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2003 J. Phys.: Condens. Matter 15 5371

(http://iopscience.iop.org/0953-8984/15/31/304)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) 5371–5382 PII: S0953-8984(03)62253-8

Onset of magnetism in $Y_{1-x}Gd_xCo_2$: effect on the heat capacity and electrical resistivity

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Received 15 April 2003
Published 23 July 2003
Online at stacks.iop.org/JPhysCM/15/5371

Abstract

Results of the electrical resistivity, magnetoresistance and specific heat measurements are presented for $Y_{1-x}Gd_xCo_2$ in the vicinity of the critical Gd concentration $x_c \approx 0.15$ for the onset of a long-range magnetic order. Pronounced maxima of the coefficient γ of the T-linear specific heat, Debye temperature and residual resistivity as a function of concentration are found to exist at $x = 0.1$, i.e. just below x_c. These anomalies, together with the existence of a minimum on the temperature dependences of the electrical resistivity at $0 < x < 0.15$, are ascribed to the substantial influence of localized spin density fluctuations induced by f–d exchange in the itinerant d-electron subsystem of Co. The maximal extra contribution to the residual resistivity $\Delta\rho_{res}^{max}$ in $Y_{1-x}R_xCo_2$ pseudobinaries is found to depend on the critical concentration as well as on the type of R ions, while the value of $\Delta\gamma_{max}$ depends only on the critical concentration of R ions. The significant upturns on C_p/T versus T^2 dependences observed in $Y_{1-x}Gd_xCo_2$ at $0 < x < 0.15$ may be indicative of a non-Fermi-liquid behaviour of the d-electron subsystem caused by the partial substitution of Gd for Y.

1. Introduction

Within the series of rare earth Laves phase compounds RCo_2, the pseudobinary systems $Y_{1-x}R_xCo_2$ are of special interest since the substitution of R ions having nonzero magnetic moment for the nonmagnetic yttrium leads to the appearance of a long-range magnetic order in the itinerant 3d-electron subsystem of Co, while the parent compound YCo_2 exhibits an exchange-enhanced Pauli paramagnetism [1]. Anomalous behaviour of different physical

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properties was observed in the concentration range around the critical value x_c at which the magnetic moment on cobalt atoms μ_{Co} appears under the influence of the molecular field, H_{eff}, associated with the f–d exchange interaction. In RCo_2 the itinerant 3d electrons of Co hybridized with 5d (4d in the case of Y) electrons of R ions reveal so-called band metamagnetism. An increase of H_{eff} up to the critical value ~70 T at substitution leads to an abrupt increase of μ_{Co} from nearly zero up to a value of about 1 μ_B. The metamagnetic transition in the itinerant d-electron subsystem may also be produced by application of a high external field [2]. It should also be noted that RCo_2 are well known objects, the properties of which are strongly influenced by spin fluctuations. Such a particular magnetic behaviour of RCo_2 originates from peculiarities of the electronic structure of these compounds. The Fermi level lies in a local valley between two peaks in the density of electronic states curve for the d-band. The spin fluctuations in the d-band are considered to develop owing to the high value and positive curvature of the density of electronic states near the Fermi level [3].

The magnetic properties of $Y_{1-x}Gd_xCo_2$ compounds have been studied in numerous works by different methods [4–9]. From bulk magnetization measurements it was suggested that the long-range ferrimagnetic order with the antiparallel alignment of Gd and Co magnetic moments starts to develop from $x \approx 0.15$ with increasing Gd content [4, 5]. Using NMR measurements it has been shown that around this concentration two kinds of Co atoms (magnetic and nonmagnetic) coexist [7]. The magnetic state of the compounds for $x < 0.15$ was classified as a spin-glass or cluster-glass type [8]. A remarkable change in the behaviour of the thermopower and magnetoresistance was observed with increasing Gd content within $0 < x \leqslant 0.3$ [10].

In the present study we focus on the behaviour of the specific heat and electrical resistivity of $Y_{1-x}Gd_xCo_2$ compounds in the vicinity of the critical Gd concentration.

2. Experimental details

The $Y_{1-x}Gd_xCo_2$ compounds were obtained by arc melting in a helium atmosphere using yttrium and gadolinium of 99.9% and cobalt of 99.99% purity. The ingots were remelted several times to achieve a good homogeneity of the samples. To avoid an appearance of foreign phases, a 6 wt% excess of rare earth and yttrium over the stoichiometric composition was used. The alloys were then annealed in an evacuated sealed quartz tube for one week at 800 °C. According to x-ray diffraction all the samples were single phase with a cubic crystal structure of $MgCu_2$ type.

