

Mean Field Analysis of Exchange Coupling in Amorphous REFe₂-B (RE = Dy, Sm) Alloy Ribbons

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Experimental magnetization-temperature curves for melt-spun ribbons of amorphous alloys (Dy_{0.33}Fe_{0.67})_{1-x}B_x (x=0, 0.05, 0.1, and 0.15) and (Sm_{0.33}Fe_{0.67})_{1-x}B_x (x=0, 0.01, 0.02, and 0.03) (in atomic fraction) are fitted with theoretical equations based on the mean field theory in order to calculate exchange couplings between constituent elements as a function of the B content. In the case of the DyFe₂-B system, the sign of the exchange coupling between Dy and Fe is negative, indicating that the magnetization direction of Dy is antiparallel to that of Fe. The sign of the other two couplings are positive indicating a parallel alignment. The exchange coupling between Fe ions are greatest, while that between Dy ions is negligible. In the case of the SmFe₂-B alloys, the sign of all the couplings are positive, indicating ferromagnetic coupling between the spins. The exchange couplings between Fe ions, and Fe and Sm are comparable to each other, but they are much greater than that between Sm ions. The high exchange coupling between Fe and Sm, which is considered to occur indirectly, is rather unexpected, but it is considered to be unique characteristics of amorphous Sm-Fe alloys. In both alloy systems, the exchange coupling between Fe ions increases with increasing B content, and this may be explained by the increase of the Fe-Fe separation with increasing B content. The exchange coupling between Fe and RE also increases with increasing B content. As the B content increases, the magnetization decreases over the whole temperature range, and the Curie temperature also decreases.

I. Introduction

Mean field theory has played an important role in understanding magnetism of various magnetic materials, in particular RE-TM (where RE and TM indicate rare earths and transition metals, respectively) alloys [1-5]. The two sublattice model has frequently been used since both elements are magnetic. Much work has been done in crystalline alloys, usually intermetallic compounds, but this is not the case for amorphous RE-TM alloys, although these alloys constitute a practically important family of magnetic materials in the fields of magneto-optics and magnetostriction [6-8]. Apart from their practical importance, amorphous RE-TM alloys are also of theoretical significance, since both elements are usually magnetic elements and the involved magnetism of the alloys is rather complex [6]. Among the magnetic parameters frequently investigated in the past are exchange interactions between the constituent

atoms [1-3]. A popular way of calculating the exchange interactions is to fit experimental M - T (where M and T denote the magnetization and temperature, respectively) curves with theoretical equations based on the mean field theory [1-5]. The theoretical equations contain three exchange interactions as fitting parameters, in the case of RE-TM alloys with two sublattices. The three exchange interactions are identified to be J_{TT} (between transition metals), J_{RT} (between rare earth and transition metal) and J_{RR} (between rare earths). In spite of its simple nature, the mean field theory describes the magnetization behavior reasonably well in most cases. Amorphous RE-TM alloys with a wide composition range were frequently investigated in the past [3, 9, 10]. The main reason for this is to examine the compositional dependence of TM magnetization which can provide information on the change of electronic state of itinerant 3d electrons on alloying. In the case of an alloy with a rather poor glass forming

ability, an amorphous phase is formed only in a limited composition range, preventing us from examining magnetic behavior over a wide composition range. In order to avoid this problem, a large amount of B (about 20 %) is usually added to RE-TM alloys [11]. The main emphasis of examining RE-TM-B alloys in the past is therefore not to investigate the role of B in affecting magnetic properties but to extend the composition range (specifically the fraction of RE with respect to TM) over which magnetic properties such as the compositional dependence of TM magnetization are examined. The present work on amorphous RE-TM-B alloys, however, differs from the past in that the B content is varied while the RE/TM ratio is fixed in order to examine the effects of B on the magnetic properties. The alloy systems investigated are $(\text{Dy}_{0.33}\text{Fe}_{0.67})_{1-x}\text{B}_x$ ($x = 0, 0.05, 0.1$), and 0.15 , and $(\text{Sm}_{0.33}\text{Fe}_{0.67})_{1-x}\text{B}_x$ ($x = 0, 0.01, 0.02$, and 0.03) (in atomic fraction). It is noted that the RE/Fe ratio is that corresponding to REFe_2 which is well-known to possess giant magnetostriction at room temperature and hence a great application potential [8].

II. Mean Field Theory

Theoretical equations describing M - T curves of RE-TM alloys by using the mean field theory in the two sublattice model can be found in the literature [1-5], but they are briefly explained here again for an amorphous REFe_2 alloy mainly to define the notations and explain their meaning. The molecular fields acting on the two sublattices of RE and Fe at temperature T (respectively denoted by $H_{RE}(T)$ and $H_{Fe}(T)$) are given by;

$$H_{RE}(T) = H + d[n_{RR}M_{RE}(T) + 2n_{RF}M_{Fe}(T)] \quad (1(a))$$

$$H_{Fe}(T) = H + d[2n_{FF}M_{Fe}(T) + n_{RF}M_{RE}(T)] \quad (1(b))$$

Here, H is the applied field, $M_{RE}(T)$ and $M_{Fe}(T)$ are the magnetic moments of RE and Fe, respectively, at temperature T in unit of the Bohr magneton (μ_B). The quantity d is a conversion factor for the magnetic moment from the Bohr magneton to gauss, and is given by;

$$d = \frac{N_A H_B \rho}{A} \quad (2)$$

Here, N_A is the Avogadro's number, ρ the density in g/cm^3 , and A is the REFe_2 formula weight. With these definitions, all the fields (H , $H_{RE}(T)$ and $H_{Fe}(T)$) are given in gauss, and the molecular field coefficients n_{RR} , n_{RF} and n_{FF} , which respectively describe the exchange interactions between the sublattices of RE-RE, RE-Fe and Fe-Fe, are in dimensionless unit. The temperature dependence of each sublattice magnetization is governed by a Brillouin function ($B_J(x)$);

