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Electrical resistivity and Mössbauer effect studies of $Y(Fe_{1-x}Co_x)_2$ intermetallics

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ABSTRACT

Synthesis, X-ray analysis (300 K), electrical resistivity and Mössbauer effect studies in the intermetallic compounds $Y(Fe_{1-x}Co_x)_2$ are reported. Cubic Laves phases $MgCu_2$-type, were observed across the series. Crystal lattice parameter is considerably reduced with Co concentration. The lattice parameters of the $Y(Fe_{1-x}Co_x)_2$ series are compared to analogous data previously observed for the $Gd(Fe_{1-x}Co_x)_2$ and $Dy(Fe_{1-x}Co_x)_2$ series. Electrical resistivity for the $Y(Fe_{1-x}Co_x)_2$ intermetallic series was measured in the temperature range 15–1000 K. Parameters characterizing the resistivity dependence on temperature including Debye temperature were determined. Residual, phonon and magnetic resistivity were separated from the measured electrical resistivity. Curie temperatures of the $Y(Fe_{1-x}Co_x)_2$ series were determined using both electrical resistivity and Mössbauer effect measurements. The Curie temperature increases with x, reaches maximum approximately at $x = 0.3$ and than strongly decreases as a result of Fe/Co substitution. The obtained Curie temperatures for the $Y(Fe_{1-x}Co_x)_2$ series are related to the data known for the $Dy(Fe_{1-x}Co_x)_2$, $Gd(Fe_{1-x}Co_x)_2$ and other series. Numerical formula, which approximates Curie temperatures of intermetallics is proposed.

1. Introduction

The intermetallic compounds $R(Fe_{1-x}Co_x)_2$ (R – heavy rare earth) with the C15 cubic Laves phase structure are intensely studied for their fundamental interest and their practical applications [1–11]. From Mössbauer effect studies it was found, that a magnetic hyperfine field observed at ^{57}Fe nuclei as a function of composition for the $R(Fe_{1-x}Co_x)_2$ (R = Y, Gd, Dy, Ho) intermetallic series, behaves similarly to the Slater–Pauling curve, in analogy to the 3d metal–3d metal alloys [2,3,12–14] with a maximum field for the $R(Fe_{0.7}Co_{0.3})_2$ compound. Usually substitution of one transition metal/another transition metal in the RM_2 system (M – transition metal) is adopted as a driving force to change across the series the number n of 3d electrons (n calculated per transition metal atom) in the transition metal sublattice and thus to change band properties and related to them magnetic and hyperfine interaction properties [13,14].

For instance, a magnetic ordering temperature observed across the exemplary $R(Fe_{1-x}Co_x)_2$ (R = Dy, Gd) series can be treated as a certain replica of the magnetic hyperfine field [15–17]. Namely, the Curie temperature in these series increases with x and a maximum Curie temperature is approached in the area of the $R(Fe_{0.7}Co_{0.3})_2$ compound. Further Fe/Co substitution strongly reduces the Curie temperature. In contrast to magnetic properties, electrical properties of the $R(Fe–Co)_2$ compounds are known only fragmentary and an origin of these properties and their relations to the magnetism of the $R(Fe–Co)_2$ compounds seems to be a open topic up to date.

Yttrium with a lack of the well-localized electron magnetic shell is often used in intermetallics as a substitute instead of rare earths [1]. An influence of yttrium on transition metal sublattice properties is less as compared to the heavy lanthanides. Thus it has been an interesting topic to test the series $Y(Fe_{1-x}Co_x)_2$ with the Y^{3+} ions as constituents which are characterized by relatively simple quantum state. Crystal structure, magnetic properties, ^{57}Fe Mössbauer effect and NMR considering local environment effects were studied for