The ac magnetic susceptibility was measured on the polycrystalline samples at the frequency 1 kHz and field amplitude of 1×10^{-4} T using PPMS (Quantum Design, USA). The specific heat was measured also using PPMS on the same samples with a mass of 5–10 mg in magnetic fields up to 5 T. The measurements of the dc electrical resistivity were performed on the parallelepiped samples of $1 \times 1 \times 5$ mm^3 using the four-contact circuit at temperatures from 2 K up to 300 K in magnetic fields up to 7 T. The experimental error of the electrical resistivity was estimated to be within ±5%.

3. Results

The temperature dependences of the ac susceptibility measured on $Y_{1-x}Gd_xCo_2$ samples with the Gd content below $x = 0.4$ show a cusp-like peak suggesting a magnetic ordering (insets in figure 1). The concentration dependence of the critical temperature T_{crit}, which can be well defined from the position of such peaks, is presented in figure 1 for the interval $0 < x \leqslant 0.4$. The change in the $T_{crit}(x)$ slope around $x \approx 0.15$ may be associated with the change in the

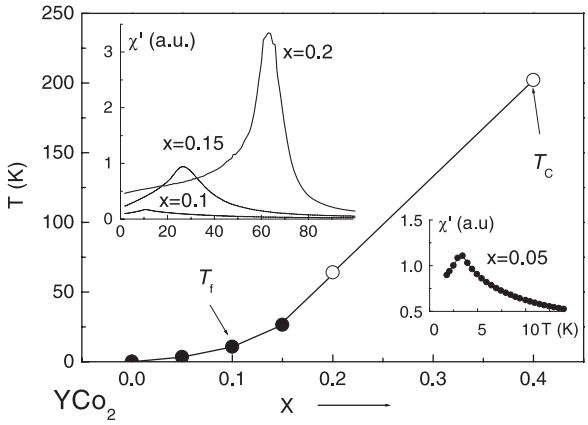

Figure 1. Low Gd concentration part of the magnetic phase diagram of the $Y_{1-x}Gd_xCo_2$ system. T_f and T_C denote the freezing temperature and Curie temperature, respectively. Insets show $\chi'(T)$ dependences at various Gd content.

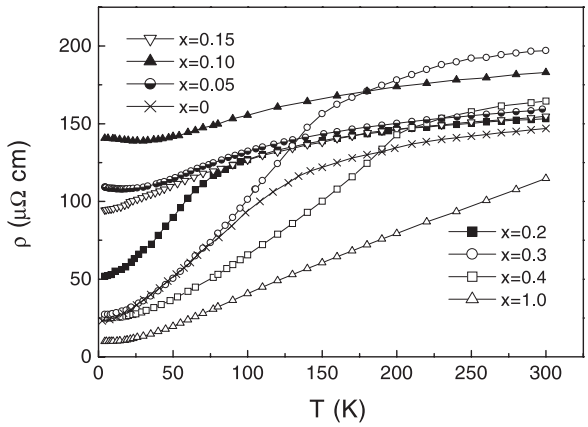

Figure 2. Temperature dependences of the electrical resistivity of $Y_{1-x}Gd_xCo_2$ compounds at various x.

magnetic state from a cluster-glass regime at $x < 0.15$ to a non-homogeneous ferrimagnetic state at the higher Gd content.

Figure 2 displays the temperature dependences of the electrical resistivity of $Y_{1-x}Gd_xCo_2$ compounds with different Gd content. As can be seen, the substitution of Gd for Y up to $x = 0.1$ increases drastically the residual resistivity ρ_{res} and significantly modifies the ρ versus T curves in the low-temperature region. Unlike the T^2 dependence of the low-temperature resistivity for YCo_2 [11] the pronounced minimum appears on $\rho(T)$ curves for $Y_{1-x}Gd_xCo_2$ compounds with $x = 0.05$ and 0.1. Nevertheless, the $\rho(T)$ dependences for the compounds with $x < 0.15$ do not show any anomaly when the temperature decreases below the critical value $T = T_f$ which is associated with freezing of the magnetic moments of ferrimagnetic clusters in random directions. An increase of the Gd content above $x = 0.1$ recovers a normal behaviour of $\rho(T)$ as well as significantly suppresses the residual resistivity. Thus we have obtained a non-monotonic change of $\rho_{res}(x)$ with a maximal value at $x = 0.1$, i.e. just below the critical concentration x_c (see figure 3).

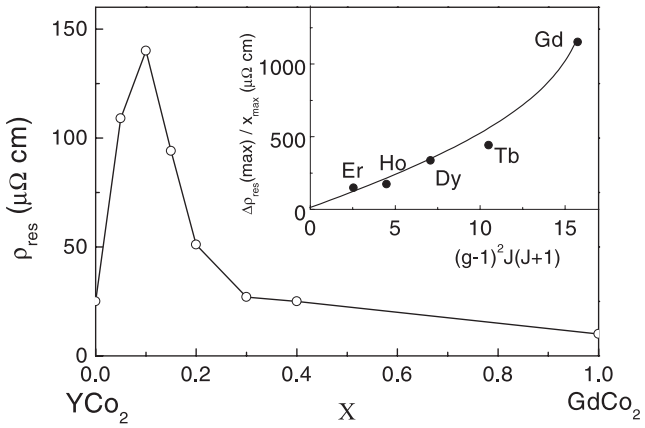

Figure 3. Concentration dependence of the residual resistivity of $Y_{1-x}Gd_xCo_2$ compounds. The inset shows the normalized extra contribution to the residual resistivity $\Delta\rho_{res}^{max}/x_{max}$ versus the de Gennes factor (the data presented in the table 1 are used).

