Magnetic properties of amorphous $Fe_{80-x}R_xB_{20}$ (with R= Pr, Nd, Sm, Gd, Dy, Ho, Er and Tm)

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Abstract

Amorphous $Fe_{80-x}R_xB_{20}$ alloys (with R=Pr, Nd, Sm, Gd, Dy, Ho, Er and Tm) have been prepared by melt spinning and their magnetic properties have been studied. The Gd moment at 4.2 K is found to be $7\mu_B$ which agrees with the theoretical value indicating a collinear spin structure. The moments of the other rare-earth are lower than the theoretical value, implying a conical spin structure. The mean-field theory has been used to explain the temperature dependence of the magnetization. The exchange interactions between Fe-Fe and Fe-R atom pairs have also been evaluated.

1. Introduction

Amorphous alloys based on rare-earth (R) and transition metals (T) and metalloids (M), such as T-R and T-R-B, show interesting magnetic properties and have been studied in the past by several authors [1-3]. They offer the possibility to study various aspects of amorphous magnetism, such as, random anisotropy, nature of magnetic interactions, the dilution of magnetic moments etc… In contrary to crystalline materials these aspects can be investigated in continuous concentration range of the rare-earth metal as well as in relation with the low local symmetry that is characteristic for the amorphous state. Rare-earth metal atoms with an orbital moment are known to give rise to large random anisotropy in amorphous alloys [4].

In T-R and T-R-B amorphous alloys, it is generally accepted that there are three types of exchange interactions, namely, the T-T interactions between magnetic moment of the T sublattice, The R-T intersublattice interactions and R-R interactions between the magnetic moments within the R sublattice. The T-T interactions are positive and lead to the ferromagnetic coupling between the 3d moments. These interactions primarily govern the temperature dependence of the 3d magnetization and the value of the Curie temperature. The R-T interactions are significantly smaller than the T-T interactions; in most cases, they determine the thermal stability of 4f moments, the temperature dependence of R sublattice anisotropy and the rare earth magnetic moment configuration. Finally, the R-R interactions are generally the weakest. The strength of the R-T exchange interactions can be determined experimentally by several methods. This constant can be determined from analysis of the ordering temperature [5-7], of the high-field magnetization

measured on free powders [8-9] and from the inelastic neutron scattering spectra by using a well established spin wave model [10-12], if the spin of the transition metal is known from other experimental methods or from band structure calculations. On the basis of these experimental results, some systematic trends such as the variation of the strength of R-T interactions with rare-earth and transition-metal nature, and with the rare-earth concentration have been found [13]. In accordance with the model proposed by Brooks and al. [14], Duc and al. [13] have proposed that the origin of these phenomena may be found in the variable degree of the 3d-5d hybridization from one compound to another. The value for R-T exchange coupling in different compounds has been correlated with the value of the 3d magnetic moment.

In order to study the influence of the addition of R on the various magnetic properties of amorphous Fe-B alloys, and to describe the character of the magnetic order in all these alloys, whereby a ferromagnetic coupling of the 3d and 4f moments of Fe and R ions is to be expected when R is a light rare-earth element and a ferrimagnetic coupling when R is a heavy rare-earth element, all those amorphous $Fe_{80-x}R_xB_{20}$ alloys with $0 \le x \le 16$ have been prepared and their magnetic properties were investigated. We have analyzed the thermal variation of magnetization in terms of the mean field model and have obtained several parameters such as the exchange integrals, etc

2. Experimental

Amorphous $Fe_{80-x}R_xB_{20}$ ribbons with $0 \le x \le 16$ and R=Pr, Nd, Sm, Gd, Dy, Ho, Er and Tm, were obtained in an inert atmosphere of Ar by melt spinning technique. The purity of the starting materials was 99.99% for B, R and 99.999% for Fe. Argon ejection pressure of 2 to 5 kPa and a substrate speed of 35 m/s were employed. The melt ejecting tubes were made of quartz glass with an ejecting orifice of about 0.4 mm in diameter. The ribbon samples were about 30 μm thick with different widths varying from 2 to 4mm. The X-ray diffraction (XRD) was used to verify the amorphous structure. The exact chemical composition of the samples was determined by electron probe microanalysis. The magnetization was measured by vibrating sample magnetometer (VSM) from 4.2 to 300 K in magnetic field up to 1.8 T. Curie temperature was determined using a VSM from 300 to 900 K in a small magnetic field of about 10^{-2} T.

