

Dedicated to Professor Marin Ivaşcu's 80<sup>th</sup> Anniversary

## MAGNETIC PROPERTIES AND EXCHANGE INTERACTIONS IN RARE-EARTH-IRON-BASED COMPOUNDS

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*Abstract.* By considering the 4f–5d–3d exchange interactions model, the contributions of local 4f–5d and short–range 5d–3d interactions to R5d band polarizations in  $R_2Fe_{14}A$  ( $A=B,C$ ) and  $R_2T_{17}$  ( $T=Fe,Ni$ ) compounds were evaluated. The induced R5d band polarizations by 4f–5d interactions are linearly dependent on De Gennes factor, but having different slopes for light and heavy rare–earth compounds, respectively. The R5d band polarizations resulting from 5d–3d short range interactions are dependent on the number of neighbouring magnetic atoms to a given R one and their moments. The Fe3d–Fe3d exchange interactions were analysed in correlation with the site occupancy and volume variations. For a large number of interstitial compounds, the effective exchange interactions,  $J_{eff}$ , shows a similar volume dependence, with  $d\ln J_{eff}/d\ln v = 18$ . The above data were correlated with the Curie temperatures, of the studied compounds, as experimentally determined. Finally, the magnetic properties of (Pr,Nd)–(Fe,Co,Si)–B nanocomposites were reported.

*Key words:* rare earth compounds, exchange interactions, band structure.

### 1. INTRODUCTION

The  $R_2Fe_{17}$  compounds, where R is a rare-earth or yttrium have low Curie temperatures, ranging from 260 K ( $R=Ce$ ) to 477 K ( $R=Gd$ ). The iron magnetic moments in  $R_2Fe_{17}$  series are almost as large as those of iron metal [1, 2]. The rare-earth anisotropy is rather weak and the contributions of rare-earths are not sufficiently strong to counteract the iron sublattice anisotropy, at room temperature, which favours an easy plane magnetization.

The  $R_2Fe_{14}A$  ( $A=B,C$ ) compounds have technical applications, being the hard magnetic components of R–Fe–B or R–Fe–C permanent magnets [3]. Despite the good price/performance ratio, they have some disadvantages, namely low Curie temperatures and high sensitivity to corrosion. The Curie temperatures of these compounds amount only to about  $T_C \cong 580$  K, which result in high negative temperature coefficients of magnetic induction and coercive field. The  $T_C$  values

show linear dependences on the De Gennes factor  $G = (g_J - 1)^2 J(J + 1)$ , with different slopes for light and heavy rare-earths, respectively.

The exchange interactions in  $R_2Fe_{17}A$  ( $A=B,C$ ) are rather complex. The exchange interactions involving iron (Fe–Fe–type) are of short range and strongly dependent on the interatomic distances [4,5], and usually described in terms of the Néel–Slater curve [6] – Fig. 1. According to this curve, the interactions between iron atoms situated at distances smaller than 2.45–2.50 Å are negative, and positive, respectively, if the distances between them are greater than above values. The negative interactions in  $R_2Fe_{17}$  and  $R_2Fe_{14}A$  ( $A=B,C$ ) are not satisfied since they are smaller than positive ones and consequently a considerable magnetic energy is stored. This brings about their low Curie temperatures.

The exchange interactions, of R–Fe type, can be described by the Campbell’s phenomenological model [7]. In this model, the 4f electrons of the rare–earth polarize their 5d bands and there are short range exchange interactions with neighbouring iron atoms. The local 4f–5d exchange interactions are positive and the 4f and 5d moments will align parallel. An additional contribution to 5d band polarization resulting from 5d–3d short range exchange interactions, through hybridization effects, was shown in  $RF_e_2$  compounds to be proportional to the number of neighbouring transition metal atoms to an R one and their moments [8].

The intensities of exchange interactions between rare–earths, in intermetallic compounds are rather small. These take place by means of conduction electrons or/and 5d–5d short range interactions. The last type of interactions were shown to be present in  $DyNi_{5-x}Al_x$  [9] or  $(Gd_xY_{1-x})Ni_5$  [10] compounds.