Table 1. The maximal value of the extra contribution to the residual resistivity $\Delta\rho_{res}^{max} = \rho_{res}^{max}(Y_{1-x}R_xCo_2) - \rho_{res}(YCo_2)$, the maximal value of the extra contribution to the coefficient of the T-linear specific heat $\Delta\gamma_{max} = \gamma_{max}(Y_{1-x}R_xCo_2) - \gamma(YCo_2)$ and the concentrations of R ions x_{max}^{ρ}, x_{max}^{γ} at which the maximal values of ρ_{res} and γ are observed for $Y_{1-x}R_xCo_2$ pseudobinaries.

$(Y_{1-x}R_x)Co_2$	$\Delta\rho_{res}$(max) ($\mu\Omega$ cm)	$x_{max}(\rho)$	$\Delta\gamma$ (mJ mol^{-1} K^{-2})	$x_{max}(\gamma)$	x_c
$(Y_{1-x}Gd_x)Co_2$[a]	117	0.1	32	0.1	0.15
$(Y_{1-x}Tb_x)Co_2$	90 [23]	0.2 [23]	32 [15]	0.1 [15]	0.2 [23]
$(Y_{1-x}Dy_x)Co_2$	69 [29]	0.3 [29]	61 [30]	0.2 [30]	0.3 [30]
$(Y_{1-x}Ho_x)Co_2$	71 [22]	0.42 [22]	124 [23]	0.2 [23]	0.43 [13]
$(Y_{1-x}Er_x)Co_2$	76 [21]	0.5 [21]	163 [1]	0.5 [1]	0.55 [21]

[a] Present work.

The great difference in the magnetic state of $Y_{1-x}Gd_xCo_2$ below and above the critical concentration is clearly shown by magnetoresistance measurements of these compounds. The field dependences of longitudinal magnetoresistance $\Delta\rho/\rho = [\rho(H) - \rho(0)]/\rho(0)$ are presented in figure 4 for compounds in the concentration range $0.1 \leqslant x \leqslant 0.3$. An application of a magnetic field H causes a gradual lowering of the electrical resistivity of $Y_{0.9}Gd_{0.1}Co_2$, which presumably exhibits a cluster-glass magnetic state at $H = 0$. A different behaviour of the magnetoresistance is observed for the compounds with a Gd content above x_c. At first, the electrical resistivity of the compounds with $x = 0.2$ and 0.3 slightly reduces under an applied magnetic field and then the resistivity shows a gradual growth, with a further increase of the field giving the positive value of $\Delta\rho/\rho$ at high magnetic fields. It should be noted that the positive longitudinal magnetoresistance (it is not shown in figure 4) was also observed for the parent compound YCo_2 in previous works (see [12] for instance). Initially there may seem to be some contradiction with the current opinion that an applied field should suppress spin fluctuations in this compound and, as a consequence, should reduce its electrical resistivity. However, a growth of ρ with increasing field in the low-temperature range seems to be quite reasonable in the case of YCo_2, bearing in mind that an applied field destabilizes the itinerant d-electron subsystem of YCo_2 when H approaches the critical value for the metamagnetic-like transition.

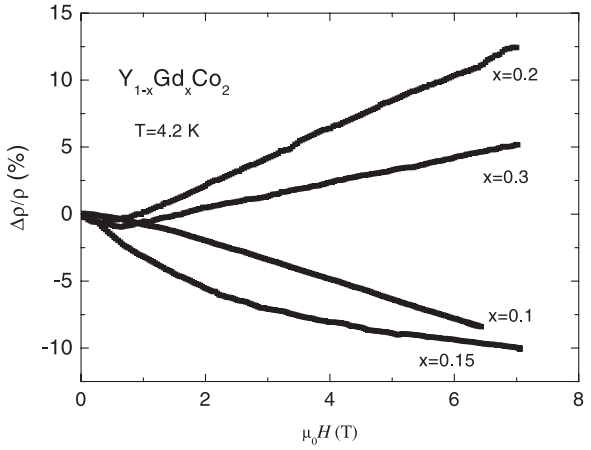

Figure 4. Field dependences of the longitudinal magnetoresistance measured at $T = 4.2$ K on $Y_{1-x}Gd_xCo_2$ compounds with $0.1 \leqslant x \leqslant 0.3$.

