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Study of the d-magnetism instability in the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ system around the critical concentration by means of resistivity measurements

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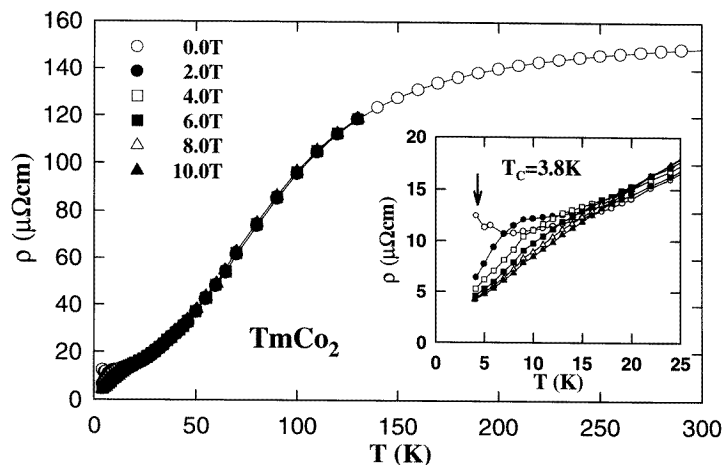
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Received 11 July 1996

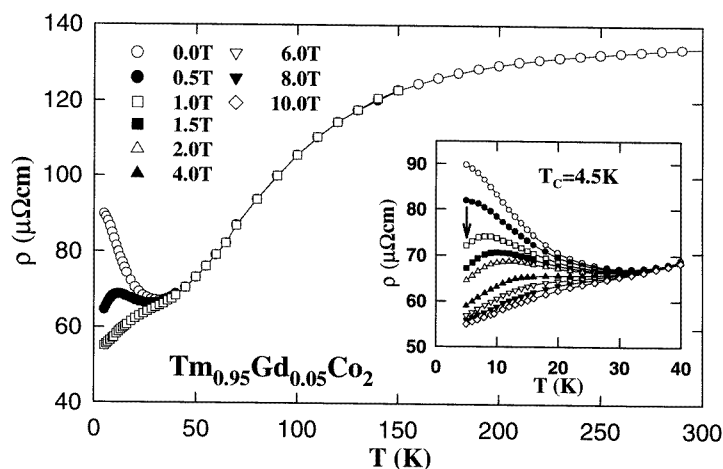
Abstract. The substitution of Gd for Tm in the pseudobinary $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ system gives rise to the onset of long-range magnetic order in the itinerant Co sublattice above a critical concentration ($x_c = 0.1$). The features of the Co magnetism around the critical concentration were studied by means of resistivity measurements in magnetic fields up to 10 T and pressures up to 15 kbar. An extremely large drop of the resistivity at the ordering temperature measured for $x \geq x_c$ is explained by a suppression of the spin-fluctuation contribution to the resistivity in the vicinity of T_C due to the intersublattice molecular field. The analysis of the magnetoresistance revealed that the spin fluctuations above T_C are additionally enhanced due to short-range correlations within the f–d-electron system in a limited temperature region above T_C . The application of external pressure results in a suppression of both the spin fluctuations and the onset of the long-range order in the Co sublattice. In $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$ the Co sublattice remains paramagnetic below the Curie temperature for $p \geq 8$ kbar. The magnetic Grüneisen parameter was found to substantially increase as the critical concentration x_c was approached.

1. Introduction

The rare-earth- (R-) based cubic Laves phase compounds with cobalt (RCO_2) attract much attention owing to the d-magnetism instability, which causes various interesting field- and temperature-dependent phenomena. In the compounds with $R = \text{Y}, \text{Lu}$ and Sc the conditions for long-range magnetic order in the itinerant d-electron subsystem are not fulfilled, in contrast to the case for the RFe_2 series. These RCO_2 compounds remain paramagnetic down to the lowest temperatures; however, they show an enhanced temperature-dependent Pauli susceptibility [1, 2]. A first-order phase transition from the paramagnetic into a ferromagnetic state—so-called itinerant metamagnetism [3–5]—occurs in YCo_2 (and LuCo_2) in a magnetic field H_{met} of about 70 T [6]. Due to the strong electron–electron correlations among the electrons in the d subsystem, spin fluctuations are present and influence the physical properties of these compounds substantially [7, 8]. In those RCO_2 compounds where R bears a permanent magnetic moment, a strong intersublattice f–d exchange coupling arises between the itinerant d subsystem (the Co sublattice) and the localized 4f subsystem (the R sublattice). In most of the RCO_2 , the intersublattice molecular field H_{f-d}^{Co} exceeds H_{met} and the Co sublattice undergoes a transition into a ferromagnetic state below T_C . As the exchange constant of the f–d interaction is negative, these compounds are collinear ferrimagnets with heavy R atoms (Gd to Er) and ferromagnets with light rare earths (Pr,