3. Results and discussion

Magnetic studies

The technical saturation is attained for H < 1 T at all temperatures. The temperature dependence of the magnetization in applied field H = 1.8 T is displayed in fig.(1). All the light rare-earth alloys show ferromagnetic behavior, the heavy rare-earth alloys are ferrimagnetic and their M(T) curves show compensation points for some alloys with x bigger than the compensation concentration. The total magnetization of the $Fe_{80-x}R_xB_{20}$ alloy consists of the sum of partial magnetizations according to the following scheme:

$$M = M_{Fe} + M_R; \quad \mu_a = ((80-x)\mu_{Fe}+x\mu_R)/100 \quad (1)$$

for all the light rare-earth alloys and

$$M=|M_{Fe} - M_R|; \quad \mu_a = | (80\text{-}x)\mu_{Fe} - x\mu_R |/100 \quad (2)$$

for all the heavy rare-earth alloys.

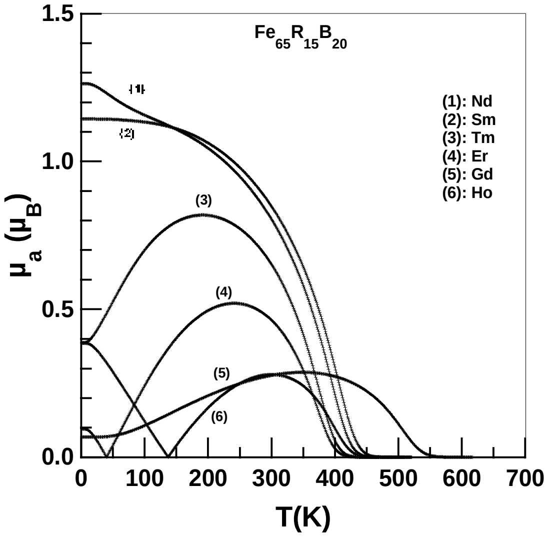

Figure (1): The temperature dependence of the magnetization for alloys $Fe_{65}R_{15}B_{20}$ with different rare-earth.

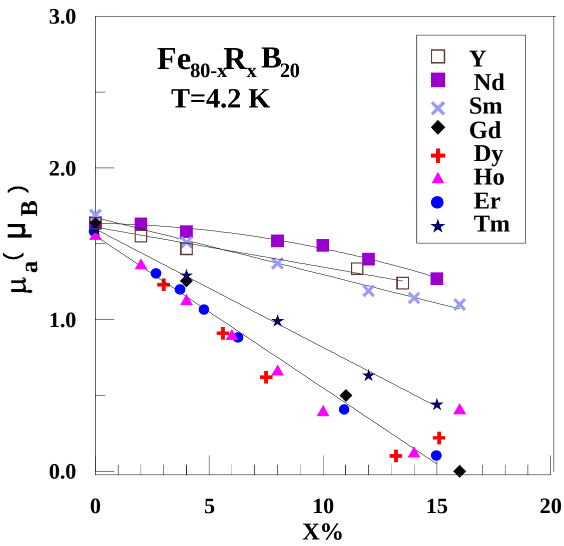

Figure (2): Concentration dependence of μ_a at 4.2 K for alloys $Fe_{80-x}R_xB_{20}$ with different rare-earth.

The concentration dependence of the magnetization (μ_a in μ_B at 4.2 K) is shown in fig.(2). There is a linear decrease in μ_a for each series of amorphous $Fe_{80-x}R_xB_{20}$ alloys versus the concentration of rare-earth R (except Nd). One notices that the slope of the decreasing for amorphous alloys with light rare-earth is smaller than that of amorphous with heavy rare-earth. The above results are characteristic of the antiferromagnetic interaction between R and Fe atoms for the heavy rare-earth alloys and of the ferromagnetic interaction for those with light rare-earth.

