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Magnetic properties of RCo5-based systems

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ABSTRACT

Band structure calculations and magnetic measurements were performed on $(Gd_xY_{1-x})Co_4Si$ as well as on YCo_4M compounds, where M = Ga, Al, and Co. The substitutions both at rare-earth and cobalt sites influence strongly the cobalt moments as well as the Curie temperatures. When Y is replaced by Gd, in $Gd_xY_{1-x}Co_4Si$ system, the mean cobalt moments increase from $0.73\mu_B$/atom to $1.25\mu_B$/atom. The computed magnetic moments per formula unit agree rather well with experimental data. The reciprocal susceptibilities for ferrimagnetic compounds show non-linear temperature dependences. The effective cobalt moments are little dependent on composition. The ratio r between the mean number of cobalt spins determined from effective and saturation cobalt moments, respectively follows a dependence on Curie temperature of the form $r \propto T_c^{-2/3}$. The magnetic behaviour of cobalt is analysed in spin fluctuations model.

1. Introduction

The RCo_5 intermetallic compounds, where R is a rare-earth or yttrium, have been intensively investigated in connection both with their technical applications as well as for a better understanding of physical properties, particularly cobalt magnetism.

The RCo_5 compounds crystallize in a $CaCu_5$-type structure. In this lattice the cobalt atoms occupy 3g and 2c sites, while the R atoms are located in 1a-type site [1,2]. These compounds have generally high Curie temperatures and in case of light rare-earths relative high saturation magnetizations, since parallel alignment of R and Co moments. The RCo_5 compounds with heavy rare-earths are ferrimagnetically ordered. Some RCo_5 compounds are known for their large magnetocrystalline anisotropy. The interesting magnetic properties arise from the presence in the same compound of both 3d-itinerant and 4f-localized magnetism of Co and R elements, respectively. The substitutions of a magnetic rare-earth by a nonmagnetic one or yttrium and cobalt by nonmagnetic elements, is a way in which information on the cobalt magnetism can be obtained [1,2].

We showed previously [3], that there is a correlation between the variation of exchange fields acting on cobalt and their moments, determined at 4.2 K. The same behaviour was shown by high field measurements in the case of $Gd_xLa_{1-x}Co_4B$ system [4]. An induced cobalt moment of $[2.5–3.0 \times 10^6]^{-1}\,\mu_B$/Oe was reported [3,4]. The effective cobalt moments determined in the paramagnetic range seem to be little dependent on composition [3,5]. Substitutions at cobalt sites, in $RCo_{5-x}A_x$, compounds, preserving the $CaCu_5$-type structure, were also performed for A = Si [5–8], Al [9–12], Ga [13,14] or Ge [15]. These substitutions influence significantly the cobalt moments, and can be correlated with site occupancy. Experimentally it has been shown that the Al and Ge atoms substitute Co atoms only in 3g sites, while Ga and Si are distributed preferentially on 3g sites.

The exchange interactions in RCo_5-based compounds were little studied. Now, it is generally accepted that in rare-earth (R)–transition-metal (T), compounds, the exchange interactions between magnetic rare-earths and T atoms can be described by a 4f–5d–3d model, as suggested by Campbell [16]. The exchange interactions between transition metal elements are of short range, while those between rare-earths take place both by means of conduction electrons or by direct 5d–5d-type ones [17].

An important matter is that played by the R5d band polarization in the exchange interactions. We already showed [17–19] that the R5d polarizations in RT_2 and RT_5-type compounds, are determined both by local 4f–5d exchange, $M_{5d}(f)$, as well as by 5d–3d hybridization effects, due to the short range exchange interactions, $M_{5d}(d)$. The $M_{5d}(d)$ values are dependent on the number of magnetic T atoms situated in the first coordination shell, to an R one, as well as on their moments. Later on Laguna-Marco [20], by using X-ray magnetic circular dichroism, confirmed the above behaviour. A general discussion on this matter was recently published [21].

In order to obtain more information on the magnetic behaviour of cobalt, we studied $Gd_xY_{1-x}Co_4Si$ system in a large temperature

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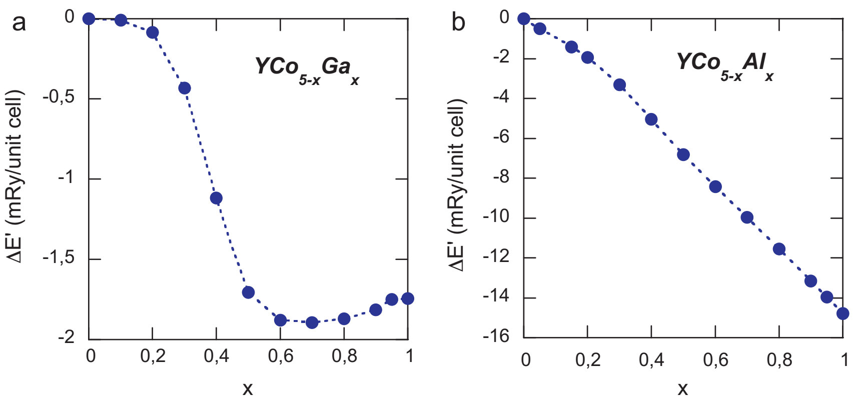

