (M_{15}) and magnetostriction ($\lambda_{//}$-$\lambda_{\perp}$) of amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys, measured at 15 kOe with respect to boron concentration.

of B remains the alloys to be amorphous.

Fig. 1 shows the changes of magnetization ratio at 15 kOe (M_B/M_0; magnetization of with and without boron, respectively) in amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ and $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys with respect to B concentration. In the case of amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys, M_0 and M_B are obtained by division of total magnetic moments of the alloy into the volume of magnetic Nd and Fe atoms, excluding that of non-magnetic B. This implies that M_B/M_0 of $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys should display the virtually same values within the compositional variations between Nd and Fe. However, the M_B/M_0 drastically decreases with small additive B by less than 60%, which means the additive B reduces the magnetization of amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys. In another case of $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys, M_0 and M_B are obtained by division of total magnetic moment of the alloys into their total weights. The large reduction of M_B/M_0 is also shown by the additive B.

As for amorphous FeB alloys, M. Matsuura, *et al.* [5] previously reported on the base of XPS measurement that addition of B less than 14 at. % firstly entered the interstitial sites of DRP (Dense Random Packing) structure, which showed the increase of magnetization by the expansion of Fe-Fe atomic distances [6] Further increase of B more than 14 at. % substituted to Fe atoms which resulted in electron transfer from 2*p* band of B to unoccupied 3*d* bands of Fe [7, 8]. This *p*-*d* electron orbits exchange coupling leads to the reduction of magnetization in the amorphus FeB alloys [9]. From the above results, the reduction of M_B/M_0 in both of the amorphous $(SmFe)_{1-x}B_x$ and $(NdFe)_{1-x}B_x$ alloys may be attributed to 2*p*-3*d* electron orbits exchange coupling between B and Fe. Because unoccupied 4*f* electron bands of RE (Sm and Nd) are well protected by outer 5*s*, 5*p*, 5*d* electron orbits, etc. against electron donors from B, less possibility to form 2*p*-4*f* exchange coupling is expected. Since the *p*-*d* coupling between Fe and B is selectively occurred in the electron bands of RE, Fe and B atoms, it is defined as "selective 2*p*-3*d* electron bands exchange coupling (SEC)". This SEC implies that because magnetization of amorphous RE (Sm, Nd)-Fe-B alloys is reduced without the change of RE magnetic moment, the unique magnetic properties of RE can be remained in the alloys.

From now on, SEC will be applied to deal with the bottlenecks of magnetic and magneto-optic properties in amorphous RE-TM alloys, such as effective giant magnetostriction in low external fields (λ_{GS}) of amorphous $SmFe_2$ alloys and polar Kerr rotation angle in blue wavelengths (θ_{KB}) of amorphous $NdFe_2$ alloys, respectively.

Fig. 2 shows the relationship between magnetization (M_{15}) and magnetostriction ($\lambda_{//}$-$\lambda_{\perp}$; $\lambda_{//}$ and $\lambda_{\perp}$ denote the magnetostriction constants parallel and perpendicular, respectively to external magnetic field) of amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys, measured at 15 kOe with respect to B concentration. Less than 2 at. % of B, $\lambda_{//}$-$\lambda_{\perp}$ proportionally increases with the initial increase of M_{15}, which can be explained by the expansion of Fe-Fe distances[6]. But further addition of B decreases magnetization with rapid increase of $\lambda_{//}$-$\lambda_{\perp}$. The reduction of M_{15} is considered by electron transferring from 2*p* electrons of B to unoccupied 3*d* bands of Fe[7,8,9]. However, evidently, this does not represent the proportional relationship[10,11] between magnetization (M) and magnetostriction (λ) as below,

$$\lambda = 1.5\lambda_S(M/M_S)^2 \quad (1)$$

λ_S: saturation magnetostriction

Ms: saturation magnetization

Even though the relationship in Eq. 1 does not entirely apply to amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys, it can separately satisfy with below and above 2 at. % of B, at which SEC is occurred.

To investigate boron added amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys for their commercial purpose, effective giant magnetostriction in low external fields $\{\lambda_{GS}=1.5(\lambda_{//}-\lambda_{\perp})\}$ is measured in Fig. 3. The maximum $\lambda_{//}$-$\lambda_{\perp}$ is -550×10^{-6} at 400 Oe, when 3 at. % of boron is added. This value is 10 times larger than that of the conventional magnetostrictive

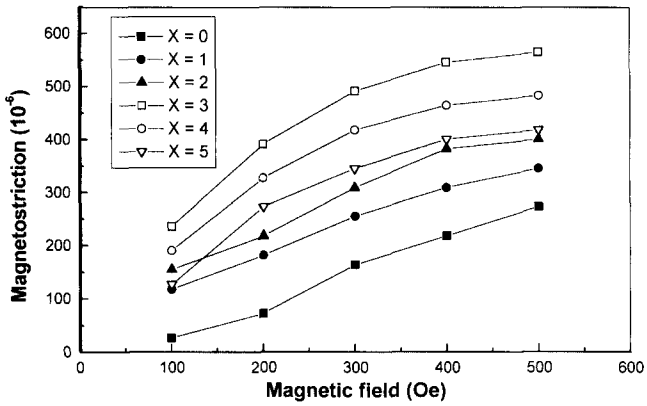

Fig. 3. Magnetostriction constants ($\lambda_{//}$-$\lambda_{\perp}$) of amorphous $(Sm_{33}Fe_{67})_{100-x}B_x$ alloys in low magnetic fields.

