JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS, Vol. 10, No. 12, December 2008, p. 3497 - 3500

Magnetic exchange interactions in $R_xFe_{80-x}B_{20}$ amorphous alloys based on Sm and Gd

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Amorphous $R_xFe_{80-x}B_{20}$ alloys (with R = Sm and Gd), in ribbon form, have been prepared by melt spinning and their magnetic properties have been studied ($0 \le x \le 18$). The mean field theory has been used to explain the temperature dependence of the magnetization. The Fe moment, the R moment, the Curie temperature and the exchange interactions between Fe-Fe and Fe-R atom pairs have been evaluated. The Sm moment at 4.2 K is found to be 0.73 μ_{BP} which is two times smaller than the theoretical value of 1.5 μ_{BP} indicating a conical spin structure. On the other hand, the Gd moment at 4.2 K is found to be 6.85 μ_{BP} which agree with the theoretical value indicating a collinear spin structure. The increase of Fe-R interaction with an increase of R content is attributed to an enhancement of the 3d-5d band hybridization.

(Received November 7, 2008; accepted November 27, 2008)

Keywords: Iron base alloys, Ternary alloys, Samarium alloy, Gadolinium alloys, Magnetism, Exchange interactions

1. Introduction

In amorphous R-T-M and, particularly, in $R_xT_{80-x}B_{20}$ alloys, the magnetic order in the transition-metal sublattice is mainly due to the strong exchange coupling between T-3d transition metals (Fe, Co, Ni) atoms while that between R ones is likely to be established through the intersublattice atomic exchange coupling [1-3]. The magnetic properties of the rare-earth metals differ from those of the iron-group transition elements because of the nature of the 4f and 3d electronic states responsible for the magnetism. In the rare-earth, the magnetic 4f electrons are buried deeply in the ionic core and are well shielded from crystal field effects. Both spin and orbital motion contribute to the total angular momentum and several of the heavier rare-earth has the highest atomic magnetic moments of all the elements. The moment of each atom is coupled to its neighbours by a relatively weak indirect exchange mechanism (conduction electron polarization) which results in low Curie temperature. The 3d electrons are responsible for the magnetic characteristics of the iron-group transition metals. These electrons are in the outermost shell and are highly perturbed by the crystal field. The field quenches orbital moments and the total moment is due to spin contributions alone. The resulting magnetic moment per atom is smaller than that of the gadolinium-group rare-earths. However, direct overlap of the 3d orbitals provide a strong direct exchange coupling of the individual atomic magnetic moments and the Curie temperatures will be relatively high. With the large magnetic moments per atom of the gadolinium group and the high Curie temperature associated with iron group, magnetic alloys of the transition metals and the rare-earths may have desirable magnetic characteristics.

It is well known that the exchange interaction between T and R is ferromagnetic for R lighter than Sm and antiferromagnetic for R heavier than Gd. In the case of the exchange interaction constants and magnetic moment investigation, the presence of strong magnetic anisotropy may disturb simple analysis. In this sense, the metal such as Gd, and, even Sm, as R is suitable for investigation because Gd (Sm) has little anisotropy as a result of negligible angular moment, $L \cong 0$. Moreover, it was shown by Krishnan et al. that in sufficiently high fields, the antiferromagnetic coupling between the T and R moments is broken [4, 5]. As a consequence, the magnetic properties investigation in high-fields yields several important magnetic parameters, such as, the exchange field on the R atoms, and the intersublattice molecular-field coefficient, etc. The value for the T-R exchange coupling parameter obtained can serve as an input parameter for the analysis of magnetization curves [6].

In this work, the magnetic properties of amorphous $R_xFe_{80-x}B_{20}$ alloys with $0 \le x \le 18$ were prepared by melt spinning technique and their magnetic properties studied in the low temperature range 4-300 K at high-fields, $B \le 30$ T. The experimental results were analysed in the terms of the exchange interactions Fe–Fe, Fe–R, and R–R, and have discussed the thermal behaviour of the magnetic transition in the high field regime.

2. Experimental

Amorphous ribbons of $R_xFe_{80-x}B_{20}$ alloys with R = Sm and Gd were prepared by melt spinning of appropriate amounts of the metals [5]. The ribbons were about 5 mm wide and about 25 μm thick. The investigated samples consist of small pieces of discoid form, cut away from the

ribbons, with diameter of 5 mm. The exact chemical composition was determined by Electron Probe Microanalysis (EPMA). The amorphous state was verified by X-ray diffraction (XRD). The magnetization of sample was measured using a vibrating sample magnetometer (VSM) from 4.18 K to 600 K in magnetic field up to 2 T. Curie temperature was determined using VSM from 300 to 900 K in a small magnetic field of about 0.02 T.