Fig. 1. Total energy difference when cobalt is located in 3g and 2c sites, respectively for YCo_4Ga (a) and YCo_4Al (b).

range, including paramagnetic region. In addition, band structure calculations were made on the above system as well as on YCo_4A compounds with A = Al, Si, Co and Ga.

2. Experimental and computing method

The $Gd_xY_{1-x}Co_4Si$ compounds were prepared by arc melting the constituent elements in purified argon atmosphere. The samples were remelted several times to ensure a good homogeneity. An excess of 4% gadolinium (yttrium) was used in order to compensate their loss during melting. In this way the formation of phases with higher Curie temperatures, than of the basic compounds, can be avoided. This requirement is important in order to obtain reliable magnetic data in paramagnetic range. The samples were thermally treated, for one week, at 1000 °C.

The X-ray analyses show the presence of only one phase having hexagonal structure of P6/mmm space group. Since the radius of Gd^{3+} (0.96 Å) and Y^{3+} (0.93 Å) are close, the lattice parameters are only slightly changed as results of substitutions. The a parameters in $Gd_xY_{1-x}Co_4Si$ system increase as function of Gd content from 4.937 Å ($x = 0$) to 4.985 Å ($x = 1$) and c lattice constants from 3.912 Å ($x = 0$) to 3.928 Å ($x = 1$). In a rigid spheres model the a parameters are mainly determined by R–T contacts and c ones by T–T contacts. The larger variation in a values (0.96%) than of c ones (0.10%) can thus be explained.

Magnetic measurements were performed in the temperature range 4.2–950 K and fields up to 90 kG. In the paramagnetic region, the susceptibilities, χ, have been determined from magnetization isotherms, according to Honda–Arrot plot [22]: $\chi_t = \chi + dM_s'H^{-1}$, by extrapolating the measured values, χ_t, to $H^{-1} \to 0$. d is denoted as presumed impurity content and M_s' is their saturation magnetization. By this method any possible alteration of the χ values, as result of the presence of small quantities of magnetic ordered phase is avoided. It is also to be mentioned that in case of $x = 0.6$ sample, around ≅2% impurity phase, with $T_c \cong 500$ K, allowed measurements only at $T > 600$ K. For other samples the content of magnetic impurity phases, present in paramagnetic range, estimated from χ versus H^{-1} plots, were very low (<0.2%).

The ground state electronic structure and magnetic properties of $Gd_xY_{1-x}Co_4Si$ and YCo_4M (M = Al, Ga, Si) compounds have been studied by tight-binding linear muffin-tin orbital method (TB-LMTO) within the atomic sphere approximation (ASA), together with the coherent potential approximation (CPA) [23]. The local spin density approximation (LSDA) was used for the exchange-correlation potential within Vosko–Vilk–Nusair parameterization [24]. All calculations have been performed using a mesh of 16 × 16 × 16 k-points in the full Brillouin zone (BZ), resulting in 274 k-points in the irreducible wedge of BZ.

The LSDA+U approximation was also used to compute the band structures of $Gd_xY_{1-x}Co_4Si$ alloys with $x = 0$, 0.5 and 1.0, where an intra-atomic Coulomb interaction U was added [25]. For Gd, the average Coulomb interaction $U_f = 9$ eV and exchange interaction $J_f = 1$ eV were chosen [21,25]. For cobalt, values $U_d = 2$ eV and $J_d = 1$ eV, which are widely accepted [21,26], have been used.

3. Band structures

The energy calculations were performed on $YCo_{5-x}A_x$ (A = Ga, Si, Al) in order to analyse the preferred substitutions on cobalt sites. The energy difference, when A elements are located in 3g and 2c sites, respectively, was analysed, as shown, as example, in Fig. 1 for $YCo_{5-x}A_x$ with A = Ga and Al. The minimum energy difference, $\Delta E'$, in $YCo_{5-x}Ga_x$ system corresponds to a substitution of $x = 0.75$ Ga atoms in 3g site. This result is in agreement with neutron studies, where 75% of Ga atoms were shown to occupy the 3g site and only 25% to be located on 2c site [14]. Similar result was obtained for Si substitution. The energy considerations showed that Al can be located exclusively in 3g sites. As a consequence, in analysing the band structures of YCo_4A (A = Al, Si, Ga), and $(Gd_xY_{1-x})Co_4Si$ compounds, the substituting elements, were located only on 3g sites.