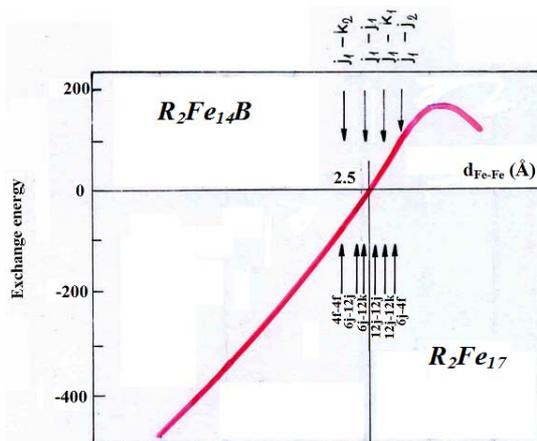


Fig. 1 – Néel-Slater curve and the distances between iron atoms situated in sites located around 250 Å.

In the first part of paper we analyse the short–range exchange interactions between iron atoms and the ways to increase the Curie temperatures by iron substitutions in sites involved in negative exchange interactions or/and as a result

of volume changes. The part played by R5d band polarizations in analysing the exchange interactions between rare-earths and iron, is a fundamental question. Consequently, this matter will be also analysed in correlation with crystal structure and Curie temperatures. In the last part, the magnetic properties of  $\text{Pr}_x\text{Nd}_{10-x}\text{Fe}_{75}\text{Co}_{9-y}\text{Si}_y\text{B}_6$  nanocomposites will be reported.

## 2. BAND STRUCTURE CALCULATIONS

The band structure calculations were carried out by using the *ab-initio* tight-binding linear muffin-tin orbital method (TB-LMTO) in the atomic sphere approximation (ASA). The detailed procedures of calculations were described elsewhere [11, 12]. In the framework of the local density approximation (LDA), the total electronic potential is the sum of external, Coulomb and exchange correlation potentials [13]. The functional form of the exchange correlation energy, used in the present work, was the free electron gas parameterization of von Barth and Hedin [14]. Relativistic corrections were also included.

## 3. EXCHANGE INTERACTIONS BETWEEN IRON ATOMS

As mentioned in introduction, both in  $\text{R}_2\text{Fe}_{17}$  and  $\text{R}_2\text{Fe}_{14}\text{A}$  ( $\text{A}=\text{B},\text{C}$ ) systems are present negative exchange interactions. These are not satisfied since the corresponding atoms are involved also in positive exchange interactions having higher intensity than the negative ones. Consequently, the Curie temperatures are much smaller than 50 % of the pure iron ordering temperature.

The increase of the Curie temperature in  $\text{R}_2\text{Fe}_{14}\text{A}$  ( $\text{A}=\text{B},\text{C}$ ) [15] and  $\text{R}_2\text{Fe}_{17}$  [1,16] compounds can be obtained by replacing iron atoms involved in negative exchange interactions or/and increase of the cell volume [16–18] – Fig. 2. Two opposite effects determine the Curie temperatures as result of substitutions. The first, is determined by the diminution of the negative exchange interactions, even if the iron was substituted by a non-magnetic atom. The second one, contributing to decrease of  $T_C$  values, is a result of magnetic dilution and diminution of iron moments due to hybridization effects with *e.g.* p states of substituting element. The first contribution dominates in case of substitution by Si, Ga, Cu, Mo, Ti, V, leading to the increase of the Curie points.

The presence of interstitial atoms as N or C, determines the increase of the cell volume and consequently of the distances between iron atoms, above the critical value of 2.50 Å. As a result, there is a high increase of the Curie temperature. In this case the second effect resulting from magnetic dilution and diminution of iron magnetic moments, as result of iron substitution, can be clearly evidenced. As example, in  $\text{Sm}_2\text{Fe}_{17-x}\text{Si}_x\text{C}_2$ , even for small silicon content, the magnetic ordering temperature decreases sensitively.

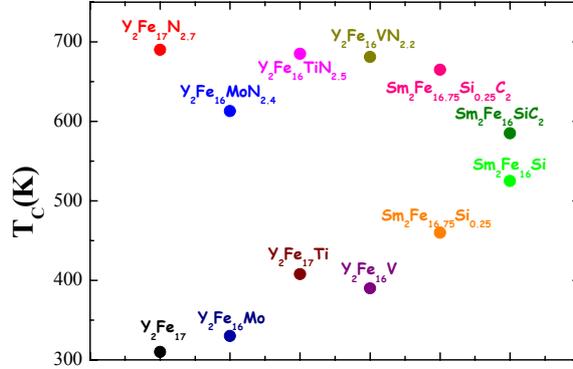


Fig. 2 – The Curie temperatures of  $Y_2Fe_{17-x}M_x$  ( $M=Fe,Mo,Ti,V$ ) and their nitrides as well as of  $Sm_2Fe_{17-x}Si_xC_y$  with  $y = 0$  and  $2$  compounds.