For small concentrations (x<8%) of R, the iron moment is not perturbed. So taking the value of μ_{Fe} = 2 μ_B obtained from the alloy with x = 0% and substituting in Eqs.(1) and (2), it is possible to determine μ_R for different rare-earth, which are plotted in fig.(3).This figure also shows a comparison between the measured values witch are a projection along the applied field and the theoretical R^{+3} moments. This value, smaller than the theoretical value $g_RJ_R\mu_B$, indicates the non collinearity in R spin structure, a phenomenon which is well known for rare-earths with strong random local anisotropy.

Using this value of μ_R then we can calculate the value of μ_{Fe} for other composition in R of all series in rare-earth alloys fig.(4). It was found that μ_{Fe} decreased when R concentration increases. This is attributed on one hand to the increased filling of 3d bands of Fe by the sp electrons from B since now the relative concentration of B with respect to Fe increases, and the other hand, to the hybridization of the 5d and 3d orbital.

In our case, we have the contribution of two sub-networks at the magnetic anisotropy in one hand the R (except Gd) which is a rare earth possessing an important magnetic anisotropy, and on the other hand the Fe for which the mean magnetic momentum

is lower than that of the metallic counterpart.

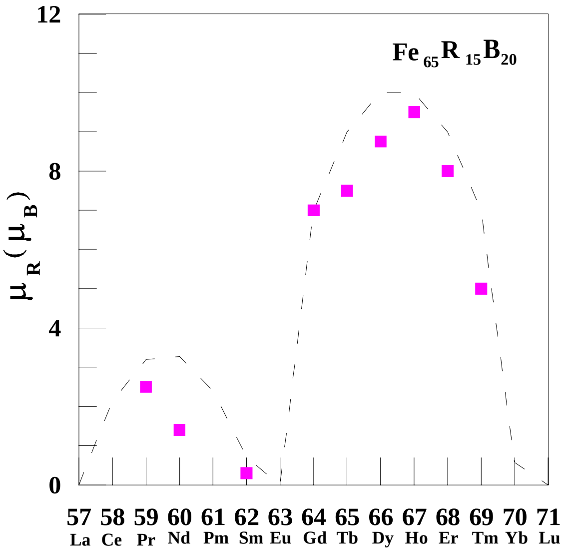

Figure (3): Saturation moment per R atom, corrected values and theoretical free R^{+3} ion values for alloys $Fe_{65}R_{15}B_{20}$.

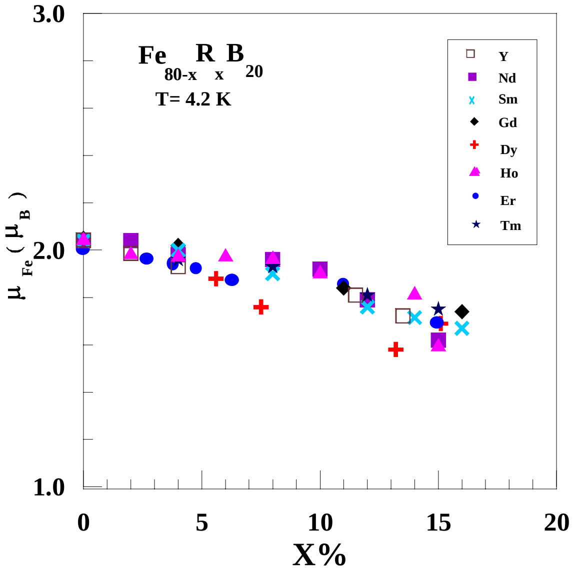

Figure (4): The iron moment as a function of R content at 4.2 K.

This situation shows that the Fe orbital momentum is incompletely quenched in the alloys, then we will find a spin-orbit interaction which will give rise to a local magnetic anisotropy in the Fe sub-network. The magnetic random local anisotropy constant evaluated by us using the effective anisotropy model is a function of the inter-sublattice exchange interactions and the sub-networks' local anisotropies [15].