The total densities of states for YCo_4A (A = Ga, Si and Al) compounds are shown in Fig. 2. The Fermi level is situated in a peak of density of states, for Co spin up 3d subband, both for 3g and 2c sites. The corresponding density of states for the spin down subband is low. The band structures of YCo_4A systems resemble to that of YCo_5 compound.

The band structure of $GdCo_4Si$, computed by using LDA+U method is shown in Fig. 3. The computed magnetic moments for Co3g and Co2c atoms are $1.27\mu_B$ and $1.23\mu_B$, respectively. There are only little different from those obtained by LMTO-ASA procedure, where values of $1.19\mu_B$ and $1.15\mu_B$, respectively were determined. For intermediate compositions, in $(Gd_xY_{1-x})Co_4Si$ system, band structures were computed considering supercells. We showed already [27,28], that the consideration of the supercell do not change the computed magnetic moments, as compared to those determined for the basic unit cell.

The energy location of the center of gravity for the spin up and spin down of the DOS for 3g and 2c sites in YCo_5, YCo_4Si, YCo_4Ga, YCo_4Al and $GdCo_4Si$ were analysed. As compared to YCo_5, the center of gravity of the Co3d spin down subbands, in case of the substituted compounds, are shifted to higher energies, and as a result the exchange splitting is diminished. The induced Y4d band polarizations, M_{4d}, in YCo_4M (M = Co, Al, Ga, Si) compounds are oriented antiparallely to cobalt moments and decrease as the cobalt moments diminishes, suggesting a correlation between these values. The Gd5d band polarization, M_{5d}, in $GdCo_4Si$ is rather high, of $0.47\mu_B$ and parallely oriented to gadolinium 4f moments. This includes contributions both from 4f–5d local exchange and 5d–3d short range exchange interactions [21]. To estimate the above contributions, band structure calculations were performed on RCo_4Si heavy rare-earth compounds. As in case of RCo_5 compounds [21], linear dependence of M_{5d} values as function of De Gennes factor, $G = (g_J - 1)^2J(J + 1)$, was shown. The determined slope, of $\beta = 0.8 \times 10^{-2}\mu_B/G$, is somewhat smaller than that evidenced in RCo_5 compounds, namely $1.4 \times 10^{-2}\mu_B/G$. [21]. The contribution to R5d band polarization, induced by R5d–Co3d hybridization was obtained by extrapolation of computed values to

$G = 0$. A value of $\cong 0.27\mu_B$ was obtained. The contribution of 4f–5d local exchange to Gd5d band polarization in $GdCo_4Si$ thus results to be the same as in $GdCo_5$ compound, of $\cong 0.20\mu_B$ [21].