The iron moments in  $R_2Fe_{14}A$  ( $A=B,C$ ) do not show a high degree of itinerancy as evidenced by the ratio between the number of spins determined from effective moments and the saturation ones [20].

The volume expansion of the lattice leads to the increase of Curie temperatures, effective exchange interactions, respectively. The variation of the Curie temperatures with volume can be characterized by the parameter  $\Gamma = d \ln T_C / d \ln v$ . The dependence of the  $\Gamma$  values on the Curie temperatures in some  $R_2Fe_{17}$  nitrides and carbides are shown in Fig. 3. In the limit of experimental errors a linear relation is obtained:

$$\Gamma = a + b T_C, \tag{1}$$

with  $a = 38$  and  $b = -0.06 \text{ K}^{-1}$ .

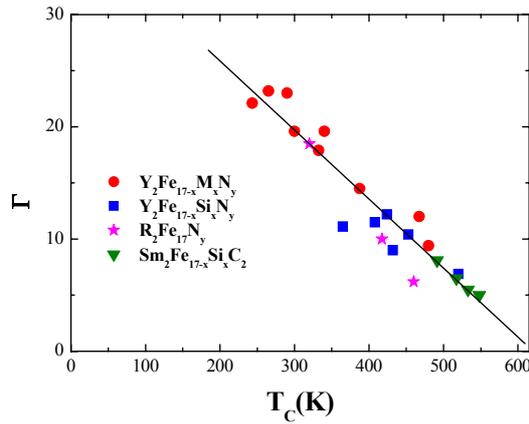


Fig. 3 – The dependence of  $\Gamma = d \ln T_C / d \ln v$  on the Curie temperatures in  $Y_2Fe_{17-x}M_xN_y$ ,  $Y_2Fe_{17-x}Si_xN_y$  [16],  $R_2Fe_{17}N_y$  [19] and  $Sm_2Fe_{17-x}Si_xC_2$  [17] compounds.

Starting from an essential localized model of iron [21], has been evidenced that the dependence of  $\Gamma$  on Curie temperatures can be described by the relation [22]:

$$\Gamma = \frac{5}{3} + 2 \frac{d \ln J_{\text{eff}}}{d \ln v} + \frac{5}{8} \frac{k_B N_0 g^2 I^2}{S(S+1) g^2 I_b} T_c, \quad (2)$$

where  $N_0$  is the Avogadro number,  $g$  the Landé factor,  $I$  the effective intra-atomic exchange integral which is reduced from its bare value  $I_b$ , by many body correlation effects and  $J_{\text{eff}}$  the effective exchange interaction.

By comparing the relations (1) and (2) a value  $d \ln J_{\text{eff}} / d \ln v = 18$  was obtained. The variation with volume of the effective exchange can be obtained also from magnetic measurements on the above compounds. By using the molecular field approximation, the  $J_{\text{eff}}$  values were determined. Taking into account the volume variations, values  $\Gamma \cong 12$  have been obtained in case of  $\text{Y}_2\text{Fe}_{17-x}\text{M}_x\text{N}_y$  and  $\Gamma = 17$  for  $\text{Sm}_2\text{Fe}_{17-x}\text{Si}_x\text{C}_2$  system. These are in reasonable agreement with the value obtained from relations (1) and (2).

The above data show a rather strong variation with the volume of exchange interactions and as the results of the Curie temperatures.

The neutron diffraction studies [3] and band structure calculations [21] showed that the mean iron moments in  $\text{R}_2\text{Fe}_{14}\text{B}$  compounds with light rare-earth were nearly constant, differing by only 1 %. The same behavior was shown in heavy rare-earth compounds although in the last case these were smaller, by 4–5 %, as compared to the values determined in light rare-earth compound. Thus in the first approximation, for a given subseries, the Fe3d–Fe3d exchange interactions are expected to have similar contributions in determining the Curie temperatures.