(a)

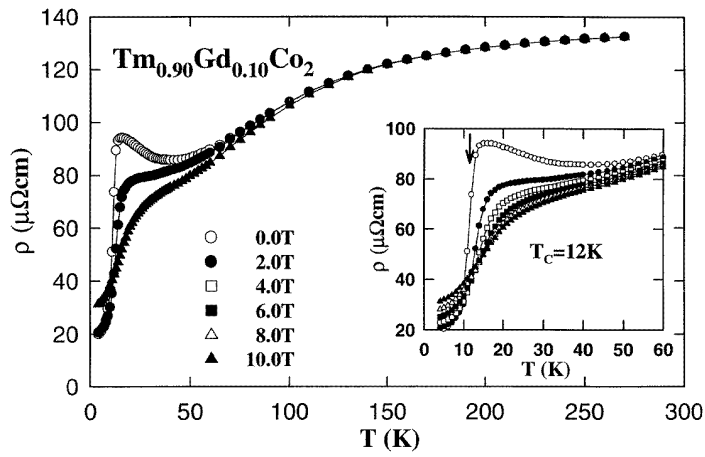


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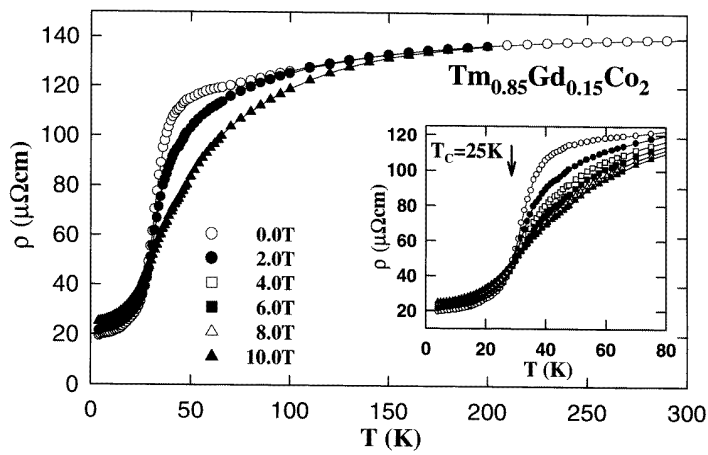
Figure 1. Temperature dependences of the electrical resistivities of the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ compounds at different external magnetic fields. The insets show the details of ρ versus T in the low-temperature region. The Curie temperatures are indicated by arrows. (a) TmCo_2 , (b) $\text{Tm}_{0.95}\text{Gd}_{0.05}\text{Co}_2$, (c) $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$, (d) $\text{Tm}_{0.85}\text{Gd}_{0.15}\text{Co}_2$ and (e) $\text{Tm}_{0.8}\text{Gd}_{0.2}\text{Co}_2$.

Nd and Sm) [9]. However, in TmCo_2 the value of H_{f-d}^{Co} (≈ 60 T) [10] is less than H_{met} and therefore the Co sublattice remains paramagnetic below $T_C = 3.8$ K where only the Tm sublattice is magnetically ordered [11, 12].