3. Results and discussion

In $R_xFe_{80-x}B_{20}$ alloys both the R and Fe atoms contribute to the magnetic properties. The temperature dependence of magnetization can do information about the magnetic behaviour of the alloy. Fig.1 shows some typical results on the temperature dependence of the magnetic moment (in units of Bohr-Procopiu magneton - μ_{BP}) for x = 12 of the $R_xFe_{80-x}B_{20}$ samples. The R-Fe-B alloys show a ferromagnetic behaviour with easy tendence of Gd alloys to a ferromagnetic behaviour (Fig.1).

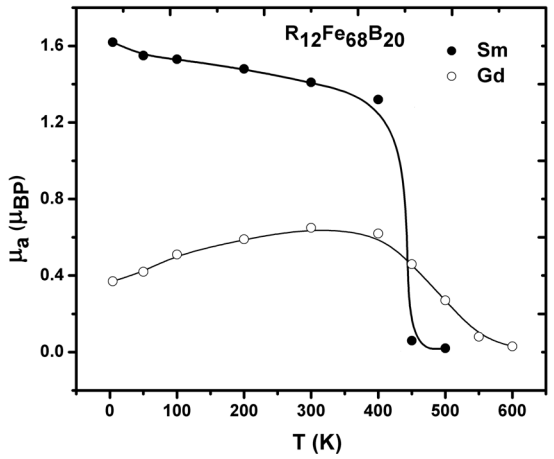

Fig.1. Typical temperature dependence of $R_xFe_{80-x}B_{20}$ alloy magnetization for x=12

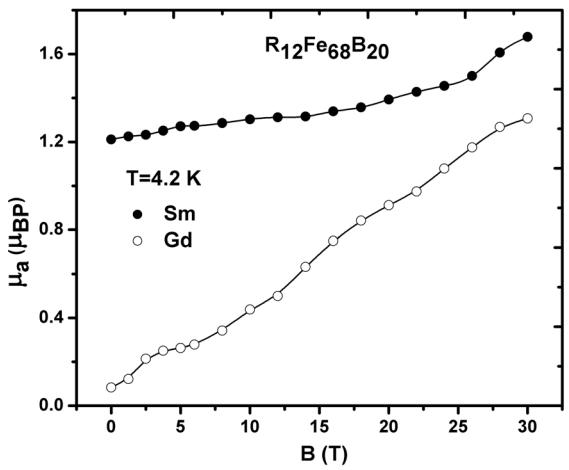

Fig. 2. High-field magnetization process of $R_{12}Fe_{68}B_{20}$ alloys at low temperature of 4.2 K.

This behaviour is confirmed by the high-field magnetization up to 30 T at low temperature of 4.2 K (Fig.2) for a typical composition of x = 12 at. %. In Fig.3 is shown the concentration dependence of the magnetic moment of the $R_xFe_{80-x}B_{20}$ samples at 4.2 K. The magnetization, at low temperature and in low-field, decreases showing a linear behaviour. The decrease is more rapidly for Gd than for Sm that is really a light rare-earth element. This linear decrease of magnetization can be associated with a ferromagnetic behaviour. Generally, the amorphous R-T-M alloys in low magnetic fields and at room temperature have a collinear ferromagnetic structure. This structure must be understood as a two-subnetwork structure containing two distinguishable groups of atoms, R and T atoms. Their spins are coupled antiparallel via a strong exchange coupling between the 3d electrons of the transition metal and 5d electrons of the rare-earth where the later are polarised by the 4f shell. This leads to a parallel alignment of the T and R moments for light rare-earths and to an antiparallel alignment for the heavy rare-earths [6]. The above results in Figs 3 and 4 are characteristic of the ferromagnetic interaction between Sm (Gd) and Fe atoms. The ferromagnetic order is perturbed by the presence of B and R-Fe interactions are different for Gd-Fe-B, i.e. more rapidly than Sm-Fe-B alloys (Fig. 3).

The magnetic moment μ_a of the $R_xFe_{80-x}B_{20}$ alloy arises from the total contribution of the moments of Fe - μ_{Fe} and R (Sm, Gd) - μ_R because they align ferromagnetically. One can hence write [5,7]:

$$\mu_a = [(80-x)\,\mu_{Fe} + x\,\mu_R]/100, \qquad (1)$$

where x is rare-earth concentration.