$$H_{RE}(T) = M_{RE}(0) B_J[M_{RE}(0) H_{RE}(T) / kT] \quad (3(a))$$

$$H_{Fe}(T) = M_{Fe}(0) B_J[M_{Fe}(0) H_{Fe}(T) / kT] \quad (3(b))$$

Here, $M_{RE}(0)$ and $M_{Fe}(0)$ denote, respectively, the zero temperature RE and Fe moments. The Brillouin function $B_J(x)$ is given by;

$$B_J(x) = \frac{2J+1}{2J} \coth\left[\frac{(2J+1) \cdot x}{2J}\right] - \frac{1}{2J} \coth\left(\frac{x}{2J}\right) \quad (4)$$

where J is the total angular momentum. The values of J used in this work are 7.5 for Dy, 2.5 for Sm [12] and 1.0 for Fe [6]. In this study, the value of $M_{RE}(0)$ is determined from the theoretical relationship, $M_{RE}(0) = g_{RE} J_{RE}$ (where g_{RE} is the g factor for rare earth and its magnitude used are 4/3 for Dy and 2/7 for Sm [12]) and $M_{Fe}(0)$ is then determined from the measured total moment at zero K. Note that the total theoretical magnetization is the vector sum of the sublattice magnetizations, and, in the present case of REFe_2 , we have

$$M_{tot}(T) = M_{RE}(T) \pm 2M_{Fe}(T) \quad (5)$$

The exchange interaction coefficients n_{ij} , which are the fitting parameters in the simulation, are determined at a minimum value of R (in %), a parameter showing the degree of fitting. The quantity R is given by;

$$R = 100 \cdot \frac{\sum |M_{exp}(T_i) - M_{tot}(T_i)|}{\sum M_{exp}(T_i)} \quad (6)$$

where $M_{exp}(T_i)$ is the measured magnetization at temperature T_i . In the absence of an applied field, a relation among the parameters including n_{ij} and T_c can

be obtained from the molecular field equations. With the condition that both $M_{RE}(T)$ and $M_{Fe}(T)$ are equal to zero at $T=T_c$, we obtain

$$(n_{RR}-T_c\alpha)(n_{FF}-T_c\beta)-n_{RF}^2=0 \quad (7)$$

where

$$\alpha \equiv \frac{[3J_{RE}/(J_{RE}+1)] \cdot [k/\mu_B d]}{M_{RE}^2(0)} \quad (8(a))$$

$$\beta \equiv \frac{[3J_{Fe}/(J_{Fe}+1)] \cdot [k/\mu_B d]}{2M_{Fe}^2(0)} \quad (8(b))$$

The Curie temperature can be calculated theoretically from Eqs. (7) and (8), and is given by;

$$T_c = \frac{(n_{FF}\alpha + n_{RR}\beta) + \{(n_{FF}\alpha + n_{RR}\beta)^2 - 4\alpha\beta(n_{FF}n_{RR} - n_{RF}^2)\}^{1/2}}{2\alpha\beta} \quad (9)$$

The calculated value of $M_{RE}(0)$ from the mean field theory is usually smaller than the value estimated from the relation $M_{RE}(0) = g_{RE}J_{RE}$ ($10 \mu_B$ for Dy and $0.714 \mu_B$ for Sm [12]). This reduction may be attributed to the non-collinear and conical spin structure of RE due to strong random anisotropy [13, 14]. The spin canting angle (θ) can be calculated from the relation [15];

$$\frac{M_{exp}(0)}{M_{cal}(0)} = \frac{M_{RE}(0)}{g_{RE}J_{RE}} = \frac{1 + \cos\theta}{2} \quad (10)$$

III. Experimental

In the case of the SmFe₂-B system, all the alloys were induction-melted. In the case of the DyFe₂-B alloys, ingots of the B-free alloy were induction-melted and then used to produce buttons of the B containing alloys by arc-melting. The melting experiments were performed in an Ar atmosphere. Melt-spinning was subsequently carried out also in an Ar atmosphere. A high wheel velocity of 50 m/s was used to obtain an amorphous phase. The composition of both melted ingots and melt-spun ribbons was determined with an inductively coupled plasma emission spectrophotometer. The composition given in this paper is that of melted ingots. No appreciable compositional change was observed during melt-quenching. The microstructure was

examined by X-ray diffraction with Cu K α radiation. The temperature dependence of magnetization was measured with a SQUID magnetometer and a VSM during heating from 5 to 700 K, with an applied field of 50 kOe.

IV. Results and Discussion

4.1 The DyFe₂-B System

All the melt-spun ribbons investigated in this work are confirmed to be amorphous, since no crystalline peaks but diffuse halos are detected by X-ray diffraction. The coercivity of the present amorphous alloys is very large at low temperatures due to the occurrence of very strong single-ion anisotropy of Dy, although it is moderate at room temperature. In the case of the B-free alloy, for example, the coercivity is 0.13 kOe at 300 K, but it is increased greatly to 23.0 kOe at the lowest temperature of 4.5 K. Expectedly, the alloys are not saturated completely at the highest applied field of 50 kOe used for the measurement of M - T curves. This is seen from the hysteresis loop of the B-free alloy, shown in Fig. 1, measured at a temperature of 4.5 K and a maximum applied field of 50 kOe. Note that the B-free alloy exhibits the highest coercivity among the alloys investigated. The steady increase of magnetization after technical saturation is typical of amorphous RE-TM alloys, and it is known that the

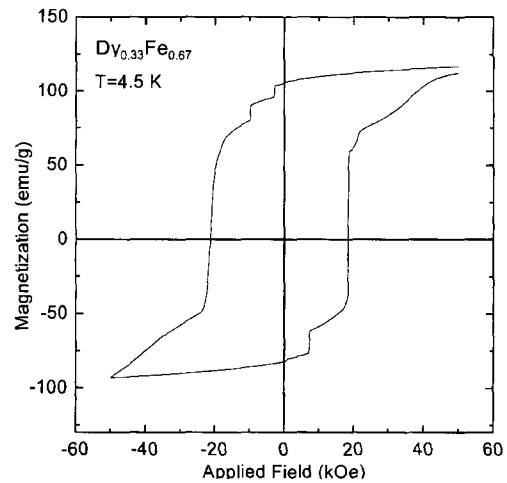


Fig. 1. The hysteresis loop of an amorphous Dy_{0.33}Fe_{0.67} alloy measured at a temperature of 4.5 K and a maximum applied field of 50 kOe.