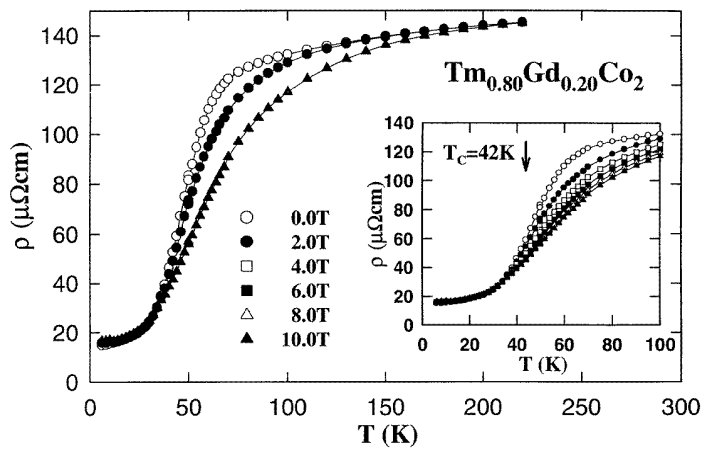
Numerous investigations have dealt with the different features of the d magnetism in the RCO_2 compounds and related substituted systems. Among them the direct observation of metamagnetic transitions in the $\text{R}(\text{Co}_{1-x}\text{M}_x)_2$ systems with $\text{R} = \text{Y}$ or Lu and $\text{M} = \text{Al}$, Ga , Sn or Si and studies of the Co magnetic state as a function of H_{f-d}^{Co} in the compounds $\text{R}_{1-x}\text{Y}_x\text{Co}_2$ should be mentioned [13–17]. The results were mainly discussed within the scope of mean-field theories, and therefore neglected spin-density fluctuations in the



(c)

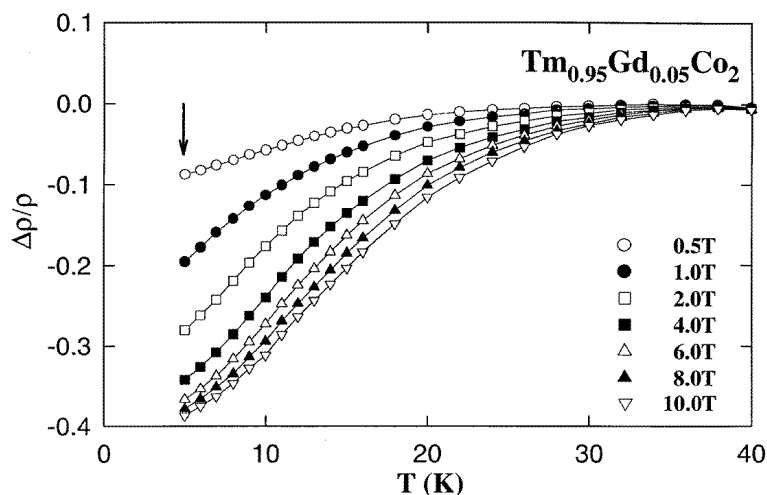


(d)

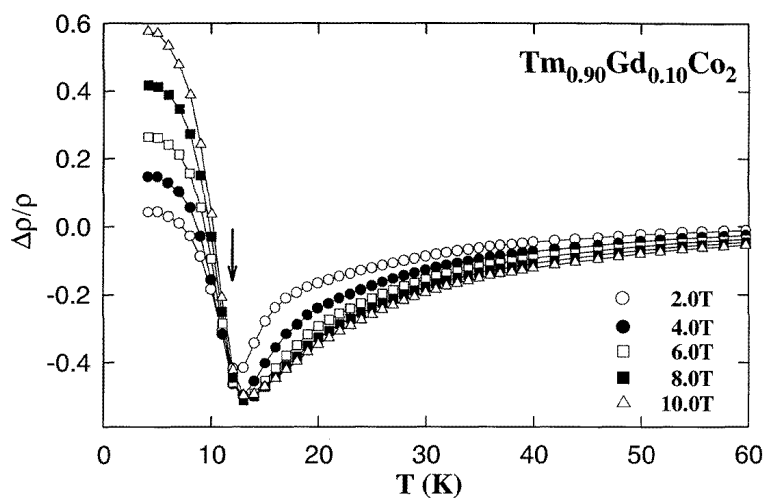


(e)

Figure 1. (Continued)



(a)



(b)

Figure 2. Temperature dependences of the magnetoresistances ($\Delta\rho/\rho$) of the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ compounds in the low-temperature region. (a) $\text{Tm}_{0.95}\text{Gd}_{0.05}\text{Co}_2$, (b) $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$ and (c) $\text{Tm}_{0.85}\text{Gd}_{0.15}\text{Co}_2$. In each panel the arrow indicates the corresponding Curie temperature.

itinerant-electron subsystem. In a more recent analysis, the low-field susceptibility of YCo_2 -based paramagnetic systems (which is dominated by the spin fluctuations) could be related to the high-field metamagnetic transition at elevated temperatures, i.e. the position of the maximum in $\chi(T)$ was found to be proportional to H_{met} [18, 19]. Furthermore, for the magnetic RCo_2 compounds, it could be demonstrated that spin fluctuations influence many of their physical properties above T_C . For instance, the pronounced enhancement of the electrical resistivity observed in the vicinity of T_C has been ascribed to strongly enhanced spin fluctuations in the d-electron subsystem due to the fluctuating f-d molecular field. This enhancement is correlated with the 3d-moment instability which increases as H_{f-d}^{Co}