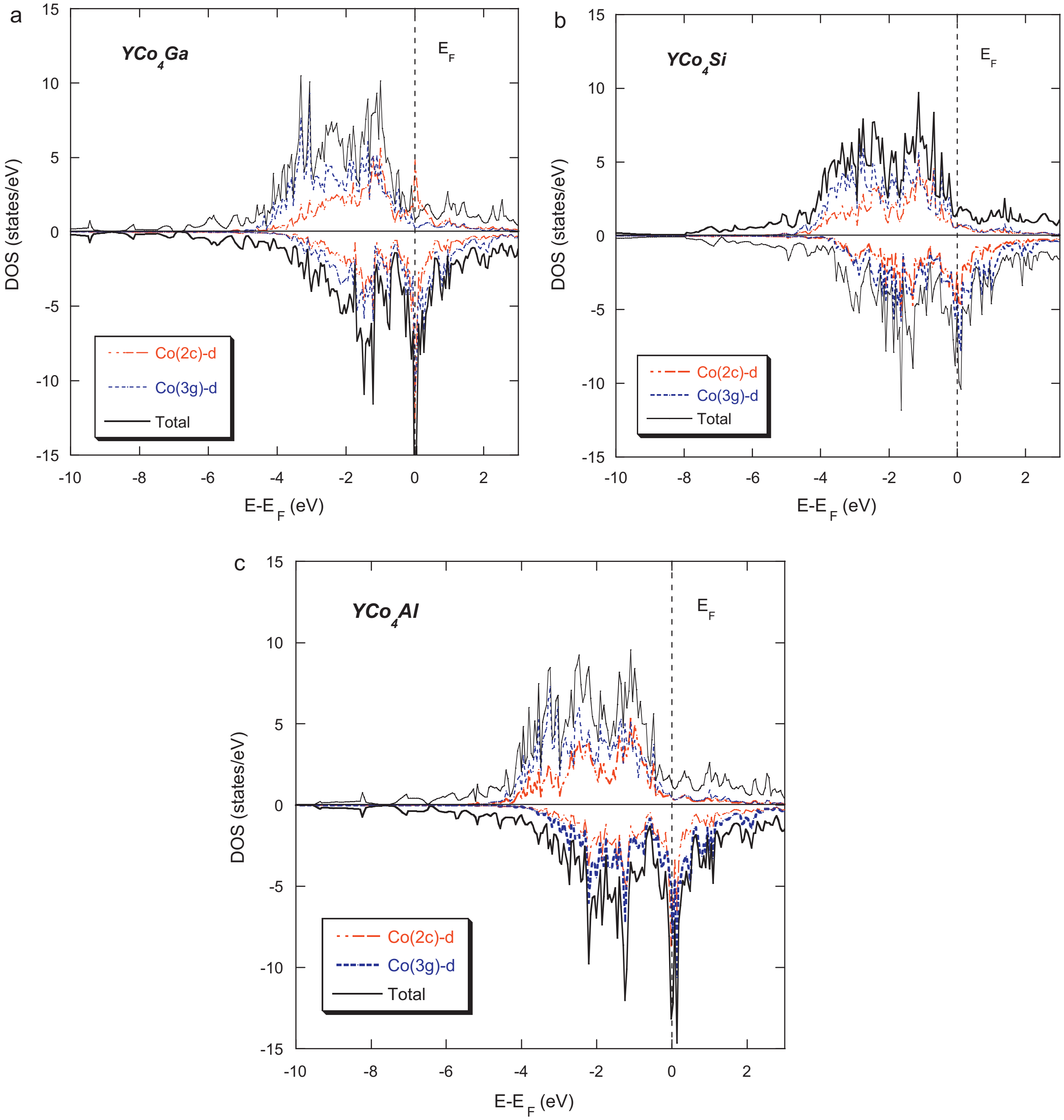

Fig. 2. The total and partial densities of states for YCo_5Ga (a), YCo_4Si (b), and YCo_4Al (c) compounds.

The magnetic moments of cobalt atoms at 2c and 3g sites in YCo_4A ($A = Co$, Al, Ga, Si) and in $GdCo_4Si$ compounds are plotted in Fig. 4 as a function of exchange splitting, ΔE, of the subbands. A linear dependence is shown suggesting that these values are strongly correlated. The determined slope, $s = 0.07$ Ry/μ_B (or 0.95 eV/μ_B), is in good agreement with that obtained from band structure calculations on Zr–T (T = Fe, Co, Ni) amorphous alloys and crystalline compounds [29]. The s value is not influenced by the valence state of transition metal partner (Zr^{4+} or Y^{3+}) or by the crystallinity degree of the samples, confirming the previous data [29].

4. Magnetic properties of $Gd_xY_{1-x}Co_4Si$ compounds

The thermal variations of spontaneous magnetizations in $Gd_xY_{1-x}Co_4Si$ compounds are shown in Fig. 5. The YCo_4Si is a ferromagnet. For the compounds with $x \geq 0.2$, the magnetizations, as function of temperatures, follow typical curves for ferrimagnetic ordering. According to Néel classification [30], these are of P-type for $x = 0.2$ and 0.4 and of N-type for compositions $x \geq 0.6$.

In analysing the magnetic behaviour of ferrimagnetic compounds, we considered a two sublattices molecular field model, corresponding to Gd and Co atoms, whose magnetizations are antiparallely oriented. Since generally, the magnetic moments at 2c and 3g sites are different, in the following we refer only to a mean magnetic moment of cobalt atoms. The magnetizations of cobalt and gadolinium sublattices, at 4.2 K, compensate at $x = 0.57$ (Fig. 6). For smaller Gd content than the above value, the resultant magnetization is oriented parallely to cobalt moments, while for higher ones, this is parallel to Gd sublattice magnetizations. The computed magnetic moments per formula unit agree rather well with exper-