The fig.(5) show the dependence of the μ_{Fe} on the rare-earth atomic number for x=15%, the average Fe moment is μ_{Fe} = $(1.7\pm0.1)\mu_B$, we can deduce that the fluctuation for this concentration of the μ_{Fe} is smaller for the light rare-earth alloys than that of the heavy rare-earth alloys.

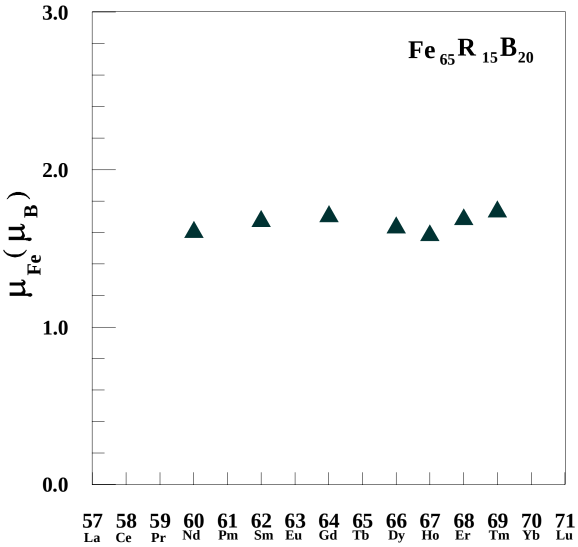

Figure (5): The Fe moment for alloys $Fe_{65}R_{15}B_{20}$ with different rare-earth

The ordering temperatures of ferromagnets and ferrimagnets will be referred to as Curie temperature in the following. For all samples they are shown in fig.(6). As expected there is a peak at $Fe_{80-x}Gd_xB_{20}$ for each concentration x=8% and x=15%.

The high value in Curie temperature for this alloy show that the exchange interaction between Fe atoms is by far the most important compared to those of the series, to the R-Fe or R-R ones and largely determines the value of T_c.

The Curie temperature T_c of amorphous $Fe_{80-x}R_xB_{20}$ alloys was determined from the thermomagnetic data. The values of T_c thus obtained are reported in fig.(7). The strong descent of T_c with x for different series of amorphous alloys can be attributed to the sensitive dependence of the Fe moment of the local environment.

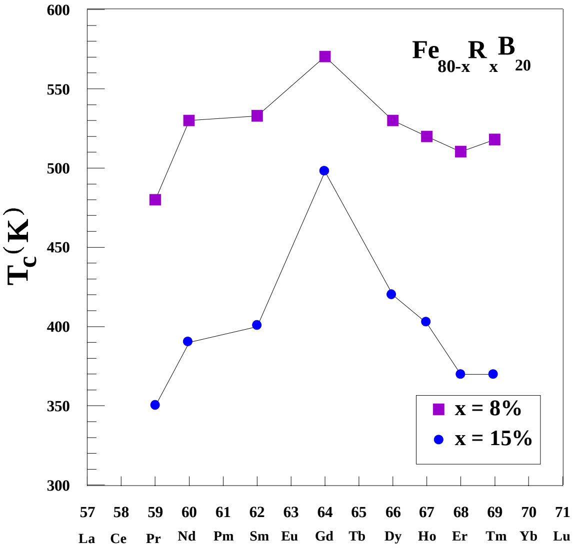

Figure (6): The variation of Curie temperature for all $Fe_{80-x}R_xB_{20}$ alloys for x=8% and x=15%.

The mean field theory has been used in the past by several authors to calculate the temperature dependence of the magnetization in many amorphous rare-earth(R)-transition metal (T) alloys [16-20]. We have performed such an analysis of the temperature dependence of the magnetization in amorphous $Fe_{80-x}R_xB_{20}$ alloys. The alloys moment μ_a can be written as:

$$\mu_a = |(80-x)\mu_{Fe} \pm x\mu_R|/100 \quad (3)$$

where (+) for the light rare earth alloys and (-) for the heavy rare earth alloys, J_R is the total momentum of rare-earth, μ_B is the Bohr magneton, g_i is the Landé factor (i = Fe, R) and we take $g_{Fe} = 2$.