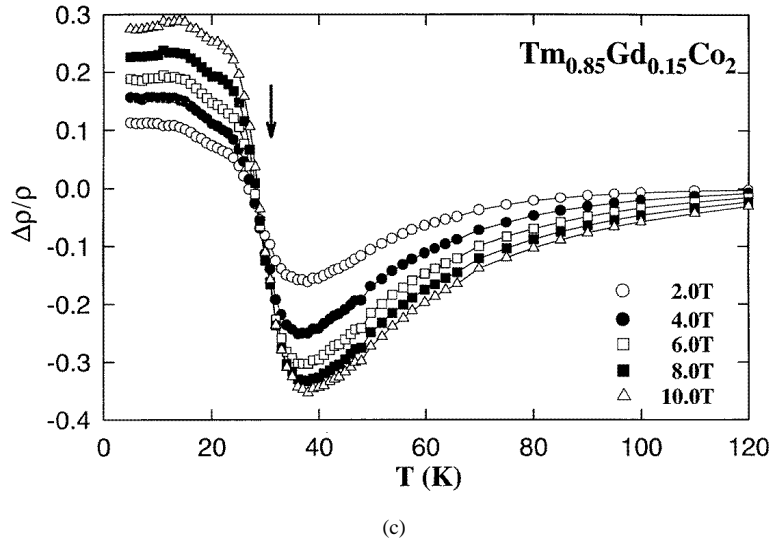


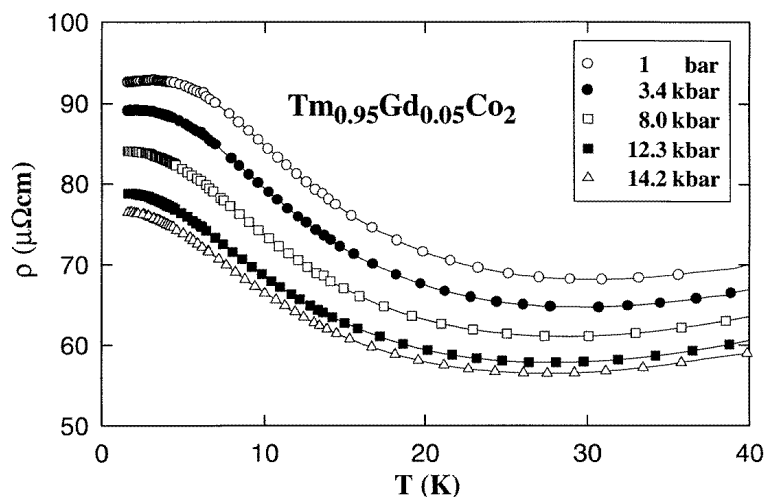
Figure 2. (Continued)

approaches H_{met} [20].

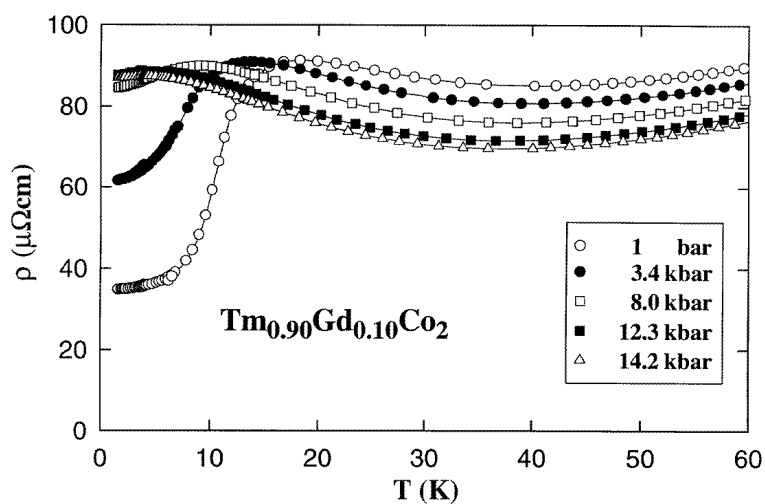
In the present paper we discuss the magnetoresistance and the electrical resistivity, under external pressure, of the pseudobinary $Tm_{1-x}Gd_xCo_2$ Laves phase compounds at around the critical concentration for long-range magnetic order in the Co sublattice. Both the magnetic field and the pressure modify the magnetic state of the itinerant-electron systems considerably. While the magnetic field suppresses the spin fluctuations and increases the magnetic order, the external pressure will broaden the *d* band, due to the decrease of the interatomic distances, and thus weaken the magnetic interactions within the Co 3*d* band. This study continues our previous work devoted to the temperature dependence of the transport properties, thermal expansion and specific heat of the $Tm_{1-x}Gd_xCo_2$ system, in which the Co sublattice becomes magnetic for $x < x_c = 0.1$ [12].