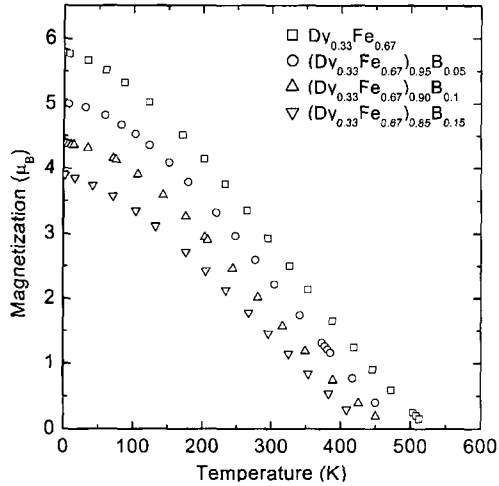


Fig. 2. Experimental M - T curves for amorphous $(\text{Dy}_{0.33}\text{Fe}_{0.67})_{1-x}\text{B}_x$ alloys with $x=0, 0.05, 0.1$, and 0.15 investigated in this work.

non-collinear spin structure as well as very large single-ion anisotropy of RE is responsible for the phenomenon [13, 14]. The magnetization obtained at the present condition is thought to be suitable for examining exchange coupling by using the mean field theory.

Experimental M - T curves for all the B contents are shown in Fig. 2. Although the shape of the curves is similar to each other irrespective of the B content, it is significantly different from that observed in ordinary ferromagnetic materials. The decrease in magnetization near 0 K is small similarly to ordinary ferromagnetic materials, but, at intermediate temperatures, the magnetization decreases nearly linearly with temperature. At temperatures near T_c , the magnetization decrease again becomes small, resulting in a so-called "inflection-like behavior" in the M - T curve. This is due to the ferrimagnetic alignment of the Dy and Fe magnetizations (this is confirmed from the present mean field analysis as will be discussed later). Recently, similar results were presented and also discussed in amorphous Gd-Fe alloys by Yano [3]. The magnetization decreases with increasing B content. The magnetization reduction is considered to be not small; near zero K, the magnetization is $5.87 \mu_B$ at $x=0$, $5.08 \mu_B$ at $x=0.05$, $4.45 \mu_B$ at $x=0.1$ and $3.95 \mu_B$ at $x=0.15$. The magnetization decrease is larger in the small B

content region than that at high B contents. In order to explain the B content dependence of the magnetization, it is necessary to consider the respective sublattice magnetization. With the present ferrimagnetic spin structure and the total magnetization dominated by the Dy sublattice, as will be shown later, the B content dependence of the magnetization can be explained by the decrease of Dy sublattice magnetization and/or the increase of Fe sublattice magnetization. The increase of Fe sublattice magnetization is not likely to occur by B addition, because B is known to act as the electron "donor" in a TM-B alloy (namely, the electrons supplied by B fill the unfilled 3d shell in TM), causing to reduce the magnetization [16]. This leaves the reduction of Dy sublattice magnetization for the possible explanation of the observed magnetization reduction. It is unlikely that the absolute magnitude of Dy sublattice magnetization is affected by B addition, because the magnetic 4f shell of Dy is located deep inside and hence shielded from outside environment. The sublattice magnetization discussed thus far is the absolute magnitude. The measured magnetization, however, depends not only on the absolute magnitude but also on the degree of spin canting. In amorphous RE-TM alloys, magnetization direction *within* the sublattice is often non-collinear due to very strong random anisotropy of RE and/or reduced exchange coupling by amorphization, resulting in a magnetic structure called "sperimagnet" [6, 14]. Since, from the present experimental results, it is not possible to estimate the non-collinearity (in the case of both Dy and Fe) and the decrease of the absolute magnitude of magnetization (in the case of Fe) as a function of the B content, it is assumed in this work that the "effective" sublattice magnetization (magnetization component in the applied field direction) of each sublattice decreases in an equal proportion. In the case of Dy sublattice where the magnetization reduction occurs solely by spin canting, the canting angle can be calculated from Eq.(10). The canting angle is rather large and increases with increasing B content; it is 16° for the B-free alloy, but it is increased to 71° at the highest B content of 15 at.%.

The fitting of the experimental data with the theoretical equations described in Section 2 is carried

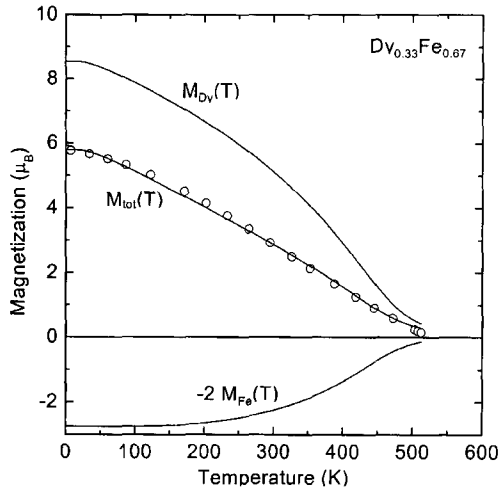


Fig. 3. Calculated M - T curves for an amorphous $Dy_{0.33}Fe_{0.67}$ alloy. The results for the total magnetization and its contribution from the Dy and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

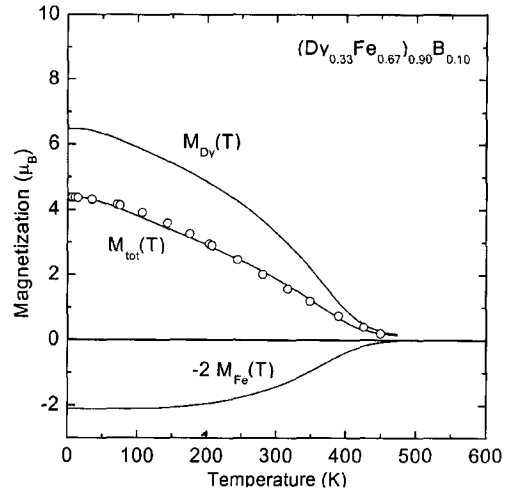


Fig. 5. Calculated M - T curves for an amorphous $(Dy_{0.33}Fe_{0.67})_{0.9}B_{0.1}$ alloy. The results for the total magnetization and its contribution from the Dy and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