#### 4. RARE-EARTH-IRON EXCHANGE INTERACTIONS

The dependences of the Curie temperatures of  $\text{R}_2\text{Fe}_{14}\text{A}$  ( $\text{A}=\text{B,C}$ ) and  $\text{R}_2\text{T}_{17}$  ( $\text{T} = \text{Fe,Ni}$ ) compounds on De Gennes factor,  $G = (g_r - 1)^2 J(J+1)$  are shown in Fig. 4. Nearly linear variations are shown with different slopes for light and heavy rare-earth compounds. For the studied system the ratio of their slopes is  $\cong 1.4$ . As discussed already the contribution of Fe3d–Fe3d exchange interactions, for a given subseries, to the Curie temperatures can be considered as constant. Consequently, the dependence of  $T_c$ , on De Gennes factor, can be attributed mainly to R–Fe exchange interactions, mediated by R5d band.

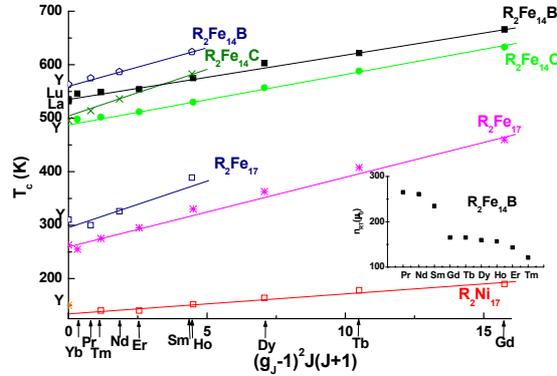


Fig. 4 – The Curie temperatures of  $R_2Fe_{14}A$  ( $A=B,C$ ) [3] and  $R_2T_{17}$  ( $T=Fe,Ni$ ) [1] compounds as function of the De Gennes factor.

The R5d bands are polarized. The computed R5d band polarizations as function of  $G = (g_j-1)^2 J(J+1)$  factor are plotted in Fig. 5 for  $R_2Fe_{14}A$  ( $A= B,C$ ) and in Fig. 6 for  $R_2T_{17}$  ( $T=Fe,Ni$ ) compounds. As in case of the Curie temperatures, linear dependences are shown, described by the relation:

$$M_{5d} = M_{5d}(0) + \alpha G, \tag{3}$$

we denoted by  $M_{5d}(0)$ , the R5d band polarizations obtained by extrapolation at  $G = 0$  and determined by 3d–5d short range exchange interactions. The second term describes the contributions due to the local 4f–5d interactions.

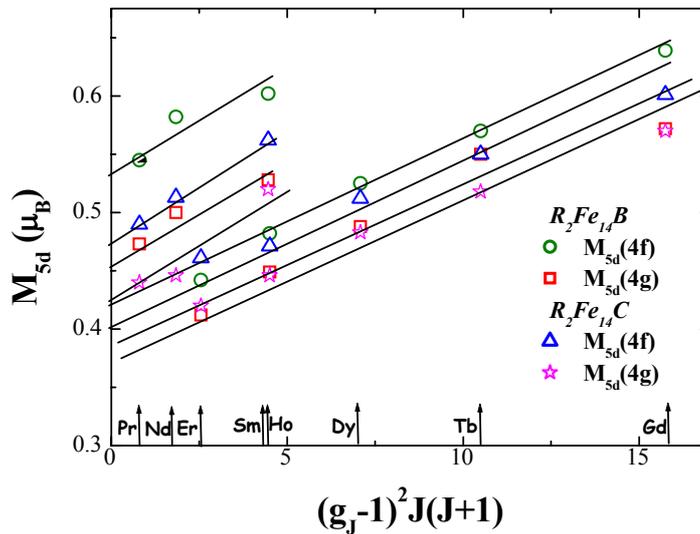


Fig. 5 – The R5d band polarizations at R(f) and R(g) sites in  $R_2Fe_{14}A$  ( $A=B,C$ ) compounds.