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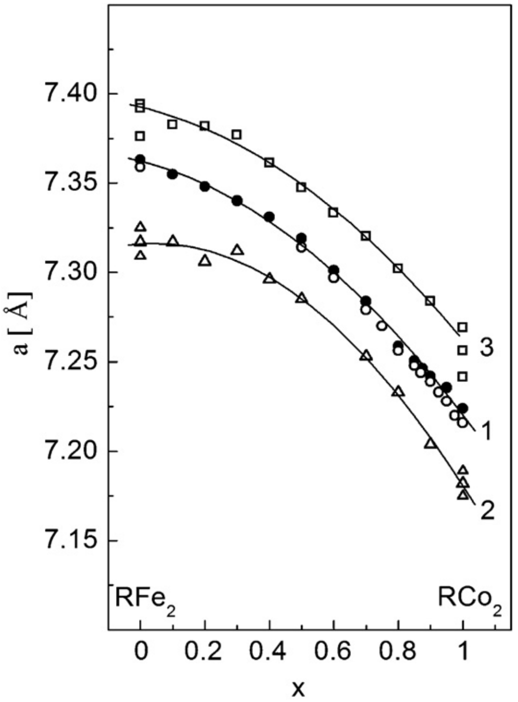

Fig. 1. The unit cell edge *a* (curve 1) observed for the Y($Fe_{1-x}Co_x$)$_2$ intermetallics (300 K). The literature data (open points) are added [1,25]. Additionally are presented data for Dy($Fe_{1-x}Co_x$)$_2$ (curve 2) [1,17,26] and Gd($Fe_{1-x}Co_x$)$_2$ (curve 3) [1,15,16].

compounds of the Y($Fe_{1-x}Co_x$)$_2$ series, especially for the borderline compounds YFe_2 and YCo_2 [18–20]. For purpose of the presented studies series Y($Fe_{1-x}Co_x$)$_2$ was prepared, X-ray, electrical measurements and high temperature Mössbauer effect measurements were performed.

2. Materials and structures

The polycrystalline compounds Y($Fe_{1-x}Co_x$)$_2$ (x = 0, 0.1, . . ., 0.85, 0.9, 0.95, and 1) were prepared by arc melting in a high purity argon atmosphere using appropriate amounts of Y(99.95% purity), Fe and Co (all 99.99% purity) as starting materials. As a next step, the synthesized ingots were annealed in a vacuum (10^{-6} Pa) at 1100 K for 10 h and then allowed to cool down along with the furnace at; approximately 250 K/h.

Crystal structures were tested with standard X-ray powder diffraction procedure using CuK_α radiation. The good quality X-ray diffractograms obtained for these materials were analyzed using the Rietveld-type method adopting both the $K_{\alpha1}$ (wave length λ_1 = 1.540560 Å) and $K_{\alpha2}$ (λ_2 = 1.544390 Å) lines [21,22]. The clean cubic, *Fd*3*m*, $MgCu_2$-type (C15) crystal Laves phases were observed for all prepared compounds of the studied series. Since additional X-ray reflexes caused by a long-range order have not been observed, thus a random distribution of Fe and Co atoms in the transition metal sublattice can be expected to exist. The C15 Laves phase has been previously described in details elsewhere [23].

As a consequence that, the atomic radius of Fe is higher than the corresponding radius of Co (r_{Fe} = 1.72 Å, r_{Co} = 1.67 Å) [24] the unit cell parameter *a* is reduced with the cobalt content *x* (Fig. 1). A numerical formula $a(x) = (-0.106x^2 - 0.036x + 7.360)$ Åthe result of a least squares fitting, describes properly experimental points (line 1 in Fig. 1). The existing literature data presented for a comparison [1,25] coincide well with the determined $a(x)$ dependence. A similar convex deviation $a(x)$ from Vegard's rule was previously observed for R($Fe_{1-x}Co_x$)$_2$ intermetallics, where R = Dy (Fig. 1, curve 2) [1,17,26] and R = Gd (Fig. 1, curve 3) [1,15,16].

3. Electrical resistivity

Bar (cuboid) shaped specimens for the electrical measurements with typical dimensions 1mm × 1mm × 15mm, were precisely cut from the ingots using precise diamond wheel saw. Electrical contacts to the bars were connected by point spark-welding of high purity thin copper wires onto the ends of the bars. Only microscopically observed cracks free samples were used for the four probe dc electrical measurements.