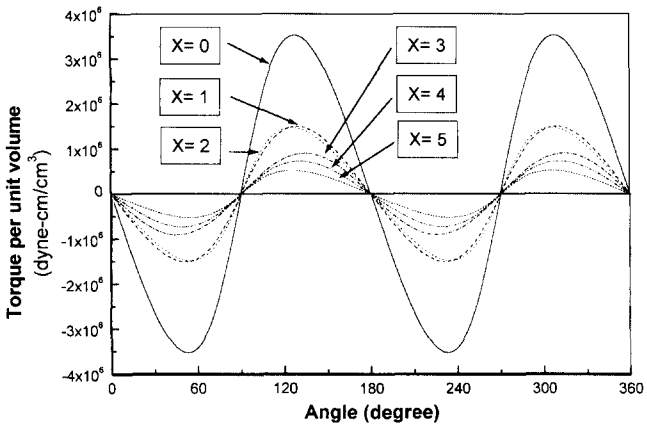

Fig. 4. Changes of effective anisotropy energy (K_{Ueff}) of amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys in terms of boron concentration.

Ni based alloys.

From this time, magneto-optical (MO) properties of boron added amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys is evaluated for blue wavelength MO recording media. Even though an amorphous NdFe alloy film is one of the promising candidates for MO recording media in blue wavelengths, large demagnetization energy ($2\pi M_S^2$) prevents from magnetic anisotropy (K_{Ueff}) perpendicular to the film planes as follows,

$$K_{Ueff}=K_U-2\pi M_S^2 \text{ (in cgs unit)} \quad (2)$$

K_{Ueff}: effective uniaxial magnetic anisotropy energy (ergs/cm^3)

K_U: intrinsic uniaxial magnetic anisotropy energy (ergs/cm^3)

M_S: saturation magnetization (emu/cm^3)

So only small polar Kerr rotation angle (θ_{KB}) is developed. To solve this problem, SEC applies to amorphous NdFe alloy films to reduce demagnetization energy and obtain large θ_{KB}. As shown in Fig. 1, the additive B of $(Nd_{33}Fe_{67})_{100-x}B_x$ alloy films decrease magnetization and then demagnetization energy in Eq. 2, which expects to increase K_{Ueff} perpendicular to the film planes.

Fig. 4 shows the changes of effective anisotropy energy

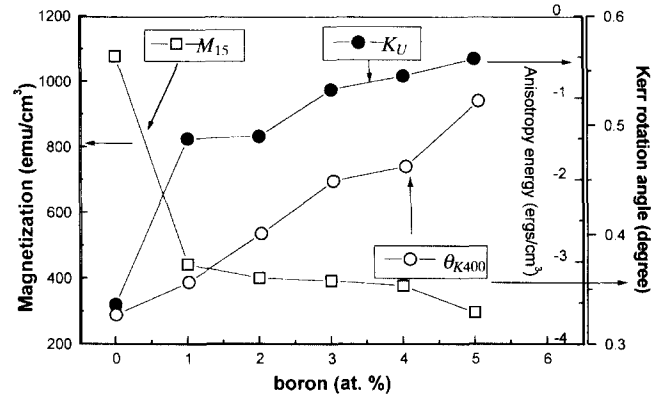

Fig. 5. Relationship of magnetization at 15 kOe (M_{15}), effective magnetic anisotropy energy (K_{Ueff}) and polar Kerr rotation angle at 400 nm (θ_{K400}) of the amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys with respects to boron concentration.

(K_{Ueff}) in amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys. The K_{Ueff} of amorphous $Nd_{33}Fe_{67}$ alloy without B is -3.5×10^6 ergs/cm^3, which has the preferential magnetic direction parallel to the film plane. However, additive 5 at. % of B into the amorphous NdFe alloy reduces in-plane K_{Ueff} to -0.5×10^6 ergs/cm^3, which implies that the preferential magnetic direction changes from in-plane to perpendicular directions. θ_{KB} at 400 nm is measured in order to confirm whether SEC is effective for magneto-optical property or not.

The relationship of M_{15}, K_{Ueff} and θ_{K400} at 400 nm of amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys is shown in Fig. 5. As explained in Fig. 1 and 4, the additive B significantly reduces M_{15} and in-plane K_{Ueff}. But the increase of θ_{K400} breaks the proportional relationship[12] between magnetization and θ_K, because θ_K is generally determined by the contribution of composed elements and the dominance of sub-magnetization in the magnetically saturated state. The θ_{K400} of NdFe alloys is more strongly affected by Nd contribution than Fe contribution, since the photo emission transition energy of Fe and Nd is about 1.613 and 4.214 eV, respectively. On the figure, the opposing trend of M_{15} and θ_{K400} with boron addition implies that the SEC successfully reduces the Fe sub magnetic moment without changing Nd sub magnetic moment. This accounts for the decrease of the demagnetization energy and sequent increase of K_{Ueff} perpendicular to the film plane. The decreased in-plane K_{Ueff} makes contribution of Nd moment towards perpendicular to the film planes and the value of θ_{K400} from 0.33 to 0.52° in the amorphous $(Nd_{33}Fe_{67})_{100-x}B_x$ alloys.

4. Conclusions

SEC produces novel effect on magnetic and magneto-optic properties in amorphous RE-Fe alloys by selective reduction of Fe magnetic moment. This achieves large effective giant magnetostriction constant of -550×10^{-6} at 400 Oe in the amorphous $(Sm_{33}Fe_{67})_{97}B_3$ alloy and polar

Kerr rotation angle of 0.52° at 400 nm in the amorphous $(Nd_{33}Fe_{67})_{95}B_5$ alloys. The SEC will provide new scientific hints to develop unique magnetic properties of rare earth elements.

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