$\mu_{Fe}(T)$ and $\mu_R(T)$ were assumed to be expressed by the Brillouin function:

$$\mu_R(T) = \mu_R(0)\, B_J(g_R J_R \mu_B H_R/K_B T) \quad (4)$$

$$\mu_{Fe}(T) = \mu_{Fe}(0)\, B_S(g_{Fe} S_{Fe} \mu_B H_{Fe}/K_B T) \quad (5)$$

where S_{Fe} and J_R are the Fe spin momentum and R total angular momentum respectively. The molecular fields H_{Fe} and H_R are given by:

$$H_{Fe} = 2J_{Fe\text{-}Fe}Z_{Fe\text{-}Fe}S_{Fe}/g_{Fe}\mu_B + 2J_{Fe\text{-}R}Z_{Fe\text{-}R}(g_R-1)J_R/g_{Fe}\mu_B \quad (6)$$

$$H_R = 2J_{R\text{-}Fe}Z_{R\text{-}Fe}(g_R-1)S_{Fe}/g_R\mu_B + 2J_{R\text{-}R}Z_{R\text{-}R}(g_R-1)^2J_R/g_R\mu_B \quad (7)$$

where Z_{ij} is the coordination number taken to be $Z_{Fe\text{-}Fe} = Z_{R\text{-}Fe} = 12(80-x)/80$, $Z_{Fe\text{-}R} = Z_{R\text{-}R} = 12x/80$ (8)

$J_{Fe\text{-}Fe}$, $J_{Fe\text{-}R}$ and $J_{R\text{-}R}$ are the exchange constants for Fe-Fe, Fe-R and R-R interactions, respectively. We assume that $J_{R\text{-}R} = 2\ 10^{-23}$J [19]

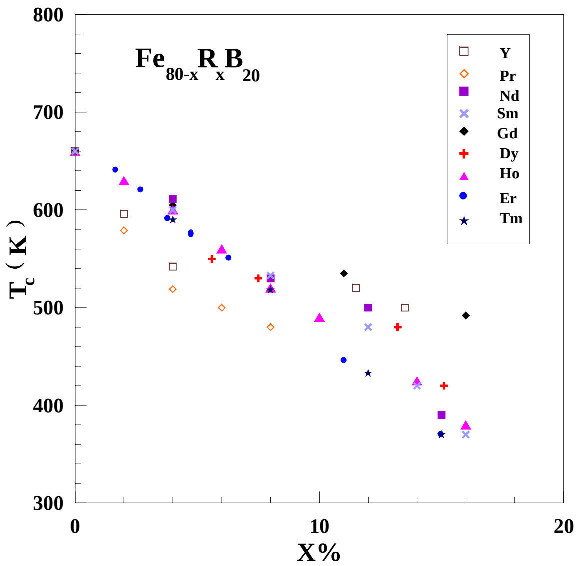

Figure (7): The R concentration dependence of Curie temperature.

The temperature dependence of the saturation magnetization can be analyzed in terms of the mean-field theory. Using the spin values determined for different series of alloys and

adjusting the exchange interactions $J_{Fe\text{-}Fe}$ and $J_{Fe\text{-}R}$, the sublattice magnetization M_{Fe}, M_R and the saturation magnetization $M=|M_{Fe}\pm M_R|$ can be calculated. Table 1 shows the various parameters obtained from the analysis of the data using the models described. From these fits the exchange interactions were extracted as function of the R content.