2. Experimental details

Polycrystalline samples of $Tm_{1-x}Gd_xCo_2$ with $x = 0.00, 0.05, 0.10, 0.15$ and 0.20 were induction melted under an argon atmosphere using a stoichiometry of 1:1.95 instead of 1:2 in order to avoid the formation of magnetic RCO_3 as a foreign phase. The samples were annealed at 800 °C for five days; the loss in weight was less than 0.1%. X-ray diffraction patterns give no indication of the presence of foreign phases. The resistivity in a field and under pressure was measured by means of a conventional four-probe technique. The sample dimensions for the magnetoresistance measurements were about $1 \times 1 \times 7 \text{ mm}^3$, while those for the pressure experiment were typically about $0.5 \times 0.5 \times 5 \text{ mm}^3$. Transverse magnetic fields up to 10 T have been applied. A hydrostatic pressure, up to 15 kbar, was generated in a liquid pressure cell using a 1:4 ethanol–methanol mixture as the pressure-transmitting medium. The pressure in the cell was measured using lead as a manometer. Because of the brittleness of these Laves phases the uncertainty in the absolute value of the resistivity is approximately 10%.



(a)

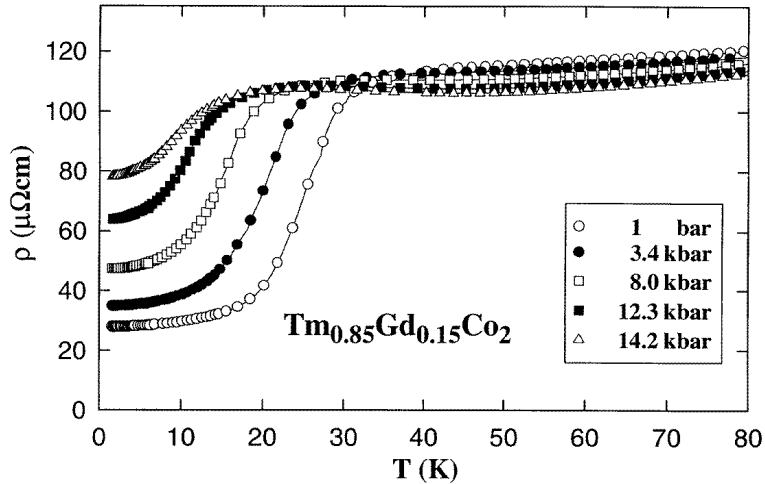


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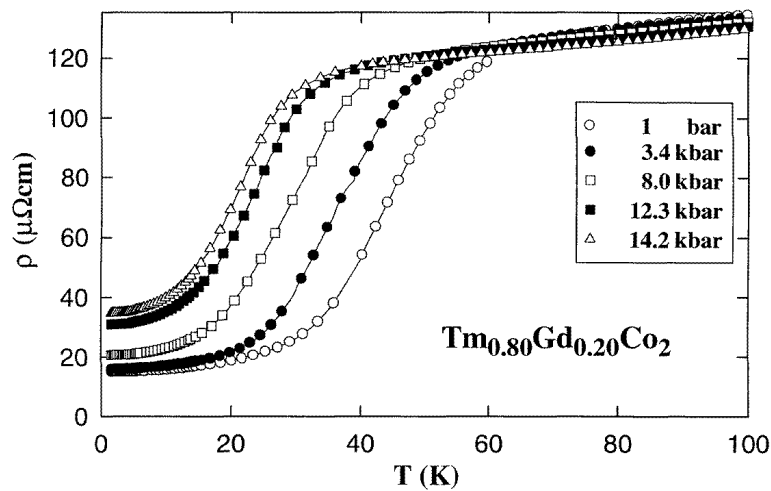
Figure 3. Temperature dependences of the electrical resistivities of $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ compounds measured under different external pressures up to 15 kbar: (a) $\text{Tm}_{0.95}\text{Gd}_{0.05}\text{Co}_2$, (b) $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$, (c) $\text{Tm}_{0.85}\text{Gd}_{0.15}\text{Co}_2$ and (d) $\text{Tm}_{0.8}\text{Gd}_{0.2}\text{Co}_2$.