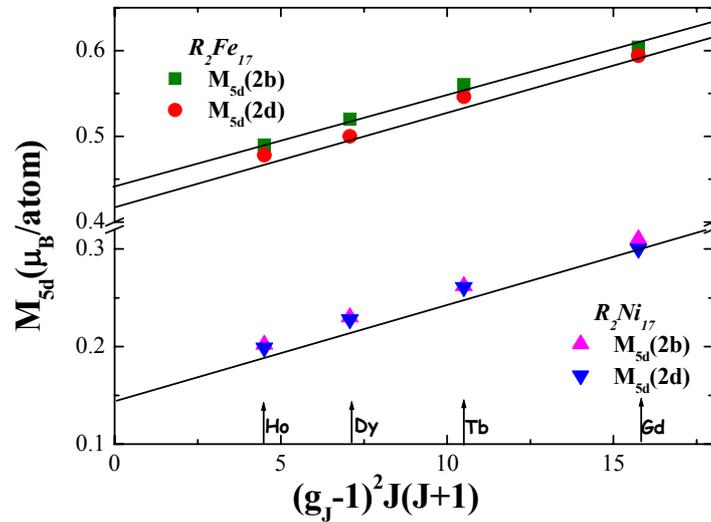


Fig. 6 – The R5d band polarizations in heavy rare-earth compounds of  $R_2T_{17}$  ( $T=Fe, Ni$ ) type.

The slopes of the dependences are higher in light rare earth compounds as compared to heavy rare earth ones, their ratio being  $\cong 1.5$ , near the same value as the slope ratio of Curie temperatures. The observed behavior can be correlated with local 4f–5d interactions. The  $r_{4f}$  radii of light rare-earths Pr–Sm decrease from 1.11 Å to 0.96 Å with a mean value of  $1.03 \pm 0.08$  Å. Smaller values can be shown for the subseries Gd to Tm, with variations of  $r_{4f}$  between 0.85 Å and 0.78 Å, with a mean value of  $0.82 \pm 0.03$  Å [24]. Although 4f–5d interactions are expected to vary along a subseries, on going from Sm– to Gd– compound there is a diminution of  $r_{4f}$  radius by 12 % which can affect substantially the local exchange interactions. Thus, it is expected to be stronger 4f–5d exchange interactions in light-rare earth compounds than in heavy rare-earth ones. Consequently, the R5d band polarizations induced by local exchange is supposed to be also higher in light rare earth compounds. The 4f–5d exchange interactions can be changed also along a subseries, although no so much as when passing from Sm to Gd. This last effect, along a subseries, cannot be unambiguously determined since of data scattering obtained from band structure calculations.

The estimated values of  $n_{R-Fe}$  exchange interactions constants [25], evidenced sudden changes in the  $R_2Fe_{14}B$  series, when Sm is substituted with Gd at the limit of the two subseries. Also variations of the  $n_{R-Fe}$  along a subseries has been shown, in agreement with the above discussion.

Since the R–Fe exchange interactions are mediated by R5d band, higher variations of Curie temperatures are expected for light rare-earth ones, for the same  $G$  values, as experimentally shown.

The increased R–Fe contributions to the exchange interactions in light rare-earth compounds can be also correlated with the evolution of iron magnetic moments. We showed already [26] that in RFe<sub>2</sub>– and RFe<sub>3</sub>– based compounds was a strong connection between the variations of exchange field,  $\Delta H_{\text{exch}}$ , and that of iron moment,  $\Delta M_{\text{Fe}}$ ,  $\Delta M_{\text{Fe}} = \beta \Delta H_{\text{exch}}$  with  $\beta = (2 \cdot 10^2)^{-1} \mu_{\text{B}}/T$ . Both by neutron diffraction studies [3] and band structure calculations [3, 23], higher values of iron moments in light R<sub>2</sub>Fe<sub>14</sub>A (A=B,C) compounds were shown. As example in Nd<sub>2</sub>Fe<sub>14</sub>B and Sm<sub>2</sub>Fe<sub>14</sub>B, the mean iron moments were 2.265 and 2.274  $\mu_{\text{B}}$ /atom, respectively, while in Ho<sub>2</sub>Fe<sub>14</sub>B and Er<sub>2</sub>Fe<sub>14</sub>B values of 2.188 and 2.157  $\mu_{\text{B}}$ /atoms have been determined. Thus, there were differences of only 1–2 % along a subseries, while the iron moments differed by  $\cong 5$  % in the two subseries. Taking into account the correlation between the iron moments and the exchange field, previously determined, the exchange fields acting on iron, is expected to be greater by 20 T in light rare earth R<sub>2</sub>Fe<sub>14</sub>B compounds as compared to that in heavy rare-earth ones.