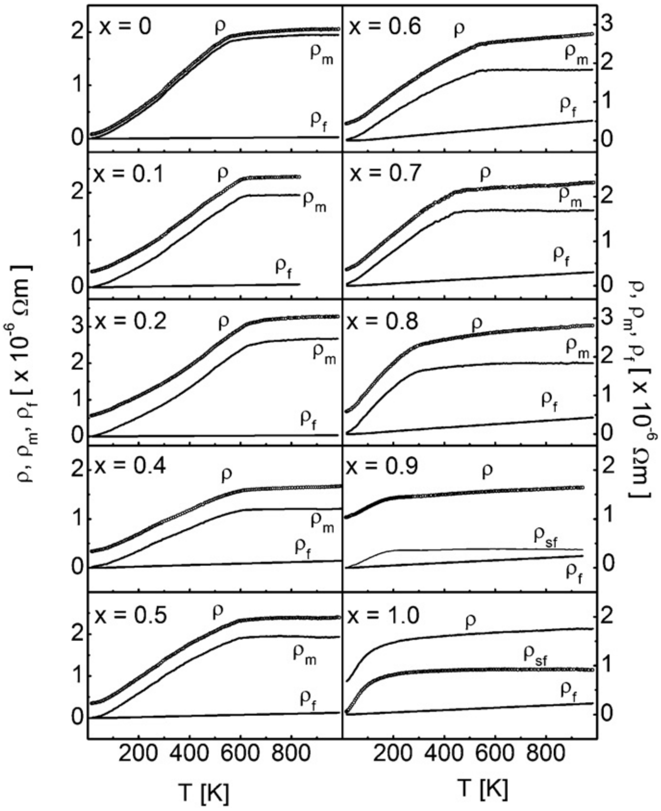

Fig. 2. Electrical resistivities: the total ρ, the phonon ρ_f and the magnetic ρ_m or ρ_{sf} observed against temperature for the Y($Fe_{1-x}Co_x$)$_2$ compounds.

The obtained good quality electrical resistivities ρ as functions of temperature *T* for the intermetallic series Y($Fe_{1-x}Co_x$)$_2$ are presented in Fig. 2.

The resistivity observed for magnetic materials can be described by the Matthiesen formula [27,28]:

$$\rho = \rho_0 + \rho_f + \rho_m \tag{1}$$

where ρ_0 is the residual resistivity, ρ_f is the phonon scattering resistivity described by the Bloch–Grüneisen formula [17,29] and ρ_m is the magnetic contribution to the total resistivity. The particular resistivity contributions present in the Matthiesen formula, have been discussed detailedly elsewhere [17]. A numerical treatment to separate ρ_0, ρ_f, ρ_m and to calculate parameters characterizing these component electrical resistivities has also been previously described elsewhere [15,17,26].

It should be mentioned that in the case of exchange-enhanced paramagnet a spin fluctuation ρ_{sf} contribution instead of the ρ_m contribution do exist [30].

As discussed previously, in the case of R–M intermetallics expression (1) for temperatures $T \ll \theta_D$ and $T \ll T_C$ (θ_D is the Debye temperature, T_C is the Curie temperature) can be rewritten in the following form [17,27]

$$\rho(T) = 497.6D\left(\frac{T}{\theta_D}\right)^5 + AT^2 + BT + \rho_0 \tag{2}$$

where *A*, *B* and *D* are parameters.

In the case of the high temperature range ($T \gg T_C$) the total resistivity formula can be approximated as [27,28]