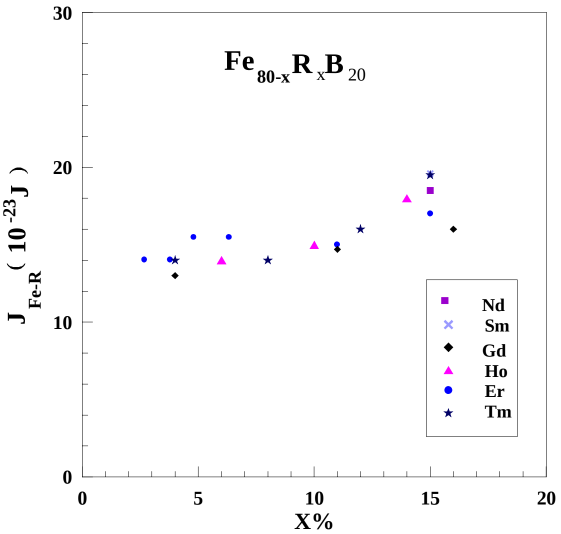

Figure (8): The R concentration dependence of the exchange interactions $J_{Fe\text{-}R}$.

It is seen that $J_{Fe\text{-}R}$ increases, when the R concentration increases, fig.(8). A similar increase in J_{RT} has been reported in intermetallic compounds and amorphous alloys[16,18]. The 3d-5d interactions depend critically on 3d-5d hybridization according to Brooks et al [14]. Therefore the increase in $J_{Fe\text{-}R}$ would indicate an increase in 3d-5d hybridization when the concentration relative to R is decreased. The exchange fluctuation caused by the structural disorder affect the shape of the M curves. This influence has been omitted because at least one additional parameter would have to be adjusted.

The fig. (9) show a dependence between the exchange interactions $J_{Fe\text{-}R}$ and moment of iron μ_{Fe}, we can remarked that the $J_{Fe\text{-}R}$ increases with the decreasing of the μ_{Fe}. These results suggest thus that hybridization effects in rare-earth transition metal alloys increase with percentage of R alloyed elements.

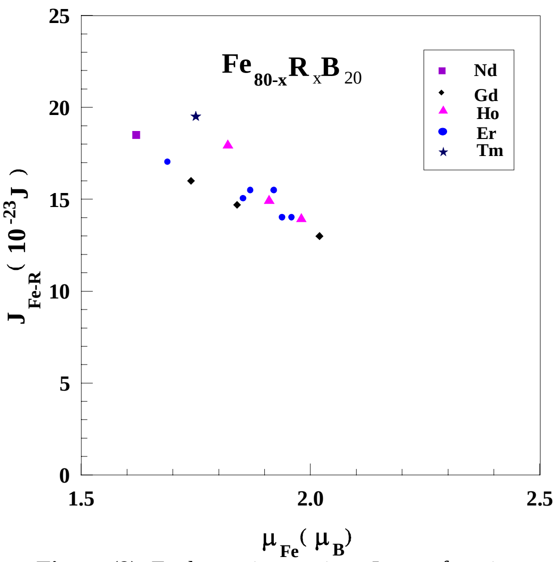

Figure (9): Exchange interactions $J_{Fe\text{-}R}$ as function μ_{Fe}.

Table 1:
Some magnetic parameters of $Fe_{65}R_{15}B_{20}$ alloy at 4.2K.

R	$\mu_a(\mu_B)$	S_{Fe}	$J_{Fe\text{-}Fe}$ (10^{-23}J)	$J_{Fe\text{-}R}$ (10^{-23}J)	T_c^{exp} (K)
Nd	1.27	0.81	56.1	18.5	390
Sm	1.12	0.845	53.3	19.6	400
Gd	0.08	0.86	56.6	16	497.7
Ho	0.40	0.80	53.7	18	402.1
Er	0.10	0.85	47.5	17	370
Tm	0.39	0.875	43	21	370

4. Conclusion

The magnetic properties of $Fe_{80\text{-}R}R_xB_{20}$ (with R=Pr, Nd, Sm, Gd, Dy, Ho, Er and Tm) alloys were investigated with respect to their composition and temperature dependence. The saturation magnetization was analyzed in terms of

mean field theory. The Fe moment, the R moment, the Curie temperature and the exchange interactions $J_{Fe\text{-}Fe}$ and $J_{Fe\text{-}R}$ were evaluated. Therefore the increase of $J_{Fe\text{-}R}$ with an increase of R content is attributable to such an enhancement of the 3d-5d band hybridization.

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