The  $M_{5d}(0)$  band polarizations obtained by band structure calculations are dependent on the rare-earth lattice sites – Fig. 5. This behavior can be correlated with the intensity of 5d–3d short-range exchange interactions with atoms situated in the neighborhood to an R one. Admitting also the presence of 5d–5d short range interactions, the corresponding system can be described by the Hamiltonian [9]:

$$\mathcal{H} = 2J_{3d5d} S_{5d}(0) \sum_i S_{3d_i}(0) - 2J_{5d5d} S_{5d}(0) \sum_j S_{5d_j}, \quad (4)$$

where  $J_{3d-5d}$  and  $J_{5d-5d}$  are exchange integrals, characterizing the 3d–5d and 5d–5d interactions with  $i$  Fe atoms and  $j$  R neighbour atoms, to a given R, and  $S_{5d}(0)$  and  $S_{3d}(0)$ , the spin values of Fe3d and R5d bands for  $G = 0$ .

The Hamiltonian (4) can be analysed in molecular field approximation. The effects of 5d–3d and 5d–5d exchange interactions are equivalent to an internal field  $H_{\text{eff}}$ , acting on R atoms,  $H_{\text{eff}} = n_{5d-3d} M_{\text{Fe}} + n_{5d-5d} M_{5d}$ , where  $n_{3d-5d}$  and  $n_{5d-5d}$  are the molecular field coefficients describing the interactions between R–Fe and R–R atoms respectively. In iron based compounds, the last term can be neglected. In R<sub>2</sub>Ni<sub>17</sub> compounds, where the nickel moments are of the same order of magnitude as R5d band polarizations, this term must to be also considered. The effective field will induce an increase of R5d band polarization, similar as for Fe3d band in magnetic rare earth compounds [26]. We suppose that the 5d–3d short range exchange interactions and the corresponding hybridization effects are not modified along a given series. The R5d band has a rather great extension, of 5.33 Å [24]. In addition the distances between the R and Fe atoms are only little changed along a series. Thus, in the above assumptions, for a given R site, the  $M_{5d} \propto \sum_i z_i M_{\text{Fe}_i}$  when 5d–5d interactions were neglected. The  $\sum_i z_i M_{\text{Fe}_i}$  values were evaluated for different R sites by using the iron magnetic moments determined from band structure calculations. In case of R<sub>2</sub>Ni<sub>17</sub> compounds the

possible R5d–R5d interactions were also considered. The R5d and Y4d band polarizations as function of  $\sum_i n_i M_i$  are plotted in Fig. 7a,b. Linear variations were shown both for 2:14:1 and 2:17 compounds, respectively, but having different slopes.

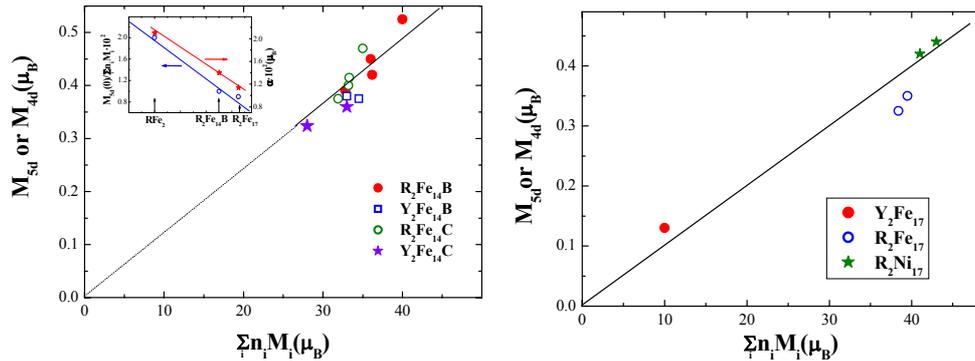


Fig. 7 – The  $M_{5d}(0)$  and  $M_{4d}$  band polarizations, as a function of  $\sum_i n_i M_i$ , in: a)  $R_2Fe_{14}A$  ( $A=B,C$ ); b)  $R_2T_{17}$  ( $T=Fe,Ni$ ) compounds. The ratio  $M_{5d}(0)/\sum_i n_i M_i$ , as function of the transition metal content corresponding to one rare-earth, is given in inset of (a).

The induced polarizations determined by short range interactions, divided by the magnetizations of the neighbor atoms to an R one,  $M_{5d}(0)/\sum_i M_i$  decrease when the number of magnetic transition metal atoms, in a given compound, is higher – Fig.7a inset. A similar variations is shown also for  $\alpha$  parameter from relation (3).