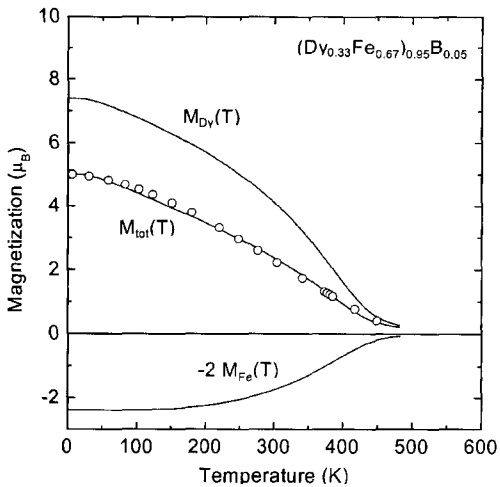


Fig. 4. Calculated M - T curves for an amorphous $(Dy_{0.33}Fe_{0.67})_{0.95}B_{0.05}$ alloy. The results for the total magnetization and its contribution from the Dy and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

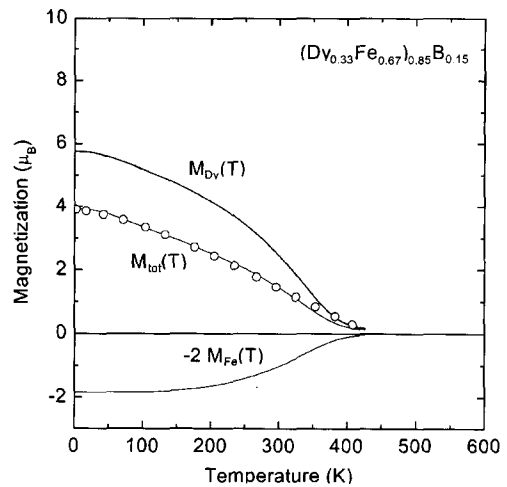


Fig. 6. Calculated M - T curves for an amorphous $(Dy_{0.33}Fe_{0.67})_{0.85}B_{0.15}$ alloy. The results for the total magnetization and its contribution from the Dy and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

out, and the results are shown in Figs. 3-6 for $x = 0, 0.05, 0.1,$ and $0.15,$ respectively. The scale of the y axis is intentionally maintained constant for all the figures, in order to clearly see the B content dependence of M - T curves. The agreement between the calculated and experimental data is very good over the whole

temperature range, except for the temperature near T_c . The discrepancy near T_c may be due to the inability of the mean field theory to explain rather complex magnetization behavior at temperatures close to T_c , a large exchange fluctuation, for example. Two main points can be noted from the results shown in Figs. 3-

6. Firstly, the magnetization contribution from the Dy sublattice is much greater than that from the Fe sublattice. Secondly, the sign of the magnetization from the two sublattices is opposite, indicating that the two sublattice magnetizations are aligned antiparallel. This is in agreement with the previous observations [17, 18], and also is in accord with the theory [6] in which the two sublattice magnetizations in amorphous RE-TM alloys are antiparallel in the case of the heavy rare earths such as Dy. Magnetic parameters including the exchange interactions optimized from the present fitting procedure are summarized in Table I. The R value is small (1.8 %) for the B-free alloy, but it increases with increasing B content. However, even the highest value of 4 % obtained at $x = 0.15$ is considered to be reasonable. The order in the absolute magnitude of the exchange coefficient is $n_{FF} > n_{DF} > n_{DD}$. This is in agreement with the previous results in crystalline alloys [17, 18]. Also the present result is well expected, since the wave function of transition metals is much more extended than that of rare earths. Because of this, the coupling between Fe and Fe is direct and strong, but the exchange interaction involving rare earth is indirect and weak. It is particularly noted that the magnitude of n_{DD} is much smaller than the other two coefficients, indicating that spin alignment is minimally affected by Dy-Dy exchange coupling.

As the B content increases, the absolute values of both n_{FF} and n_{DF} increase monotonically. Due to the much smaller magnitude of n_{DD} and also equally smaller contribution to the calculated $M-T$ curve (in other words, the value of R is little affected by n_{DD}), it is impossible to accurately obtain the B content dependence of this exchange coefficient. Due to the itinerant nature of Fe atoms, the present B content dependence of n_{FF} should be explained by considering

the change in electronic state with the addition of B, but this is not a simple problem and beyond of the scope of the present paper. One possible and simple reason for this may be related with the change in the distance between Fe atoms with the addition of B. Exchange interaction between Fe atoms is known to be a sensitive function of the distance between the atoms [6, 16]. In the distance range of ferromagnetic exchange interaction, the coupling increases with increasing separation between the atoms. One good and recent example is the results observed in amorphous binary Gd-Fe alloys by Yano [3] and Yano *et al.* [19], and in amorphous ternary Gd-Fe-B₂₀ alloys by Hassini *et al.* [11]. They observed an increase of n_{FF} with the increase of Gd content, and explained this increase of n_{FF} by the increased Fe-Fe separation. The increase of Fe-Fe separation is well expected in amorphous Gd-Fe alloys, since the atomic radius of Gd is greater than that of Fe. The present result for the increase of n_{FF} with the increase of B content can be explained similarly, since B is expected to reside in interstitial sites, and hence to increase the Fe-Fe separation with the increase of the B content.