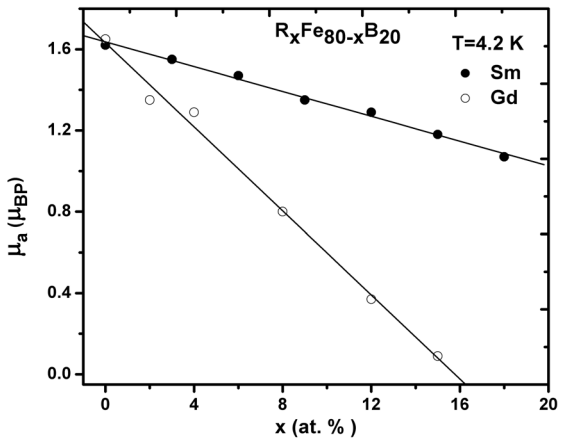

Fig. 3. Rare-earth element concentration dependence of magnetization for $R_xFe_{80-x}B_{20}$ alloys at 4.2 K.

We have calculated the Sm magnetic moment μ_{Sm} for the critical composition x = 55.5 at. % Sm when magnetic moment of Sm-Fe-B alloy becomes $\mu_a = 0$ by taking the value of μ_{Fe}=1.66 μ_{BP} obtained for the alloy with x = 0 and using it in Eq. (1). We found μ_{Sm} = 0.73 μ_{BP}, which is

lower than the theoretical ($g_RJ_R\mu_{BP}$) value of $1.5\mu_{BP}$ [8]. This value, smaller than the theoretical value $g_RJ_R\mu_{BP}$, indicates the non collinearity in Sm spin structure, a phenomenon which is well known for rare-earths with strong random local anisotropy [9]. Analogously, Gd magnetic moment μ_{Gd} was calculated for the critical composition x = 16.2 at. % Gd when magnetic moment of Sm-Fe-B alloy becomes $\mu_a = 0$ by taking the value of μ_{Fe}=1.74 μ_{BP} obtained for the alloy with x=0. We found μ_{Gd} = 6.85 μ_{BP}, which is close to the theoretical ($g_RJ_R\mu_B$) value of $7\mu_{BP}$, indicating the collinear spin structure for Gd. However, the fact that the calculated value μ_{Gd} is smaller than the theoretical value can means the existence of a sperimagnetic (non-collinear spin) structure within the Gd sublattice at higher concentrations [5].

Using the value of μ_R we can calculate the value of μ_{Fe} for different composition of R (Sm, Gd). The variation of the Fe moment with the R concentration at 4.2 K is shown in Fig.4.

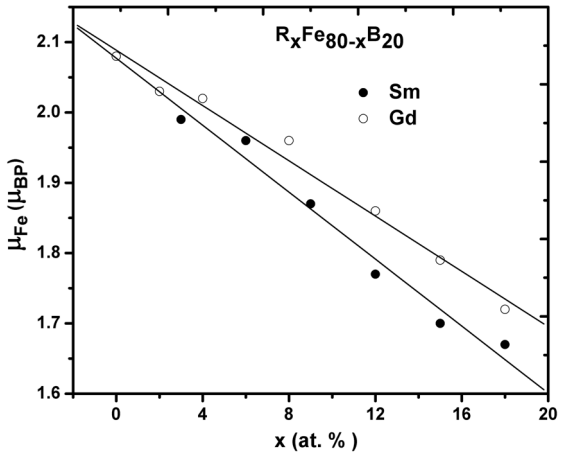

Fig. 4. Iron magnetic moment as a function of R content at 4.2 K.

The average Fe moment μ_{Fe}, in $R_xFe_{80-x}B_{20}$ alloys is determined by the structural and chemical environment and by hybridisation and mixing effects. The magnetic moment of Fe, μ_{Fe}, decreases 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 5d and 3d orbital. In the case of Sm alloys we have the contribution of two sub-networks at the magnetic anisotropy in one hand the Sm, on the other hand the Fe for which the mean magnetic moment is lower than that of the metallic counterpart.

This situation shows that Fe orbital moment is incompletely quenched in the alloy, and 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 [9] using effective anisotropy model is a function of the inter-sublattice exchange interactions and the sub-networks' local anisotropies. The above assumption is not valuable in the case of Gd alloys because Gd is a rare earth possessing an important magnetic anisotropy [5].