The present analysis of R5d band polarizations and their correlation with Curie temperatures, involved some assumptions generated by the complexity of exchange interactions. Thus the 4f–6s exchange interactions were neglected. Also, the effects of 4f–5d and 5d–3d interactions were considered as additive, although these can be reciprocal influenced. In spite of the above assumptions, the present analysis gives a rather good descriptions of the contributions of 4f–5d–3d exchange interactions to R5d band polarizations as well as a method to separate the corresponding 4f–5d and 5d–3d contributions.

## 5. (Pr,Nd)–(Fe,Co,Si)–B NANOCOMPOSITE MAGNETS

The alloy composition and processing conditions have a strong influence on the magnetic properties of R–Fe–B magnets [3, 27]. For isotropic microcrystalline R–Fe–B alloys, the remanence  $B_r$  is limited to half of the saturation induction,

although the coercivity is rather high. The grain refinements into the nanocrystalline regime enhance the remanence above the theoretical Stoner-Wohlfarth value ( $B_s/2$ ) for randomly oriented non-interacting grains [28].

The phase compositions and magnetic properties of nanocrystalline  $\text{Pr}_x\text{Nd}_{10-x}\text{Fe}_{75}\text{Co}_{9-y}\text{Si}_y\text{B}_6$  alloys with  $x = 2, y = 4$  and  $x = 4, y = 4$  were studied. The samples were obtained by high energy ball milling. The above system is constituted essentially from a hard magnetic phase having composition  $(\text{Pr,Nd})_2(\text{Fe,Co,Si})_{14}\text{B}$  and soft magnetic ones. As discussed already, the substitution of Fe by Co or Si will increase the Curie temperatures. The presence of cobalt decreases the magnetic anisotropy while silicon, in small quantities, increases the anisotropy field [3].

The XRD pattern of the  $\text{Pr}_2\text{Nd}_8\text{Fe}_{75}\text{Co}_4\text{Si}_5\text{B}_6$  shows the presence of the 2:14:1 hard magnetic phase and of the soft ones,  $\alpha\text{-Fe, (Fe-Co)}$  and  $\text{Fe}_2\text{B}$  (Fig. 8).

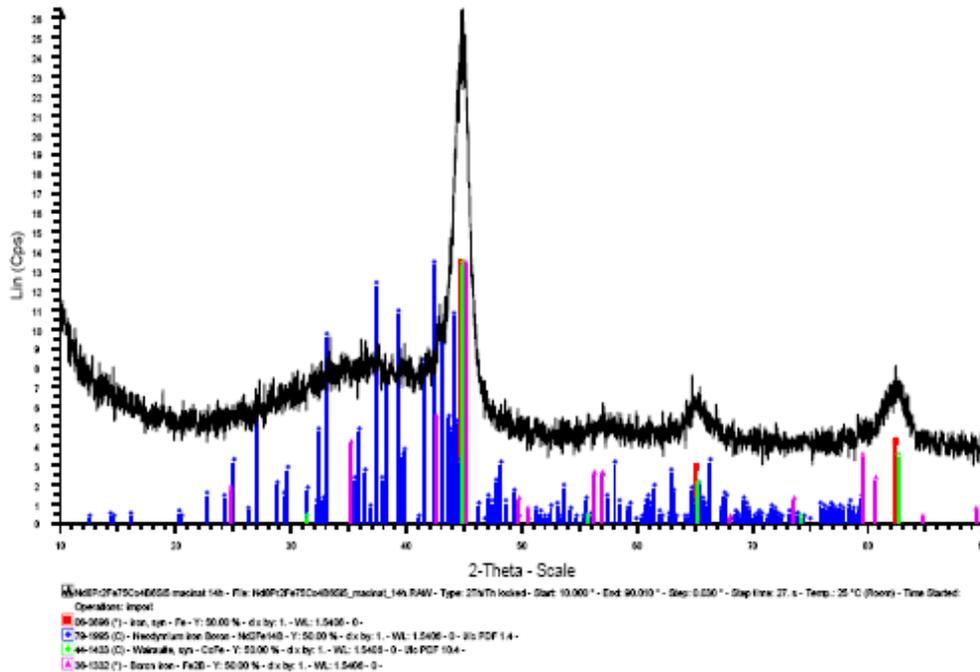


Fig. 8 – XRD pattern of nanocrystalline  $\text{Pr}_2\text{Nd}_8\text{Fe}_{75}\text{Co}_4\text{Si}_5\text{B}_6$  alloy.