The B content dependence of n_{DF} is much larger than that of n_{FF} ; in the investigated B content range, the increase of n_{FF} is 12 %, but that of n_{DF} is 117 %. Resultantly, the absolute magnitude of n_{DF} becomes about one half of n_{FF} at the highest B content of 15 at.%, and this high value is considered to be rather unexpected. Normally, the coupling between RE and TM is much smaller than that between transition metals. In crystalline REFe₂ compounds (RE = Tb, Dy, Ho, Er, and Tm), for example, the absolute magnitude of the coupling coefficient between RE and TM is 10~20 % of that between transition metals [18]. This can be understood since the exchange interaction between RE and TM is known to be

Table I. The exchange interaction coefficients n_{ij} , the Curie temperature (both measured and calculated) T_c and the fitting parameter R for the DyFe₂-B system.

	n_{FF}	n_{DF}	n_{DD}	Measured T_c (K)	Calculated T_c (K)	R (%)
Dy _{0.33} Fe _{0.67}	1000	-2390	145	509	512	1.80
(Dy _{0.33} Fe _{0.67}) _{0.95} B _{0.05}	10810	-3140	145	473	486	2.68
(Dy _{0.33} Fe _{0.67}) _{0.90} B _{0.1}	10990	-4180	100	458	468	3.40
(Dy _{0.33} Fe _{0.67}) _{0.85} B _{0.15}	11990	-5190	100	437	445	4.02

indirect through polarized spins in the conduction band. The trend appears similar in amorphous RE-TM alloys, but the gap seems to decrease significantly with amorphization, although, with a paucity of available data, it is not easy to say something definite at this stage. In an Er-Fe system where data are available in both crystalline (ErFe₂) and amorphous (ErFe₃) states, the absolute magnitude of the coupling coefficient between Er and Fe is 10.8 % of that between Fe and Fe in crystalline state [18], but it becomes comparable to that between Fe atoms (70 %) in amorphous state [10]. Another example is an amorphous GdFe₂ (the data for crystalline GdFe₂ are not available, however), the exchange coupling between Gd and Fe is about 40 % of that between Fe atoms [3]. Considering this, the relatively high value of n_{DF} at $x = 0.15$ is thought to be not that extraordinary.

The Curie temperature decreases from 509 to 437 K as the B content increases from 0 to 0.15. Although the measured T_c values are slightly lower than the calculated ones over the whole B content range, the agreement between the two is considered to be reasonable.

4.2. The SmFe₂-B System

X-ray diffraction patterns indicate that all the melt-spun ribbons investigated in this work are confirmed to be amorphous. Similarly to the DyFe₂-B system, the coercivity of the present amorphous alloys is very large at low temperatures. In the case of the B-free alloy, the coercivity is 1.0 kOe at 250 K, but it is increased greatly to 20.1 kOe at the lowest temperature of 4.5 K. Expectedly, the alloys are not saturated completely at the highest applied field of 50 kOe used for the measurement of M - T curves. This is seen from the hysteresis loop of the B-free alloy, shown in Fig. 7, measured at a temperature of 4.5 K and a maximum applied field of 50 kOe. Note that the B-free alloy exhibits the highest coercivity among the alloys investigated. The steady increase of magnetization after technical saturation is typical of amorphous RE-TM alloys, and it is known that the non-collinear spin structure as well as very large single-ion anisotropy of RE is responsible for the phenomenon [14]. The magnetization obtained at the present condition is

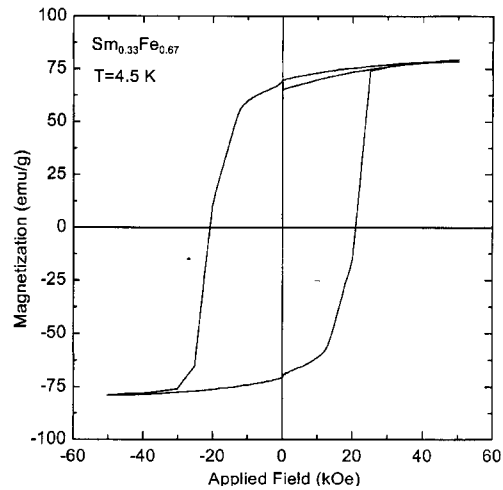


Fig. 7. The hysteresis loop of an amorphous Sm_{0.33}Fe_{0.67} alloy measured at a temperature of 4.5 K and a maximum applied field of 50 kOe.

thought to be suitable for examining exchange coupling by using the mean field theory.

Experimental M - T curves for all the B contents are shown in Fig. 8. The shape of the curves is similar to each other irrespective of the B content. However, the absolute value of magnetization decreases with increasing B content. The magnetization reduction is considered to be not small considering that the

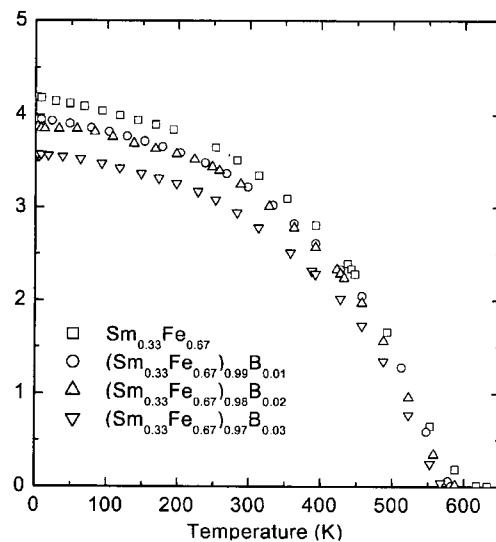


Fig. 8. Experimental M - T curves for amorphous (Sm_{0.33}Fe_{0.67})_{1-x}B_x alloys with $x = 0, 0.01, 0.02,$ and 0.03 alloys investigated in this work.