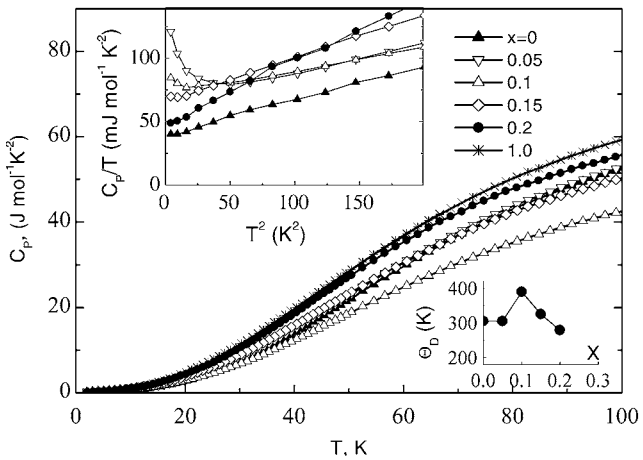

Figure 5. Temperature dependences of the specific heat of $Y_{1-x}Gd_xCo_2$. The insets show the C_p/T versus T^2 curves and concentration dependence of the Debye temperature at $0 \leqslant x \leqslant 0.2$.

Figure 5 presents the results of specific heat measurements on $Y_{1-x}Gd_xCo_2$ compounds in the temperature range 2–100 K. As is seen, the $C_p(T)$ curves do not reveal any apparent indications which could be associated with the change of the magnetic state of these compounds. Meanwhile, the C_p/T versus T^2 dependences plotted for compounds with different x show a dramatic change in the value of the coefficient γ of the T-linear term of the specific heat. The value of γ for the parent compound YCo_2 was estimated to be equal to 37 mJ mol^{-1} K^{-2}, which is in agreement with previous data (34–36.5 mJ mol^{-1} K^{-2} [13, 14]). The value of $\gamma = 39$ mJ mol^{-1} K^{-2} obtained in the present work for $GdCo_2$ is close to that observed for YCo_2 as well as for other RCo_2: 39.9 mJ mol^{-1} K^{-2} for $TbCo_2$ [15]; 37 mJ mol^{-1} K^{-2} for $HoCo_2$ [13]; 38 mJ mol^{-1} K^{-2} for $ErCo_2$ [16], although YCo_2 is an exchange-enhanced Pauli paramagnet, whereas other compounds mentioned above (RCo_2) are ferrimagnets exhibiting a long-range magnetic order in the itinerant d-electron subsystem at low temperatures. Assuming a similarity of electronic structures of all these compounds

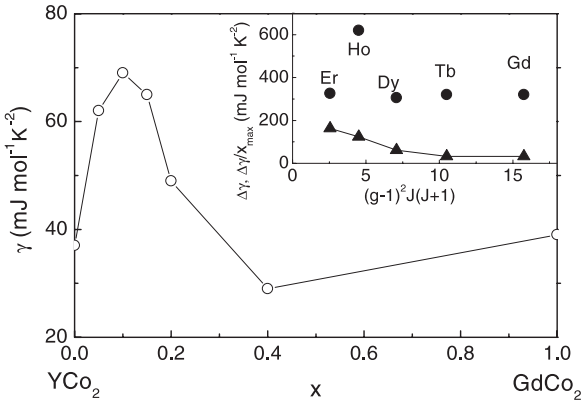

Figure 6. Concentration dependence of the coefficient γ of the linear term of the specific heat of $Y_{1-x}Gd_xCo_2$. The inset shows $\Delta\gamma_{max}$ (triangles) and $\Delta\gamma_{max}/x_{max}$ (circles) versus the de Gennes factor.

one can conclude that the d-band splitting does not appreciably influence the γ value and that the spin fluctuations give the predominant contribution to the T-linear specific heat also in the magnetically ordered state of RCo_2. The concentration dependence of the coefficient γ (figure 6) for $Y_{1-x}Gd_xCo_2$ looks similar to the $\rho_{res}(x)$ dependence and shows a pronounced maximum at $x = 0.1$.

In the inset in figure 5 one can see the low-temperature upturns on the C_p/T versus T^2 dependences for $Y_{1-x}Gd_xCo_2$ compounds with a low Gd concentration ($x < 0.15$). In order to evaluate an additional magnetic contribution to the heat capacity arising from the substitution of Gd for Y we determined the difference between the $C_p(T)$ curves for the Gd-substituted compounds and $C_p(T)$ for YCo_2: $\Delta C_m = C_{alloy} - C_{YCo_2}$. Such a contribution divided by the temperature, $\Delta C_m/T$, is displayed in figure 7 for the compound $Y_{0.95}Gd_{0.05}Co_2$ as a function of the temperature. This extra contribution to the specific heat is characterized by the presence of a broad Schottky-type anomaly around 30 K and of an additional low-temperature upturn. Both these anomalies demonstrate a significantly different behaviour under application of a magnetic field. As is shown in figure 7, an applied field 5 T does not noticeably influence the low-temperature anomaly while it increases appreciably the $\Delta C_m/T$ values in the vicinity of the broad high-temperature maximum. The broad maximum of $\Delta C_m/T$ lies at a significantly higher temperature than the peak of the ac susceptibility that indicates the freezing of cluster magnetic moments at $T_f = 3.4$ K (see the inset in figure 7). This is the usual situation for dilute magnetic systems [17].