The Fe moment decrease with the Gd concentration increase can be understood as due to an increase of the 3d spin-up band filling in Fe atom by the $6s^2/5d$ electrons of Gd. The rapidly decrease of μ_{Fe} for Gd alloys may be a result of the fact that with concentration increasing Gd change from a collinear structure to a non-collinear (sperimagnetic) ones which arises from the strong random local anisotropy [5].

The magnetic moments lined up in the magnetic alloys are constantly vibrating due to heat fluctuation. As the temperature rises, the heat fluctuation becomes stronger and proportional to this, the magnetic moment alignment becomes disrupted. Finally, this order is completely lost and the temperature at which this happens is the Curie temperature. The Curie temperature in $R_xFe_{80-x}B_{20}$ alloys for different R concentration is shown in Fig. 5.

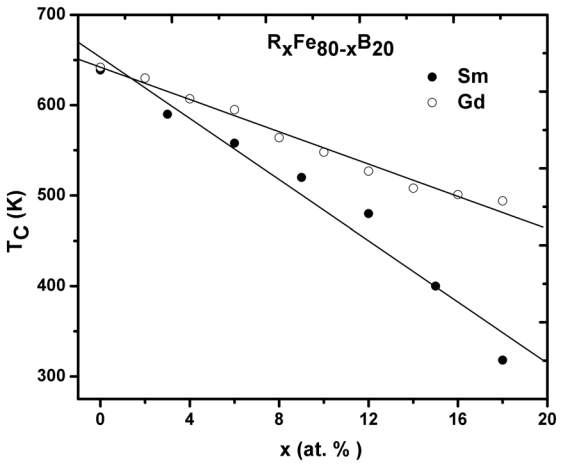

Fig. 5. Rare-earth (Sm, Gd) concentration dependence of the Curie temperature in $RxFe80_{-x}B_{20}$ alloys.

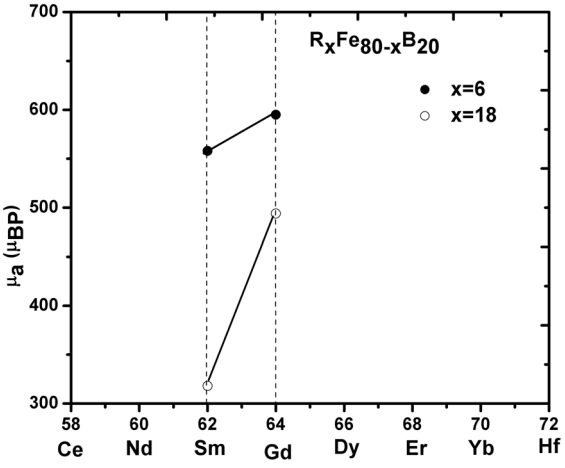

Fig. 6. Curie temperature of $R_xFe_{80-x}B_{20}$ alloys for two more different concentration of rare-earth element R.

The high Curie temperature, $T_C > 300$ K, in this kind alloys shows that the exchange interaction between Fe atoms is the most important compared to those of the R-Fe or R-R ones and largely determines the value of T_C.

Fig. 6 shows the Curie temperature for Sm and Gd for two more different concentrations, namely x = 6 at. % and x = 18 at. %.

For low R concentration the difference in T_C for Sm and Gd is small and this difference is bigger at high Gd concentrations indicating a non-collinear spin structure for Gd. This structure arises from the strong random local anisotropy and the amorphous Gd-Fe-B alloys tend to have a ferromagnetic behaviour.

4. Conclusions

The magnetic properties of $R_xFe_{80-x}B_{20}$ alloys were investigated with respect to their composition and temperature in low and high field. The Fe moment, the R moment and the Curie temperature were evaluated. Fe orbital moment is incompletely quenched in the alloy, and 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 effective anisotropy model is a function of the inter-sublattice exchange interactions and the sub-networks' local anisotropies.

The increase of Fe-R interaction with an increase of R content is attributed to an enhancement of the 3d-5d band hybridization.

Acknowledgements

This work was partially supported by European Commission through the Human Potential Programme under the contract no.HPRI-1999-CT00030 and by Grenoble High Magnetic Field Laboratory-GHMFL through the Project no.MA0903. The authors would like to acknowledge the technical support and valuable discussions of Prof. G. Chouteau, Joseph Fourier University, Dr. Ilya Sheikin and Dr. S. Diaz, CNRS-GHMFL, Grenoble, France.

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