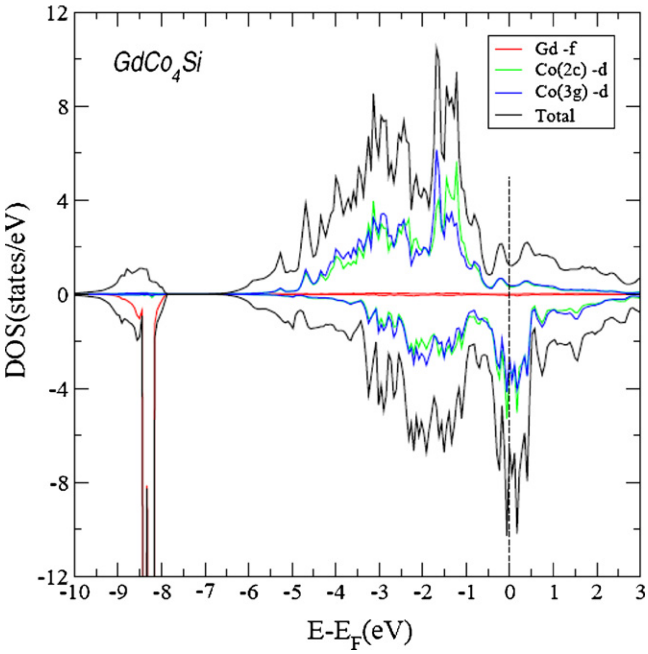

Fig. 3. The band structure of $GdCo_4Si$ compound, computed by using LDA+U method ($U_f = 9$ eV, $J_f = 1$ eV, $U_d = 2$ eV, $J_d = 1$ eV).

imental data (Fig. 6). Although there are not significant differences between the computed values when using LDA+U and LMTO-ASA methods, respectively, those obtained by LDA+U are more close to the experimental ones.

The Curie temperatures, T_c, increase from $T_c = 332$ K ($x = 0$), up to ≅450 K in $GdCo_4Si$, reflecting the increase of the exchange interactions as the non-magnetic Y is replaced by Gd (Fig. 6).

Assuming that the magnetic moment of Gd is $7\mu_B$, the mean cobalt moments were determined (Fig. 7). These show a rather high increase as the Gd content is higher, from $0.73\mu_B$ ($x = 0$) to $1.25\mu_B$/atom ($x = 1$).

The thermal variations of reciprocal susceptibility, for YCo_4Si, follows a Curie–Weiss type behaviour (Fig. 8). From Curie constant we determined a mean effective cobalt moment of $3.13\mu_B$/atom. For ferrimagnetic compounds, non-linear temperature dependences are shown, which can be described by a Néel relation [30]:

$$\chi^{-1} = \chi_0^{-1} + TC^{-1} - \sigma(T - \theta)^{-1} \qquad (1)$$

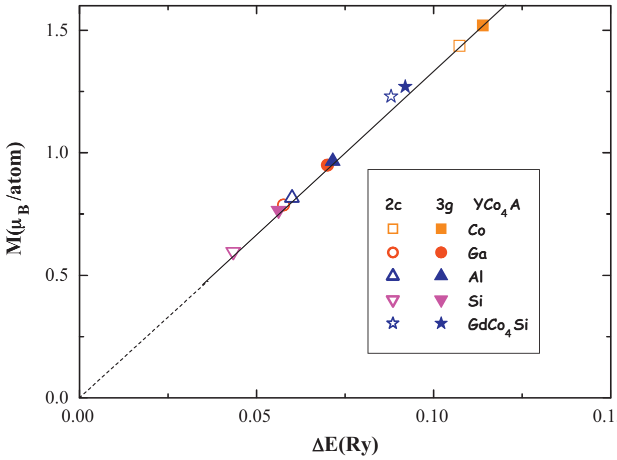

Fig. 4. The cobalt moments at 2c and 3g sites in YCo_4A (A = Al, Si, Ga, Co) and $GdCo_4Si$ compounds as a function of exchange splitting of their 3d bands.

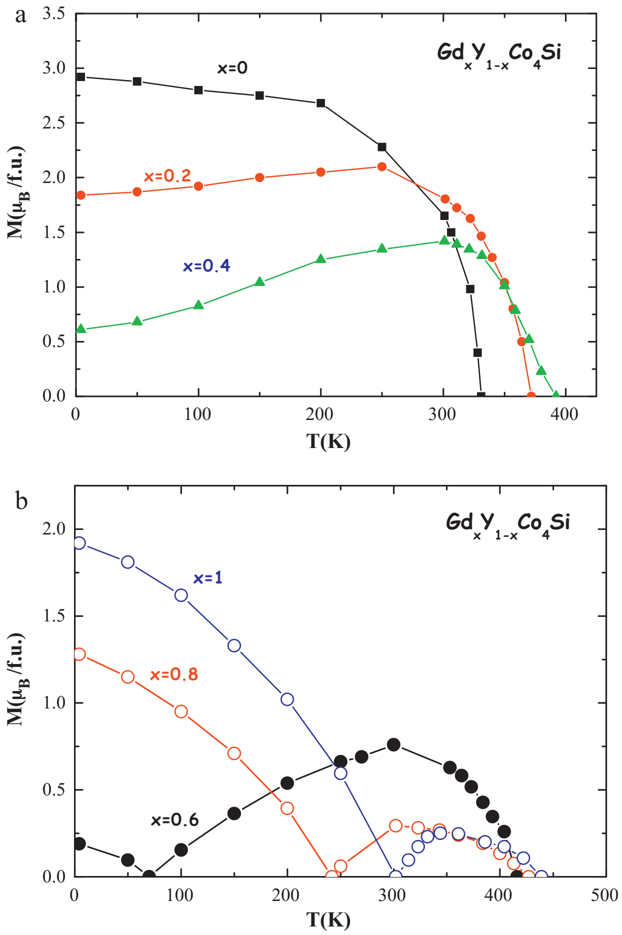

Fig. 5. Thermal variations of spontaneous magnetizations in $Gd_xY_{1-x}Co_xSi$ compounds.