Another feature of the $Y_{1-x}Gd_xCo_2$ system is a reduced lattice contribution to the total specific heat in compounds with a Gd content around $x = 0.1$, where γ and ρ_{res} have maximal values (see figure 5). Such a reduction corresponds to a significant enhancement of the Debye temperature, Θ_D. The change of Θ_D at the Gd substitution in a low concentration range is shown in the inset in figure 5. For YCo_2, the value Θ_D was estimated to be about 263 K from the slope of the C_p/T versus T^2 dependence in the temperature interval 2–15 K. This value is close to previous results ($\Theta_D = 226$ K [13], 252 K [18]), which were obtained in the same way. However, we have found that the best fitting of the experimental curve $C_p(T)$ for the wide temperature range (up to 100 K) may by achieved with $\Theta_D = 305$ K. The last value is consistent with $\Theta_D = 300 \pm 5$ K which was obtained from the $C_p(T)$ dependence in a temperature range up to 300 K in [19]. Such a discrepancy in values of Θ_D determined for the

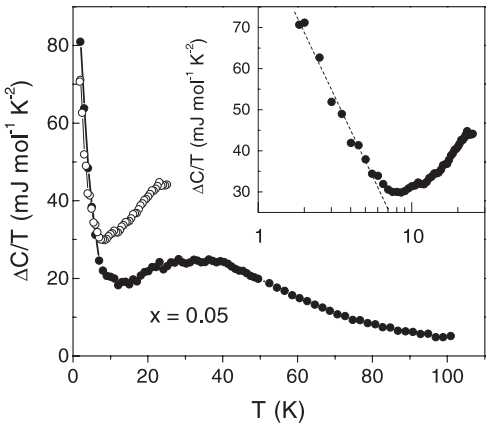

Figure 7. $\Delta C_p/T$ versus T for $Y_{0.95}Gd_{0.05}Co_2$ at zero magnetic field (full circles) and at $\mu_0 H = 5$ T (open circles). Inset: $\Delta C_p/T$ versus $\log T$ for $Y_{0.95}Gd_{0.05}Co_2$ at zero magnetic field.

different temperature intervals may be attributed to the enhancement of the spin fluctuation contribution to the lattice specific heat with increasing temperature owing to an increase of the local amplitude of spin fluctuations. According to the theory of the magnetoelastic effects in weakly itinerant ferromagnetic systems, the spin fluctuations may give rise to a positive magnetic pressure that implies a stiffening of the lattice and an increase of Θ_D [20].

In order to evaluate the Debye temperatures for Gd-substituted compounds in the concentration range $0 < x \leqslant 0.2$ we have used a high-temperature part of $C_p(T)$ curves between 60 and 100 K where all these compounds are paramagnetic and the magnetic contribution to the total specific heat becomes insignificant. As concerns the coefficient γ, we have taken it to be equal to 37 mJ mol^{-1} K^{-2} as for the parent YCo_2. As is seen from the inset in figure 5, the substitution of Gd for Y up to $x = 0.1$ leads to the dramatic increase of Θ_D up to 390 K. A further increase of the Gd content reduces Θ_D down to 279 K at $x = 0.2$. The last value is close to that for $GdCo_2$ ($\Theta_D = 264$ K), which was estimated taking into account the difference in molar mass between YCo_2 and $GdCo_2$.

4. Discussion

The magnetic structure of $Y_{1-x}Gd_xCo_2$ compounds cannot be studied in detail by neutron diffraction because of the very large absorption of neutrons by Gd. The existence of a non-homogeneous ferrimagnetic structure was established by means of neutron diffraction studies of isostructural compounds $Y_{1-x}R_xCo_2$ (R = Er, Ho, Tb) [21–24]. In all these systems a coexistence of the long-range and short-range order was observed close above the critical concentration of R ions. At $x < x_c$ only the broad maxima of the magnetic diffuse scattering were found to exist in $Y_{1-x}R_xCo_2$ together with Bragg reflections associated with nuclear scattering. The magnetic diffuse scattering was attributed to the presence of magnetic short-range correlations in $Y_{1-x}R_xCo_2$ at $x < x_c$. Therefore, taking into account the NMR data [7] the magnetic structure of $Y_{1-x}Gd_xCo_2$ with x above $x_c \approx 0.15$ can also be described as a ferrimagnetic matrix with the antiparallel alignment of Gd magnetic moments and Co magnetic moments induced by H_{eff}. Moreover, because of a non-homogeneous distribution of Gd ions over the lattice, this matrix presumably includes the regions (clusters) consisting of partially disordered Gd magnetic moments and Co ions with zero magnetic moment, since the value of