The magnetization isotherms of the nanocrystalline samples, aligned in the external field, saturate at relative low field ( $\cong 1$  T). The saturation magnetizations of the samples with  $x = 2, y = 4$  is of  $182 \mu_B/\text{f.u.}$  while of the sample with  $x = 4$  and  $y = 3$ , it was  $179 \mu_B/\text{f.u.}$  The above values are very close to the expected ones based on the initial composition of alloys, showing that the Fe and Co magnetic moments, in this system, are little changed by alloying. The sample with  $x = 2$  and

$y = 4$  shows a remanent induction  $B_r \cong B_s/2$  (Fig. 9). Consequently, we expect that the grain size is somewhat greater than the  $\cong 40$  nm, as expected for obtaining an enhanced remanence. The sample with  $x = 4$  and  $y = 3$  shows a remanent induction  $B_r = 0.55 B_s$  suggesting a good exchange coupling between hard and soft magnetic phases. The coercive field is of 475 kA/m ( $\mu_0 H_c \cong 0.6$  T), higher than determined in sample with  $x = 2, y = 4$ . The greater value is probably related to the higher silicon content in the sample.

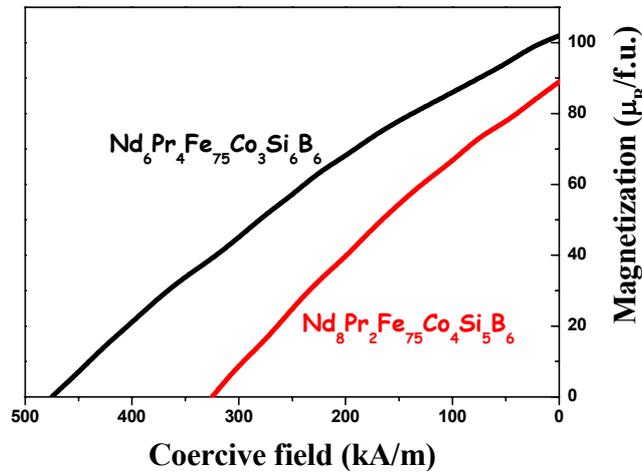


Fig. 9 – Demagnetization curves for  $\text{Pr}_x\text{Nd}_{10-x}\text{Fe}_{75}\text{Co}_{9-y}\text{Si}_y\text{B}_6$  alloys with  $x = 2, y = 4$  and  $x = 4, y = 3$ .

## 6. CONCLUSIONS

The composition dependences of the Curie temperatures in  $\text{R}_2\text{Fe}_{14}\text{A}$  ( $\text{A}=\text{B},\text{C}$ ) and  $\text{R}_2\text{T}_{17}$  ( $\text{T}=\text{Fe},\text{Ni}$ ) compounds can be well described, by considering 4f–5d–3d exchange interaction model. The Fe3d–Fe3d exchange interactions were analysed in correlation with the distances between iron atoms, site substitutions as well as volume variations. The effective exchange interaction constant,  $J_{\text{eff}}$ , shows a volume variation of the form  $\text{dln}J_{\text{eff}}/\text{dln}v = 18$ , for the nitrides and carbides of the 2:17 studied series. Values close to the above were obtained also by analysing their magnetic properties, in molecular field model.

The exchange interactions between rare-earths and iron (nickel) are mediated by R5d band. The polarization of this band is determined by two additive effects, determined by local 4f–5d and 3d–5d short range interactions. The induced R5d band polarizations, by 4f–5d exchange interactions, are linearly dependent on De Gennes factor with different slopes for light- and heavy-rare earth compounds. The  $M_{5d}(0)$  polarizations resulting from 5d–3d short range interactions are dependent

on the number and magnetic moments of the atoms situated in the first coordination shell to a given R one. For each series of compounds as  $R_2Fe_{14}A$  ( $A=B,C$ ),  $R_2T_{17}$  ( $T=Fe,Ni$ ) or  $RT_2$  ( $T=Fe,Co,Ni$ ) [29], similar behavior was shown. The R5d band polarizations are essential to stabilize the magnetic order in rare-earth-transition metal compounds. The Y4d band polarizations induced by 4f–3d short-range exchange interaction showed the same behavior as the  $M_{5d}(0)$  values.

The study of the magnetic properties of  $Pr_xNd_{10-x}Fe_{75}Co_{9-y}Si_yB_6$  nanocomposite alloys suggested that the magnetic moments of transition metals (Fe,Co) are little affected by alloying. An increase of the coercive field was shown by adding silicon.

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