maximum amount of B added is only 3 at.%. Although the B content increases at a constant step of 1 at.%, the magnetization reduction is not linear. This does not seem to represent intrinsic properties, but quite likely an experimental error, particularly, in the B content, an accurate analysis of which is not easy to make. In order to explain the B content dependence of the magnetization, it is necessary to consider the respective sublattice magnetization. As will be discussed later, the sublattice magnetization directions of Sm and Fe are parallel to each other. This indicates that the sublattice magnetizations of Sm and/or Fe should be reduced by B addition. It is well-known in a TM-B system that B acts as the electron “donor”, namely, the electrons supplied by B fill the unfilled 3d shell in TM, causing to reduce the magnetization [16]. A similar behavior is expected to occur in the Fe sublattice and resultantly the magnetization of the Fe sublattice decreases with increasing B content. However, the magnetization of the Sm sublattice, the magnetic 4f shell of which is located deep inside, will not be affected by B addition. The sublattice magnetization discussed thus far is the absolute magnitude. The measured magnetization, however, depends not only on the absolute magnitude but also on the non-collinearity of the magnetization. In amorphous RE-TM alloys, magnetization direction *within* the sublattice is often non-collinear due to very strong random anisotropy of RE and/or reduced exchange coupling by amorphization, resulting in a magnetic structure called “sperimagnet” [5]. Since, from the present experimental results, it is not possible to estimate the non-collinearity (in the case of both Sm and Fe) and the decrease of the absolute magnitude of magnetization (in the case of Fe) as a function of the B content, it is assumed in this work that the “effective” sublattice magnetization (magnetization component in the applied field direction) of each sublattice decreases in an equal proportion. In the case of Sm sublattice where the magnetization reduction occurs solely by spin canting, the canting angle can be calculated from Eq. (10). The canting angle is rather large and increases with increasing B content; it is 38° for the B-free alloy, but it is increased to 61° at the highest B content of 3 at.%. The fitting of the experimental data with the

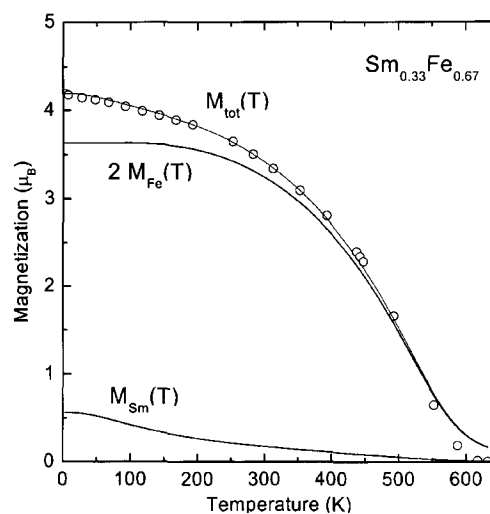


Fig. 9. Calculated M - T curves for an amorphous $\text{Sm}_{0.33}\text{Fe}_{0.67}$ alloy. The results for the total magnetization and its contribution from the Sm and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

theoretical equations described in Section 2 is carried out, and the results are shown in Figs. 9-12 for $x = 0, 0.01, 0.02$ and 0.03 , respectively. The scale of the y axis is intentionally maintained constant for all the figures, in order to clearly see the B content dependence of M - T curves. The agreement between the calculated and experimental data is very good over the whole temperature range, except for the temperature near T_c . The discrepancy near T_c may be due to the inability of the mean field theory to explain rather complex magnetization behavior at temperatures close to T_c , a large exchange fluctuation, for example. Two main points can be noted from the results shown in Figs. 9-12. Firstly, the magnetization contribution from the Fe sublattice is much greater than that from the Sm sublattice. Secondly, the sign of the magnetization from the two sublattices is identical to each other, indicating that the two sublattice magnetizations are aligned parallel. This is in agreement with the previous observations [17, 20], and also is in accord with the theory [5] in which the two sublattice magnetizations in amorphous RE-TM alloys are parallel in the case of the light rare earths such as Sm. Magnetic parameters including the exchange interactions optimized from the present fitting procedure are summarized in Table

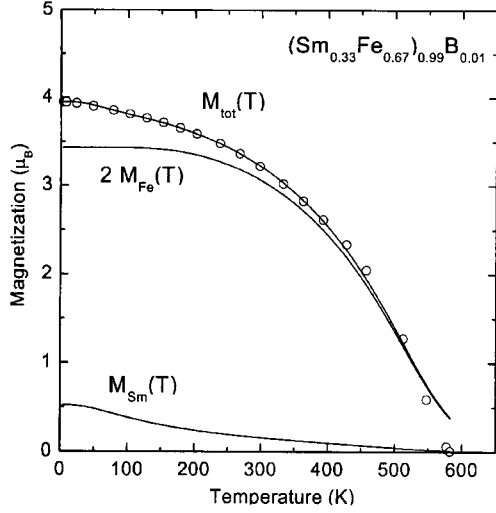


Fig. 10. Calculated M - T curves for an amorphous $(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.99}\text{B}_{0.01}$ alloy. The results for the total magnetization and its contribution from the Sm and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

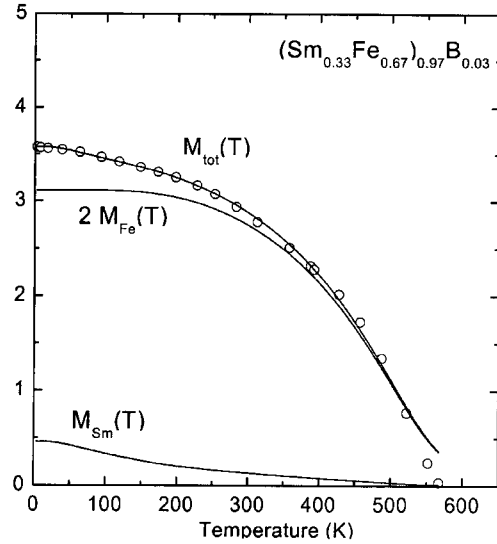


Fig. 12. Calculated M - T curves for an amorphous $(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.97}\text{B}_{0.03}$ alloy. The results for the total magnetization and its contribution from the Sm and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

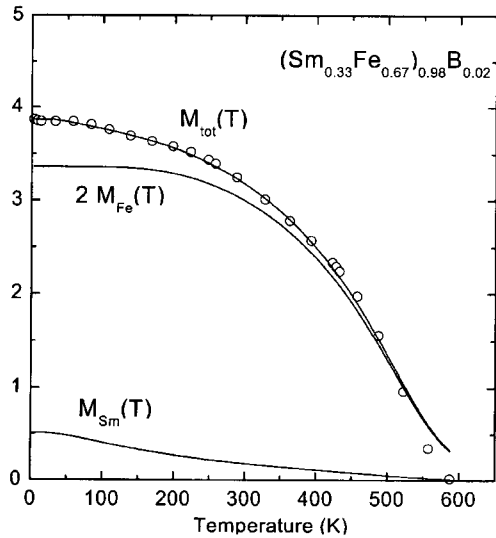


Fig. 11. Calculated M - T curves for an amorphous $(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.98}\text{B}_{0.02}$ alloy. The results for the total magnetization and its contribution from the Sm and Fe sublattices are shown together in the figure. Experimental M - T curves are also shown for comparison (indicated by circles).