H_{eff} for these clusters is lower than the critical value. The ratio between the volume of the matrix with long-range magnetic order and the volume of clusters with short-range order will change at the substitution. The critical concentration of R ions (Gd in our case) may be considered as a percolation limit [22]. As to the magnetic state of $Y_{1-x}Gd_xCo_2$ with $0.05 \leqslant x < x_c$ one can assume the presence of small regions (clusters) in the volume of the sample in which $\mu_{Co} \neq 0$, while in the matrix $\mu_{Co} = 0$. An appearance of such magnetic clusters and localized spin density fluctuations (LSF) in the d-electron subsystem can be associated with the 'microscopic' metamagnetic phase transitions which take place only in such regions of the sample where the molecular field H_{eff} acting from Gd ions exceeds the critical value.

Bearing in mind the above picture of the magnetic state of $Y_{1-x}Gd_xCo_2$ compounds with $0.05 \leqslant x < 0.15$, the conduction electron scattering on LSF in the d-electron subsystem can be considered as a main reason for the giant enhancement of the residual resistivity ρ_{res} at $x = 0.1$. The contribution from s-electron scattering on Gd magnetic moments seems to be negligible in this concentration range. An appearance of a minimum on $\rho(T)$ dependences in the compounds with $x = 0.05$ and 0.1, in our opinion, originates from the superposition of an extra contribution caused by LSF and contributions arising from the scattering on phonons and on the thermally activated spin fluctuations. Both latter contributions to the resistivity grow with increasing T in the low-temperature range. Unlike this, an additional contribution to the resistivity from the scattering on LSF increases with lowering temperature. Such a temperature dependence of the extra contribution may be attributed to an increasing number and volume of clusters where the effective field H_{eff} exceeds the critical value. This suggestion is confirmed by the correlation between the resistivity and intensity of the small-angle neutron scattering, which was observed for $Y_{1-x}Er_xCo_2$ compounds exhibiting an analogous minimum on the $\rho(T)$ dependence at $x < x_c$ [22].

A decrease of ρ observed in $Y_{1-x}Gd_xCo_2$ at $0 < x < x_c$ under application of a magnetic field may be associated with suppression of LSF. The applied field causes the rotation of the cluster magnetic moments toward the direction of H and suppresses the magnetic moment on Co atoms since H is directed against the effective field H_{eff} acting from Gd. As a result, the amplitude of LSF is reduced, leading to a gradual decrease of the contribution from scattering on such fluctuations. This assumption is supported by magnetostriction measurements presented in [9], where the negative magnetovolume effect in $Y_{1-x}Gd_xCo_2$ ($0.025 \leqslant x < 0.15$) was associated with a reduced magnetization of the Co subsystem under application of an external field.

Suggesting that the ferrimagnetic matrix of $Y_{1-x}Gd_xCo_2$ compounds with the Gd content just above x_c includes clusters consisting of partly disordered Gd magnetic moments and Co ions with $\mu_{Co} = 0$, a non-monotonic change of the electrical resistivity in an applied field (shown in figure 4), may be explained by the following. At first, an application of H makes an ordering effect on the partly disordered Gd magnetic moments in clusters, which results in growth of H_{eff} up to the critical value and leads to enhancement of the magnetic moments on Co atoms situated in these clusters up to the value that is close to μ_{Co} in the matrix. Therefore, the electrical resistivity decreases in this field region owing to the suppression of LSF. An increase of an external field hereafter causes an inverse effect on the Co subsystem. Because of the antiparallel alignment of magnetizations of Gd and Co sublattices, a further increase of H reduces the effective field acting on Co atoms. Since the itinerant d-electron subsystem becomes unstable when H_{eff} diminishes approaching the critical value for a collapse of the magnetic moment on Co atoms, the spin fluctuations contribution develop to growth resulting in a positive change of the electrical resistivity. It should be noted that the above suggested enhancement of μ_{Co} under application of a small external field was actually revealed by the neutron diffraction measurements in $Y_{1-x}R_xCo_2$ compounds (R = Er, Ho) with R content

just above the critical value [21–24]. As to the high field effect, the measurements of the volume magnetostriction performed by Yamaguchi *et al* [25] have shown that the high magnetic field significantly reduces the Co magnetic moment in $Y_{1-x}Gd_xCo_2$. A disappearance of the magnetic moment on Co atoms or the so-called inverse metamagnetic transition in RCo_2-based compounds under application of high magnetic fields was observed in several works by means of magnetization, magnetoresistance and magnetostriction measurements [26–28].