where χ_0, σ and θ are denoted parameters related to molecular field coefficients, describing the interactions inside and between magnetic sublattices and C is the Curie constant.

At temperatures $T > 700$ K, linear dependences are shown and accordingly C values were obtained. The contributions of cobalt atoms to the Curie constants, C_{Co}, were determined according to addition law of magnetic susceptibilities. We considered that the effective gadolinium moment is given by that of free ion value, as evidenced previously [31]. The effective cobalt moments are only little dependent on composition (Fig. 7). Taking into account the

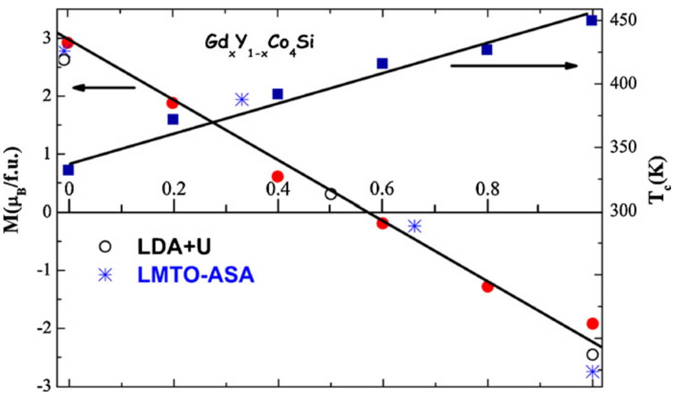

Fig. 6. Composition dependences of the magnetization at 4.2 K and of Curie temperatures, T_c in $Gd_xY_{1-x}Co_4Si$ compounds. The computed mean magnetizations by using LDA+U and LMTO-ASA methods are also plotted.

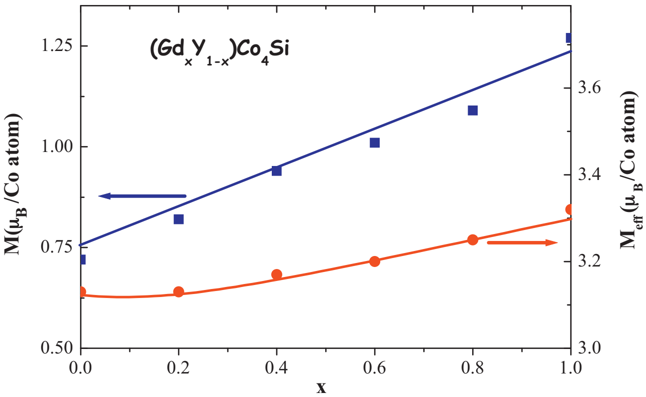

Fig. 7. Composition dependences of the mean cobalt moments at 4.2 K and of mean effective moments.

possible experimental errors, these values can be situated in the range $M_{\mathrm{eff}}(\mathrm{Co}) = 3.23 \pm 0.10\mu_B$/atom. The same type behaviour was shown in R–Co–B series [32].

The ratio $r = S_p/S_o$, between the number of the spins determined from effective cobalt moments, S_p, $\mu_{\mathrm{eff}}(\mathrm{Co}) = g\sqrt{S_p(S_p+1)}$ and from saturation data, $M_{\mathrm{Co}} = gS_o$, were determined. The r values increase from 1.97 ($x = 1$) to $r = 3.16$ ($x = 0$).