II. The R value is well within 2 % in all cases, indicating that the present fitting is reasonably good. The magnitudes of n_{FF} and n_{SF} are comparable to each other but they are much greater than n_{SS} , indicating

that spin alignment is dominated by the coupling between Fe and Fe, and Sm and Fe. Since the wave function of rare earths is very much localized, the small value of n_{SS} is well expected. However, the comparable magnitudes of n_{FF} and n_{SF} observed in this work are rather unexpected. Normally, in RE-TM systems, the coupling between transition metals is significantly greater than that between rare earth and transition metal, since the wave function of transition metals is much more extended and hence the exchange interactions between transition metals are more direct [5].

As the B content increases, the value of n_{FF} increases continuously from 10370 to 13730. The values of n_{SF} and n_{SS} also tend to increase with increasing B content. It is noted that the maxima in these values occur at $x = 0.02$, but the physical meaning of this is considered to be not significant. Due to the itinerant nature of Fe atoms, the present B content dependence of n_{FF} should be explained by considering the change in electronic state with the addition of B, but this is not a simple problem and beyond of the scope of the present paper. One possible and simple reason for this may be related with the change in the

Table II. The exchange interaction coefficients n_{ij} , the Curie temperature (both measured and calculated) T_c and the fitting parameter R for the $\text{SmFe}_2\text{-B}$ system.

	n_{FF}	n_{SF}	n_{SS}	Measured T_c (K)	Calculated T_c (K)	R (%)
$\text{Sm}_{0.33}\text{Fe}_{0.67}$	10370	10590	2150	620	626	1.74
$(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.99}\text{B}_{0.01}$	11700	11500	2490	587	592	1.48
$(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.98}\text{B}_{0.02}$	11730	15500	3400	582	588	1.61
$(\text{Sm}_{0.33}\text{Fe}_{0.67})_{0.97}\text{B}_{0.03}$	13730	14090	2790	567	570	1.44

distance between Fe atoms with the addition of B. Exchange interaction between Fe atoms is known to be a sensitive function of the distance between the atoms [5, 16]. In the distance range of ferromagnetic exchange interaction, the coupling increases with increasing separation between the atoms. One good and recent example is the results observed in amorphous binary Gd-Fe alloys by Yano [3] and Yano *et al.* [19], and in amorphous ternary Gd-Fe-B₂₀ alloys by Hassini *et al.* [11]. They observed an increase of n_{FF} with the increase of Gd content, and explained this increase of n_{FF} by the increased Fe-Fe separation. The increase of Fe-Fe separation is well expected in amorphous Gd-Fe alloys, since the atomic radius of Gd is greater than that of Fe. The present result for the increase of n_{FF} with the increase of B content can be explained similarly, since B is expected to reside in interstitial sites, and hence to increase the Fe-Fe separation with the increase of the B content.

It appears not easy to explain the very large values of n_{SF} observed in this work. Normally, the coupling between RE and TM is much smaller than that between transition metals. In *crystalline* REFe_2 compounds (RE = Tb, Dy, Ho, Er, and Tm), for example, the absolute magnitude of the coupling coefficient between RE and TM is 10~20 % of that between transition metals [18]. This can be understood since the exchange interaction between RE and TM is known to be indirect through polarized spins in the conduction band. The trend appears similar in amorphous RE-TM alloys, but the gap seems to decrease significantly with amorphization, although, with a paucity of available data, it is not easy to say something definite at this stage. In an Er-Fe system where data are available in both crystalline (ErFe_2) and amorphous (ErFe_3) states, the absolute magnitude of the coupling coefficient

between Er and Fe is 10.8 % of that between Fe and Fe in crystalline state [18], but it becomes comparable to that between Fe atoms (70 %) in amorphous state [10]. Another example is an amorphous GdFe_2 (the data for crystalline GdFe_2 are not available, however), the exchange coupling between Gd and Fe is about 40 % of that between Fe atoms [6]. Considering this, the present results for n_{SF} are thought to be not that extraordinary. This is further supported from reported results in *crystalline* Sm-Fe intermetallics. In a SmFe_3 compound, the values of n_{FF} and n_{SF} are respectively 14500 and 11200 [17]. Also in a $\text{Sm}_2\text{Fe}_{17}$ compound, the values of n_{FF} and n_{SF} are 3890 and 3200, respectively [20]. These results clearly indicate that the exchange coupling between Sm and Fe is actually comparable to that between Fe atoms, which is in agreement with the present results.

The Curie temperature decreases from 620 to 567 K as the B content increases from 0 to 0.03. The reduction of T_c is considered to be large; the reduction is greater than 50 K at 3 at.% B. This is actually expected from the large change in the exchange interaction coefficients as a function of the B content. A good agreement is achieved between experimental and calculated Curie temperatures over the whole B content range, as can be seen from Table II.