$$\rho(T) = D\left(\frac{T}{\theta_D}\right) + C \tag{3}$$

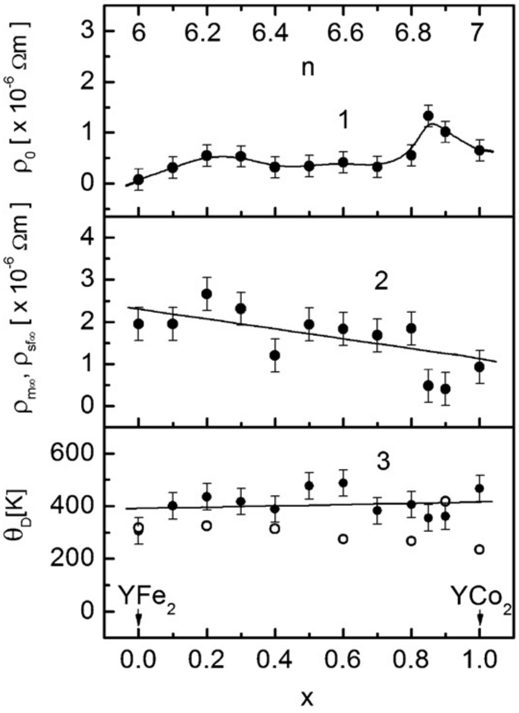

Fig. 3. The residual resistivity ρ_0 (curve 1), the asymptotic magnetic resistivity $\rho_{m\infty}$ or $\rho_{sf\infty}$ (curve 2) and the Debye temperature θ_D (curve 3) as functions of the composition parameter x for the $Y(Fe_{1-x}Co_x)_2$ compounds. The literature Debye temperatures θ_D (open points) are added [31].

where C is a parameter.

An analogous method of calculations following formulae (1)–(3) as described elsewhere [15,17] was applied to separate particular contributions to the total electrical resistivity.

Results of the resistivity measurements and of the performed calculations are presented in Figs. 2 and 3 and in Table 1.

The determined residual resistivity $\rho_0(x)$ for the $Y(Fe_{1-x}Co_x)_2$ series is presented in Fig. 3, curve 1. The $\rho_0(x)$ dependence has two not too high maxima approximately at $x = 0.25$ and $x = 0.85$ and flat minimum in the area $x = 0.4–0.7$. To some extent similar dependence of $\rho_0(x)$ for the $Dy(Fe_{1-x}Co_x)_2$ series related to statistical disorder among the Fe, Co atoms has been observed and discussed previously [17].

The high temperature asymptotic values $\rho_{m\infty} = \rho_m(T \to +\infty)$ of magnetic resistivity for the $Y(Fe_{1-x}Co_x)_2$ series are presented in Fig. 3, line 2. The $\rho_{m\infty}$ resistivity decreases with x and can be approximated by the numerical formula $\rho_{m\infty}(x) = (-1.169x + 2.304) \times 10^{-6}$ Ωm. It is worth noticing, that what is known as the high temperature magnetic contribution to resistivity in 3d-metals, which is a counterpart of $\rho_{m\infty}$, approximately equals: 0.8×10^{-6} Ωm for Fe metal and 0.5×10^{-6} Ωm for Co metal [28]. The value of Fe metal resistivity is higher compared to the Co metal value. Therefore it can be expected that the tendency of $\rho_{m\infty}(x)$ to decrease, which is a result of Fe/Co substitution in the $Y(Fe_{1-x}Co_x)_2$ series, has an origin analogous to the case of pure metals [28]. Similar dependence $\rho_{m\infty}$ has previously been observed for the $Dy(Fe_{1-x}Co_x)_2$ series [17]. In the case of spin fluctuations, compounds with $x = 0.9$ and $x = 1.0$, $\rho_{sf\infty}$ instead of $\rho_{m\infty}$ is introduced.

The $\theta_D(x)$ determined points for the $Y(Fe_{1-x}Co_x)_2$ series (Fig. 3, curve 3) are described by the numerical formula $\theta_D(x) = (24x + 391)$ K. A tendency to slightly grow across the $Y(Fe_{1-x}Co_x)_2$ series can be observed. These θ_D values (Fig. 3) are situated closer the Debye temperatures known for Fe (423 K), Co (385 K) than temperature known for Y (214 K) metal [27]. The obtained $\theta_D(x)$ values coincide satisfactorily with the literature data obtained using specific heat studies performed for this series (Fig. 3, curve 3, open points) [31].