5. Discussion

The idea of Rhodes–Wohlfarth plot [33], where the ratio between the number of spins determined from Curie constants and saturation magnetizations versus Curie temperatures, T_c, is given, should be to distinguish between metallic systems obeying a localized model, from those showing an itinerant magnetic behaviour. According to Wohlfarth [34], the application of itinerant electron model of ferromagnetism gives the ratio $r = S_p/S_o \cdot \alpha \cdot T_c^{-1}$. This relation predicts a hyperbolic increase of r values as the Curie temperatures diminishes. Moriya [35] developed a spin fluctuations model of magnetism which takes into account both band and localized features. In the local moment limit there is $r = 1$, while in the opposite limit, the band model predicts a divergence of this ratio as the spin $S_o \to 0$ and $T_c = 0$. Generally, in metallic systems, there is a large range of r values, showing that the nature of spin fluctuations can changes continuously between the opposite extremes. The non-local nature of spin fluctuations in itinerant magnets is generally far more important than in magnetic insulators. When the spin fluctuations model [35] is considered, the mathematical development approach, gives $r \propto T_c^{-2/3}$. In this case, the approximate hyperbolic form, suggested by Rhodes–Wohlfarth, is distorted. Such a dependence has been shown previously in $\mathrm{Ni}_{100-x}\mathrm{Y}_x$ amorphous alloys with $3 \le x \le 17$ [36]. The r values as function of T_c^{-1} and $T_c^{-2/3}$ are plotted in Fig. 9. Linear variations can be seen in both cases. The dispersions of the data are greater when T_c^{-1} model is considered, suggesting that the presence of spin fluctuations behaviour is most probable.

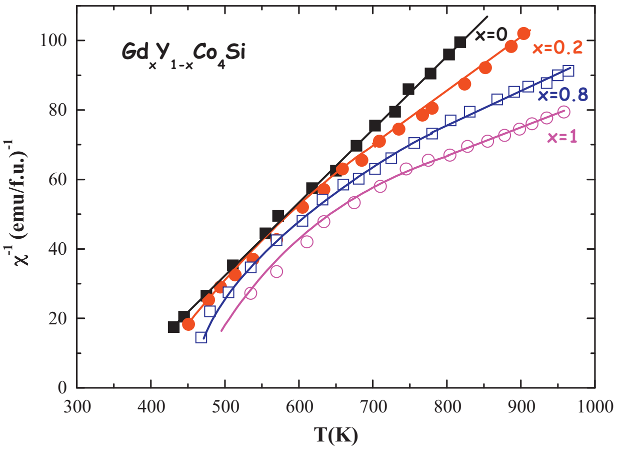

Fig. 8. Thermal variations of reciprocal susceptibilities for $\mathrm{Gd}_x\mathrm{Y}_{1-x}\mathrm{Co}_4\mathrm{Si}$ compounds.

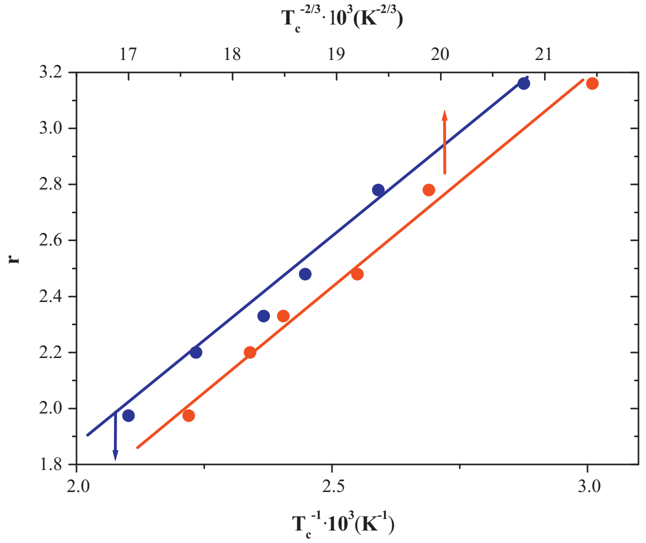

Fig. 9. The r values determined in $\mathrm{Gd}_x\mathrm{Y}_{1-x}\mathrm{Co}_4\mathrm{Si}$ compounds, plotted as function of T_c^{-1} and $T_c^{-2/3}$.

In order to obtain more information on the behaviour of cobalt moments in $\mathrm{Gd}_x\mathrm{Y}_{1-x}\mathrm{Co}_4\mathrm{Si}$ system we computed the exchange field, H_{exch}, acting on cobalt. A nearly linear variation is shown between cobalt moments and exchange fields, as previously reported in some Gd–Co–B compounds [33], with a slope of $(2.5 \times 10^6)^{-1}\mu_B$/Oe (Fig. 10). This shows the importance of exchange field in splitting the cobalt subbands, the cobalt moments being linearly correlated both on exchange field and exchange splitting of 3d bands, respectively. Also, the previous reported value, $s = 0.95\ \mathrm{eV}/\mu_B$ [29], describing the correlation between the local cobalt moments and the exchange splittings of their 3d states was confirmed.