The non-monotonic concentration dependences of the residual resistivity ρ_{res} and the coefficient γ of the T-linear specific heat with a maximum just below x_c were observed in all $Y_{1-x}R_xCo_2$ systems with heavy rare earths, except for Tm (see [23] and references therein). In our opinion, the presence of such maxima on $\rho_{res}(x)$ and $\gamma(x)$ originates in complementary contributions to both these characteristics from LSF induced by the effective field acting from R ions at the concentration just before the onset of a long-range magnetic order in the Co subsystem. Table 1 displays the maximal values of the additional contribution to the residual resistivity $\Delta\rho_{res}^{max} = \rho_{res}^{max}(Y_{1-x}R_xCo_2) - \rho_{res}(YCo_2)$ for $Y_{1-x}R_xCo_2$ as well as the concentrations x_{max}^{ρ} at which these maxima were observed. The value of the residual resistivity of the parent compound YCo_2 was taken to be equal to 23 $\mu\Omega$ cm (see figure 2). The additional contribution to the γ value $\Delta\gamma_{max} = \gamma_{max}(Y_{1-x}R_xCo_2) - \gamma(YCo_2)$, as well as the concentration of R ions x_{max}^{γ} at which the maximal value of γ_{max} is observed, are also presented in table 1. It seems reasonable to suggest that the extra contributions $\Delta\rho_{res}^{max}$ and $\Delta\gamma_{max}$ will depend on the local amplitude of LSF and accordingly on the spin of the R ion that induces these fluctuations. Moreover, one could expect that $\Delta\rho_{res}$ and $\Delta\gamma_{max}$ will be dependent on the concentration of R ions which induce these LSF. Therefore, we plotted $\Delta\rho_{res}^{max}/x_{max}^{\rho}$ and $\Delta\gamma_{max}/x_{max}^{\gamma}$ as a function of the de Gennes factor $G = (g_J - 1)^2 J(J+1)$. As it turned out, the normalized extra contribution to the residual resistivity $\Delta\rho_{res}^{max}/x_{max}^{\rho}$ reveals a strong dependence on the G value indeed (see the inset in figure 3). As to the contribution to the T-linear specific heat, the ratio $\Delta\gamma_{max}/x_{max}^{\gamma}$ does not exhibit an apparent dependence on the type of the R ion, unlike the normalized extra contribution to the resistivity. This means that the value of $\Delta\gamma_{max}$ depends only upon the concentration of R ions which is necessary to produce the effective field close to the critical one. Some scatter of $\Delta\gamma_{max}/x_{max}^{\gamma}$ values arises probably from difficulties in the evaluation of the coefficient γ as well as in the determination of x_{max}^{γ} from the low-temperature specific heat data bearing in mind the low values of the magnetic ordering temperature and complicated magnetic behaviour of $Y_{1-x}R_xCo_2$ compounds in the concentration range around x_c. As follows from table 1 (see also [23]) the values of x_{max}^{ρ} and x_{max}^{γ} are lower than the critical concentrations x_c for the onset of a long-range magnetic order in all $Y_{1-x}R_xCo_2$. Moreover, in some cases the position of ρ_{res}^{max} is shifted to higher R content with respect to the position of γ_{max} for the same $Y_{1-x}R_xCo_2$ system. This is presumably because the s-electron scattering on LSF depends not only on the amplitude of LSF but also on its extent in comparison with the electron free path in the sample.

The low-temperature upturn of C_p/T was detected only for the $Y_{1-x}Gd_xCo_2$ compounds with Gd content below x_c. One may assume that it originates from a second Schottky-type anomaly caused by magnetic clusters. On the other hand, such a divergence of C_p/T with decreasing temperature may be associated with a non-Fermi liquid (NFL) behaviour of the itinerant d-electron subsystem in the vicinity of a magnetic phase transition that occurs near $T = 0$ K. Since some NFL theories predict a logarithmic T dependence of C_p/T (see [31], for example), we have used this form ($C/T = \gamma + A\log T$) to fit the data for $Y_{0.95}Gd_{0.05}Co_2$. The $\Delta C_m/T$ versus T data for this compound in a logarithmic scale are plotted in the inset in figure 7 where the logarithmic form below 6 K describes the data quite well. However, further measurements of the specific heat at lower temperatures are needed in order to specify a scaling behaviour. Usually, together with the specific heat, the NFL behaviour may be detected by the

electrical resistivity and magnetic susceptibility measurements. Unfortunately, it seems to be difficult to make a definite conclusion from analysis of these characteristics in our case, since in $Y_{1-x}Gd_xCo_2$ the situation is complicated by the presence of magnetic clusters and freezing phenomena as well as by the metamagnetism of the 3d-electron subsystem of Co.