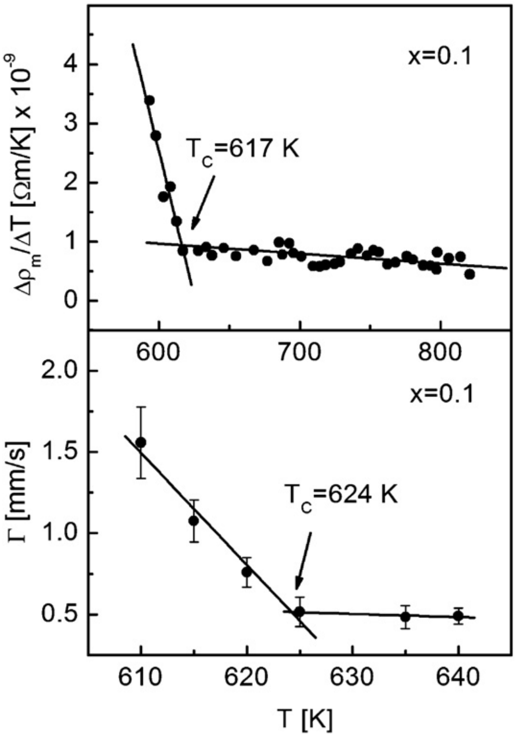

Fig. 4. The exemplary $\Delta\rho_m/\Delta T$ ratio (upper case) and the half-width Γ of the Mössbauer line (lower case) as functions of temperature T are presented for the $Y(Fe_{0.9}Co_{0.1})_2$ compound in the Curie temperature region. The intersection of lines determines the Curie temperature T_C.

4. Curie temperatures

A method based on the $\rho_m(T)$ dependence, presented previously elsewhere [17,28], was used to determine the magnetic ordering temperatures of the studied intermetallic series. Namely, the magnetic ordering temperature T_C is situated with a temperature range with the maximal change of numerically estimated parameter $\Delta\rho_m/\Delta T$ (Fig. 4, upper case). The intersection of the two fitted straight lines determines the magnetic ordering temperature. Experimental error depends on the quality of the $\Delta\rho_m/\Delta T$ dependence and a maximal error in this case can be estimated as $\Delta T_C \cong 17$ K.

The compound $Y(Fe_{0.1}Co_{0.9})_2$ has a Curie temperature below 15 K (a limit low temperature of performed measurements) or probably is an exchange-enhanced paramagnet similarly to the YCo_2 intermetallic. The compound YCo_2 without magnetic ordering temperature is an exchange-enhanced paramagnet reported in literature [30,32,33]. At a low temperature, the resistivity of these compounds follows the relation $\rho_{sf} = AT^2$ in agreement with spin fluctuation theory [30,32]. It is very interesting that a paramagnetic compound undergoes a first-order phase transition to a ferromagnetic state at a high magnetic field ($B_C = 70T$) with a discontinuous jump in the magnetization [34]. The spin fluctuations can be gradually quenched by the strong, internal magnetic field when the exchange interactions increase [35]. Moreover, the exchange interactions increase, when the Co atoms are substituted by Fe atoms, because Fe atoms in the $Y(Fe_{1-x}Co_x)_2$ series have individual magnetic moments. In the RCo_2 compounds, the cobalt moment exhibits an itinerant electron metamagnetic transition and it is induced by the exchange field originating from the

Table 1
The electrical resistivity parameters determined for the $Y(Fe_{1-x}Co_x)_2$ series.

x	n	A [Ωm/K^2] $\times 10^{-10}$	B [Ωm/K]$\times 10^{-9}$	C [Ωm]$\times 10^{-6}$	D [Ωm]$\times 10^{-10}$
0	6	0.101	0.319	2.019	97.715
0.1	6.1	0.064	1.020	1.852	130.589
0.2	6.2	0.066	1.522	3.200	309.380
0.3	6.3	0.097	0.924	2.043	838.154
0.4	6.4	0.011	1.076	1.518	566.025
0.5	6.5	0.196	0.080	2.284	615.110
0.6	6.6	0.099	1.189	2.234	2577.294
0.7	6.7	0.089	2.130	2.008	1187.431
0.8	6.8	0.404	1.471	2.381	1794.474
0.85	6.85	0.700	−1.313	1.794	1547.470
0.9	6.9	0.075	−0.078	1.883	1341.47
1	7	0.102	0.110	1.564	3477.03

localized 4f moments of the rare earth atoms through 3d–4f exchange interaction [36].