V. Conclusions

In this work, the mean field theory has been used to obtain exchange coupling between constituent elements in melt-spun ribbons of two amorphous alloy systems: $(\text{Dy}_{0.33}\text{Fe}_{0.67})_{1-x}\text{B}_x$ with $x = 0, 0.05, 0.1$, and 0.15 , and $(\text{Sm}_{0.33}\text{Fe}_{0.67})_{1-x}\text{B}_x$ with $x = 0, 0.01, 0.02$, and 0.03 (in atomic fraction). In the case of the $\text{DyFe}_2\text{-B}$ system, the magnetization direction of Dy is antiparallel to

that of Fe and the magnetization is dominated by the Dy sublattice magnetization. The exchange coupling between Fe ions are greatest, while that between Dy ions is negligible. The exchange couplings between Fe ions, and between Dy and Fe increase with increasing B content, the increase of the latter being much greater than the former. Resultantly, the exchange coupling between Dy and Fe becomes about one half of that between Fe ions at the highest B content. In the case of the SmFe₂-B system, the sign of all the couplings are positive, indicating ferromagnetic alignment between the spins. The total magnetization is dominated by the Fe sublattice magnetization. The exchange couplings between Fe ions, and Fe and Sm are comparable to each other, but they are much greater than that between Sm ions. Considering no direct coupling between Fe and Sm, the high exchange coupling between Fe and Sm is rather unexpected, but the present results may be supported from the following two facts; (1) The exchange coupling between Fe and Sm in crystalline Sm-Fe alloys is slightly small compared with that between Fe ions, and (2) amorphization generally increases the coupling between Fe and RE (rare earth) in RE-Fe alloys. In both systems, the increase of the exchange coupling between Fe ions may be explained by the increase of the Fe-Fe separation with the increase of the B content. As the B content increases, the magnetization decreases over the whole temperature range, and the Curie temperature also decreases.

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References

- [1] R. Hasegawa, B. E. Argyle and L. J. Tao, AIP Conf. Proc. **24**, 110 (1975).
- [2] Y. Mimura, N. Imamura, T. Kobatashi, A. Okada, Y. Kushiro, J. Appl. Phys. **49**, 1208 (1978).
- [3] K. Yano, J. Magn. Magn. Mater. **208**, 207 (2000).
- [4] A. Gangulee and R. J. Kobliska, J. Appl. Phys. **49**, 4896 (1978).
- [5] R. Hajjar and M. Mansuripur, IEEE Trans. Magn. **25**, 4021 (1989).
- [6] P. Hansen, in Handbook of Magnetic Materials, Edited by K. H. J. Buschow (North-Holland, Amsterdam, 1991), vol. 6, Chap. 4.
- [7] P. Chaudhari, J. J. Cuomo, R. J. Gambino, IBM J. Res. Dev. **17**, 66 (1973).
- [8] A. E. Clark, in Ferromagnetic Materials, Edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980), vol.1, Chap. 7.
- [9] P. Hansen, C. Clausen, G. Much, M. Rosenkranz and K. Witter, J. Appl. Phys. **66**, 756 (1989).
- [10] J. Benjelloun, A. Itri, H. Lassri, M. Omri and M. Ayadi, J. Alloys Comp. **299**, 27 (2000).
- [11] A. Hassini, H. Lassri, A. Bouhdada, M. Ayadi, R. Krishnan, I. Mansouri and B. Chaker, Physica B, **275**, 295 (2000).
- [12] J. Jensen and A. Mackintosh, Rare Earth Magnetism, Clarendon Press, Oxford, p. 13.(1991).
- [13] A. E. Clark, Appl. Phys. Lett. **23**, 642 (1973).
- [14] J. J. Rhyne, J. H. Schelleng, N. C. Koon, Phys. Rev. B **10**, 4672 (1974).
- [15] J. Benjelloun, H. Oukris, H. Lassri, M. Baran, R. Krishnan, M. Omri, M. Ayadi, J. Alloys Comp. **283**, 66 (1999).
- [16] R. C. OHandley, in Amorphous Metallic Alloys, Edited by F. E. Luborsky (Butterworths, London, 1983), Chap. 14.
- [17] J. F. Herbst and J. J. Croat, J. Appl. Phys. **53**, 4304 (1982).
- [18] Y. J. Tang, X. P. Zhong and H. L. Luo, J. Magn. Magn. Mater. **127**, 378 (1993).
- [19] K. Yano, Y. Akiyama, K. Tokumitsu, E. Kita and H. Ino, J. Magn. Magn. Mater. **214**, 217 (2000).
- [20] W. H. Qiao, Q. Wang, X. P. Zhong and H. L. Luo, J. Magn. Magn. Mater. **110**, 170 (1992).

비정질 REFe₂-B (RE=Dy, Sm) 합금 리본에서 평균장 이론에 의한 교환상호작용 계산

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(2001년 6월 5일 받음, 2001년 6월 13일 최종수정본 받음)

급속응고에 의해 제조된 비정질 (Dy_{0.33}Fe_{0.67})_{1-x}B_x (x = 0, 0.05, 0.1, 0.15) 및 (Sm_{0.33}Fe_{0.67})_{1-x}B_x (x = 0, 0.01, 0.02, 0.03) (원자 비) 합금 리본에 대하여 자화(온도 곡선)을 구하였으며, 이를 평균장 이론에 근거한 이론적인 식을 사용하여 fitting함으로써 구성 원소들 사이의 교환상호작용의 크기를 계산하였다. DyFe₂-B계의 경우, Dy와 Fe 사이의 교환상호작용은 음의 부호를 가지는데, 이는 Dy와 Fe의 자화 방향이 반대임을 의미한다. Fe 원소 사이의 교환상호작용이 가장 크며, 반면에 Dy 원소들 사이의 상호작용이 가장 작게 나타났다. SmFe₂-B계의 경우, 계산된 교환상호작용은 전부 양의 부호를 가지는데, 이는 스핀들 사이에 강자성 상호작용이 있음을 의미한다. Fe 원소들 사이의 상호작용과 Fe와 Sm 사이의 상호작용은 매우 크며, 그 크기 또한 유사하다. Fe와 Sm 사이의 상호작용이 큰 것은, 이 원소들 사이의 상호작용이 간접적임을 고려할 때 의외의 결과이며, 비정질 Sm-Fe 합금의 고유 성질인 것으로 생각된다. 두 합금계 모두에서 Fe 원소들 사이의 교환상호작용은 B 함량이 증가함에 따라 증가하였는데, 이는 B 함량의 증가에 따라 Fe-Fe 간격이 증가하였기 때문으로 생각된다. 두 합금계 모두에서 자화와 큐리온도는 B의 함량이 증가함에 따라 감소하였다.