An enhanced value of Θ_D is observed at the same Gd content ($x = 0.1$), at which γ and ρ_{res} also exhibit the maximal values. Thus we suggest that such an enhancement originates also from the presence of LSF induced by f–d exchange in the itinerant d-electron subsystem. As to the decrease of Θ_D with increasing Gd content above $x = 0.1$, it cannot be ascribed to the expansion of the lattice caused by the appearance of the long-range magnetic order in the Co subsystem since the Θ_D values were determined by using the high-temperature data of $C_p(T)$. Assuming that the above anomalies of different physical properties of $Y_{1-x}Gd_xCo_2$ around $x = 0.1$ originate from the presence of LSF, which are localized in real space, a softening of the lattice at $x > 0.1$ may be attributed to extension (delocalization) and mutual interaction of LSF with increasing Gd content. Moreover, the possible change in the temperature dependence of the mean-square local amplitude of the spin fluctuation in the d-electron subsystem [32] cannot be neglected when analysing the effect of spin fluctuations in the paramagnetic region of $Y_{1-x}Gd_xCo_2$ at various Gd concentrations.

It should be noted that a non-monotonic change of ρ_{res}, γ and Θ_D with maxima around the critical concentration for the appearance of the long-range magnetic order was also observed for the $Lu(Co_{1-x}M_x)_2$ and $Y(Co_{1-x}M_x)_2$ compounds [33–36] at the partial substitution of M(=Al, Ga) atoms for Co. In our opinion, the presence of such anomalies in these systems is also associated with the influence of LSF. Nevertheless, in $Y(Co_{1-x}M_x)_2$ and $Lu(Co_{1-x}M_x)_2$ such fluctuations originate from the distribution in the local density of electronic states and/or from the distribution in the d–d exchange owing to the partial substitution of M atoms for Co, unlike the $Y_{1-x}R_xCo_2$ compounds, where the LSF are produced by f–d exchange owing to the substitution of R ions having a local magnetic moment for nonmagnetic Y.

5. Conclusion

The measurements of the electrical resistivity, magnetoresistance and specific heat performed in the present work for the $Y_{1-x}Gd_xCo_2$ system have revealed a set of peculiarities in the vicinity of the onset of the long-range magnetic order at the substitution of Gd for Y. It has been found that the concentration dependence of the residual resistivity, the coefficient γ of the T-linear specific heat and the Debye temperature exhibit pronounced maxima at $x = 0.1$, i.e. just below the critical concentration $x_c \approx 0.15$. The minimum on the temperature dependence of the electrical resistivity is found to exist for compounds with a Gd content between $0.05 \leqslant x < 0.15$. The longitudinal magnetoresistance reveals a significantly different behaviour below and above the critical concentration. The extra contribution to the specific heat $\Delta C_m/T$ caused by substitution of 5 at.% Gd for Y shows a broad maximum at high temperatures (above the freezing temperature indicated by the ac susceptibility peak) and low-temperature upturn having a $\log T$ behaviour. Mainly, the above anomalies were also observed in other $Y_{1-x}R_xCo_2$ compounds with heavy rare earths except Tm. In our opinion, these anomalies around x_c originate from the presence of the LSF induced by f–d exchange in the itinerant d-electron subsystem due to the non-homogeneous distribution in the lattice of R ions having a localized magnetic moment. The magnetic state of $Y_{1-x}Gd_xCo_2$ as well as of other $Y_{1-x}R_xCo_2$ below x_c may be pictured as a paramagnetic matrix which includes small regions (clusters) where LSF is induced on Co atoms by the local molecular field arising from Gd ions via f–d exchange interaction. Above the critical concentration these compounds exhibit a non-homogeneous ferrimagnetic state formed by an antiparallel alignment of Gd

and Co magnetic moments. This ferrimagnetic matrix includes regions (clusters) with partly disordered Gd moments and Co ions with nearly zero magnetic moment.

A comparative analysis of the electrical resistivity and specific heat of $Y_{1-x}R_xCo_2$ pseudobinaries has shown that the maximal additional contribution to the residual resistivity caused by the influence of LSF strongly depends on the critical concentration as well as on the de Gennes factor of R ions, while the additional contribution to the coefficient γ of the T-linear specific heat $\Delta\gamma_{max}$ depends only upon the concentration of R ions, which should be enough to produce an effective field close to the critical one and to generate LSF in the itinerant d-electron subsystem. A step-like character of the $\mu_{Co}(H_{eff})$ dependence in $Y_{1-x}R_xCo_2$, when H_{eff} approaches the critical value with increasing x, causes an abrupt development of LSF just before the critical concentration for the onset of the long-range magnetic order. The low-temperature upturns of $\Delta C_m/T$ versus T observed for $Y_{1-x}Gd_xCo_2$ ($0 < x < 0.15$) may be indicative of the NFL behaviour of the itinerant 3d-electron subsystem of Co near the quantum critical point as well as for magnetic cluster effects. Such a finding requires further specific heat measurements of these compounds at lower temperatures.

Acknowledgments

This work was supported by the Swiss National Science Foundation (grant SCOPES no 7 IP 65598) and by grant N REC-005 of US CRDF.

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