The Curie temperature dependence $T_C(x)$ for the $Y(Fe_{1-x}Co_x)_2$ series is presented in Fig. 5 (black points – from electrical resistivity). The $T_C(x)$ dependence was additionally confirmed (Fig. 5, open points) using a Mössbauer effect scanning method. The method was previously described elsewhere [37–39]. In this case a half-width $\Gamma(T)$ of the Mössbauer effect line as a function of temperature has been used. The intersection of two $\Gamma(T)$ lines determines Curie temperature (Fig. 4, lower case). Data of both the methods coincide well (Figs. 4 and 5). Given for a comparison, open squares, triangles and stars correspond to literature data [25,40,41]. The relatively high ordering temperature at $x = 0$ (576 K) shows a further strong increase with x (655 K for $x = 0.3$) and then rapidly drops in the cobalt rich region. It should be mentioned, that for $x = 0.9$ Curie temperature close to zero value has been reported [40]. Nevertheless from Mössbauer effect studies it has been found that even for $x = 0.978$ a magnetic order appears [2,12]. Moreover magnetic moment for $x = 0.9$ and $x = 1.0$ i.e. for rich cobalt region is observed [42]. For $Y(Fe_{0.1}Co_{0.9})_2$ compound the spin fluctuation contributions to the electrical resistivity is observed above 15 K similarly to the YCo_2 compound [30,32] (Fig. 2, $x = 0.9$, 1.0). The upper horizontal axis shows the average number n of 3d electrons calculated per transition metal atom following formula $n(x) = 6(1 - x) + 7 \cdot x$ (6 and 7 are the numbers of 3d electrons in the Fe and Co atoms correspondingly).

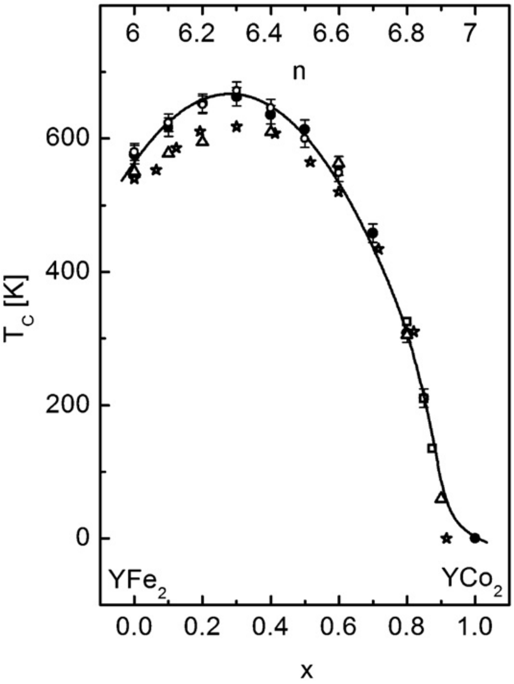

Fig. 5. The Curie temperature $T_C(x)$ of the $Y(Fe_{1-x}Co_x)_2$ series (black points-electrical resistivity measurements, open points-Mössbauer effect measurements). Open squares, triangles and stars correspond to literature data [25,40,41]. Upper axis shows number n.

5. Discussion

Electrical resistivity $\rho(T)$ is considerably influenced by Fe/Co substitution in the $Y(Fe_{1-x}Co_x)_2$ series. As a consequence of this substitution the parameters characterizing dependence of electrical resistivity on temperature and composition, in the majority, change across the series. Curie temperature T_C, which strongly depends on composition of the compound, divides $\rho(T)$ and thus $\rho_m(T)$ for particular compounds into two parts. Below T_C electrical resistivity grows up strongly and nonlinearly as thermal disorder among magnetic moments of atoms is introduced step by step with temperature. Above T_C there is no long-range magnetic order and the magnetic part of electrical resistivity is practically constant vs. temperature. At cobalt rich side of the $Y(Fe_{1-x}Co_x)_2$ series ($x = 0.9$, 1.0) there is no long range magnetic order and thus Curie temperature is not observed. In this case the magnetic contribution to electrical resistivity is related to spin fluctuations. It should be mentioned that there is no distinct relation between Fe/Co substitution and dynamical properties of crystal lattice, namely Debye temperature is practically constant across the $Y(Fe_{1-x}Co_x)_2$ series.