The above data suggest that the cobalt moments in $\mathrm{Gd}_x\mathrm{Y}_{1-x}\mathrm{Co}_4\mathrm{Si}$ compounds can be better described in spin fluctuations model [35]. In this model the Curie constants are

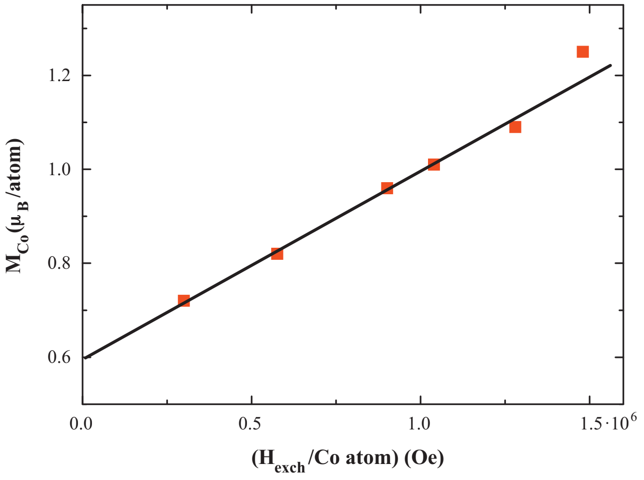

Fig. 10. The dependences of the cobalt moments at 4.2 K on exchange fields.

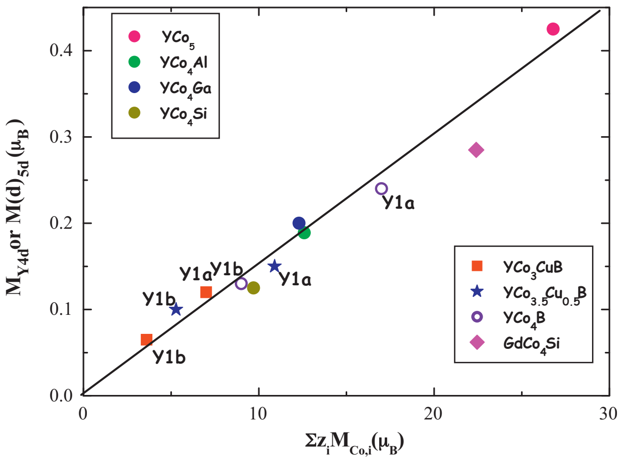

Fig. 11. The M_{5d}(d) in GdCo$_4$Si and Y4d band polarizations in YCo$_4$A (A = Co, Al, Ga, Si) compounds as function of the number of Co atoms situated in first coordination shell to an Y or Gd one, as well as on their moments. The data on YCo$_{4-x}$Cu$_x$B with x = 0, 0.5 and 1.0 are also included in figure [37,38].

independent on the saturation moment, at T = 0, but depends on the band structure around the Fermi energy. Therefore, the Curie constant is not modified when the effective exchange constants vary, keeping the close related band structures. The exchange interactions however influence the transition metal moments at 4.2 K and Curie temperatures. This behaviour is really evidenced in Gd$_x$Y$_{1-x}$Co$_4$Si system. The forms of Co partial densities of states in the above system, are rather similar although the magnetic moments and T_c values cover a rather large range of values.

The R–Co exchange interactions in Gd$_x$Y$_{1-x}$Co$_4$Si ($x \geq 0.2$) samples take place through R5d band polarizations. As in case of RT$_2$ (T = Fe, Co, Ni) and RT$_5$ (T = Co, Ni) compounds [18,21], two contributions to R5d band polarizations are shown, which in first approximation are additive. The 4f–5d local interactions induce a contribution to polarization which is linearly dependent on De Gennes factor, while M_{5d}(d) one seems to depend on the cobalt moments of neighbouring atoms to an R or Y one. We showed already [17,21] that the induced polarization by short-range T3d–R5d (Y4d) exchange interactions, can be described by the relation:

$$M_{5d}(d) \propto \Sigma_i z_i M_{Co,i} \qquad (2)$$

where z_i is the number of cobalt atoms situated in i positions and the $M_{Co,i}$ are their magnetic moments.

Such a behaviour can be shown in the studied systems as seen in Fig. 11. The induced Y4d band polarizations determined in YCo$_4$A (A = Al, Si, GaCo) as well as in YCo$_{4-x}$Cu$_x$B system [37] follow the same trend as that of M_{5d}(d) one determined in GdCo$_4$Si, as can be seen in Fig. 11.

We conclude that the cobalt moments in Gd$_x$Y$_{1-x}$Co$_4$Si and in YCo$_4$A (A = Si, Ga, Al, Co) compounds where R is a heavy rare-earth or yttrium can be better analysed in the spin fluctuations model. The R–Co exchange interactions can be described by 4f–5d–3d model. The R5d band polarizations are due both to local 4f–5d and 3d–5d short range interactions. A linear relation between Y4d and Gd5d(d) band polarizations and the total magnetizations of the cobalt atoms situated in the first coordination shell to an Y(Gd) one is shown, in agreement with the presence of short range 5d(4d)–3d interactions and the resulting hybridization effects.

Acknowledgments

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