As a result of Fe/Co substitution the magnetic ordering temperature strongly varies across the $Y(Fe_{1-x}Co_x)_2$ series. As a rigorous expression for the magnetic ordering temperature related to the band structure of metallic ferromagnets or ferrimagnets is not

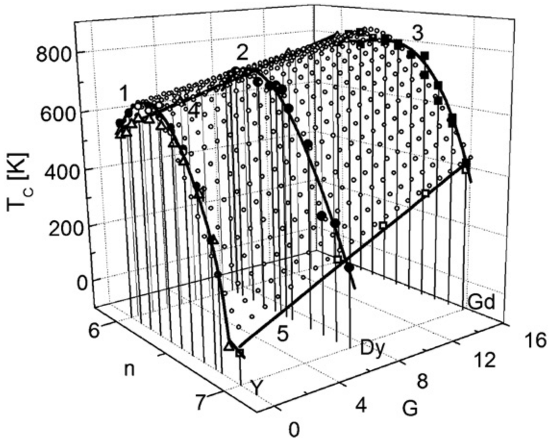

Fig. 6. The covering function $T_C(n, G)$ (marked by small open points) based on the series: $Y(Fe_{1-x}Co_x)_2$ (curve 1), $Dy(Fe_{1-x}Co_x)_2$ (curve 2) [17], $Gd(Fe_{1-x}Co_x)_2$ (curve 3) [15,16], $(Y_{1-x}Gd_xFe_2)$ (curve 4) [46] and $(Y_{1-x}Gd_xCo_2)$ (curve 5) [47].

known up to date there is not an easy task to explain the observed change in these Curie temperatures. Considering an approximate formula for the Curie temperature of intermetallics ($T_C = T_R + T_M$, T_R is rare earth sublattice contribution, T_M is transition metal sublattice contribution) [1,43,44] it can be expected that Fe/Co substitution mainly modifies the density of 3d states, especially 3d states at Fermi level, modifies the splitting energy between the 3d subbands, the transition metal magnetic moment, the exchange interactions and thus varies the Curie temperature across the series. To consider all these factors precisely is an open problem for further studies.

In Fig. 6 it can be seen a covering function $T_C(n, G)$, where G is the de Gennes factor [45], fitted to the Curie temperature experimental data of the series: $Y(Fe_{1-x}Co_x)_2$ (curve 1), $Dy(Fe_{1-x}Co_x)_2$ (curve 2) [17], $Gd(Fe_{1-x}Co_x)_2$ (curve 3) [15,16], $(Y_{1-x}Gd_x)Fe_2$ (curve 4) [46] and $(Y_{1-x}Gd_x)Co_2$ (curve 5) [47]. The $T_C(n, G)$ surface (Fig. 6, marked by small open points) approximates satisfactorily experimental points and is described by numerical formula:

$$T_C(n, G) = \left\{ \frac{682.162 - 0.147n}{0.899 + 0.021n} \cdot \exp\left[\frac{-2(n - 6.311)^2}{(0.899 + 0.021n)^2} \right] + 10.613G + 1.341 \right\} \text{K}. \tag{4}$$

The $T_C(n, G)$ numerical formula could be helpful to predict magnetic ordering temperature of an unknown compound corresponding to the considered (n, G) area. It should be mentioned that without knowing band structure and rigorous theoretical T_C formula (to compare with expression (4)) a direct physical interpretation cannot be related to determined numerical parameters occurring in the above formula (4).

Summarizing, it should be emphasized that a more exhaustive discussion of Curie temperatures of the $Y(Fe_{1-x}Co_x)_2$ series or more generally of $R(Fe_{1-x}Co_x)_2$ series could be possible after knowing at least band structure and exchange interactions for these compounds.

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