3. Experimental results

The temperature dependences of the electrical resistivities $\rho(T)$ of the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ compounds in various applied magnetic fields are presented in figures 1(a)–1(e). For all compounds the resistivity saturates at around 150 K as found for YCo_2 [7]. This saturation can most clearly be seen for those Gd concentrations with low T_C (i.e. $0 \leq x \leq 0.1$). The insets show details of the low-temperature behaviour for all of the magnetic field values for which the transverse magnetoresistance has been studied. In each panel the arrow indicates the onset of the long-range magnetic order in zero field. The values for T_C have been



(c)



(d)

Figure 3. (Continued)

obtained from specific heat and thermal expansion measurements and the point of inflection of the ρ versus T curves as discussed in [12]. For low Gd concentrations, $\rho(T)$ shows pronounced minima just above T_C , which vanish gradually with increasing magnetic fields.

Figures 2(a)–2(c) show the magnetoresistances $\Delta\rho/\rho = (\rho(H) - \rho(0))/\rho(0)$ for $x = 0.05$ (below the critical concentration), $x = 0.1$ (the critical concentration) and $x = 0.15$ (above the critical concentration). In each panel the arrow indicates T_C in zero field. In the paramagnetic temperature range the magnetoresistance is negative. For $x = 0.1$ and 0.15 $\Delta\rho/\rho$ shows a minimum above T_C followed by a sharp increase towards positive values on cooling below T_C . This behaviour is more pronounced when the field is increased. However, for $Tm_{0.95}Gd_{0.05}Co_2$ the minimum is not observed because of the low value of T_C .

Figures 3(a)–3(d) display the temperature dependences of the resistivities for the samples with $0.05 \leq x \leq 0.2$ at different pressures. In the paramagnetic range at elevated temperatures (not shown in figures 3) an external pressure of 15 kbar hardly influences the shape of the ρ versus T curves; however, the pressure has a strong influence on $\rho(T)$ in the vicinity of the magnetic transition. Assuming that the point of inflection represents the Curie temperature, it follows that the ordering temperature decreases under applied pressure. Simultaneously the resistivity at the lowest temperature measured increases drastically. The ρ versus T curve for the sample with $x = 0.05$ does not show the characteristic decrease at T_C either at ambient pressure or under pressures up to 15 kbar. However, most interesting is the case of $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$, where this decrease of $\rho(T)$ at the ordering temperature vanishes for pressures higher than about 8 kbar. Above 8 kbar the resistivity of this sample is very similar to that of the sample with $x = 0.05$. Note that, while the maximum in ρ versus T is strongly influenced by the magnetic field, it is hardly changed by pressure.

4. Discussion

A comparison of the resistivity behaviour of the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ system with that of the isostructural R series with non-magnetic partner elements, e.g. $\text{Gd}_{1-x}\text{Y}_x\text{Al}_2$ [21], shows substantial differences in the $\rho(T)$ dependences, which is concerned with the Co subsystem and its contribution to the resistivity. In a previous study of the transport properties of RCo_2 compounds an analysis of the various contributions to the resistivity has been given [20]. For the paramagnetic compounds YCo_2 and LuCo_2 the spin-fluctuation contribution was derived by subtracting the $\rho(T)$ dependence of the isostructural simple intermetallic LuNi_2 or YAl_2 from the total resistivity of these RCo_2 compounds. It was shown that with increasing temperature the spin fluctuations within the d subsystem give rise to an enhanced T^2 -increase at low temperatures, followed by a strong saturation of the $\rho(T)$ dependence at around 150 K. In the magnetic RCo_2 compounds the total magnetic contribution to the resistivity can be decomposed into two parts: one is caused by the scattering of conduction electrons on 4f moments, $\rho_{\text{spd}}(T)$ (neglecting crystal-field effects, ρ_{spd} is considered to be temperature independent above T_C and proportional to the de Gennes factor $(g-1)^2 J(J+1)$), and the other is due to spin fluctuations within the d subsystem, $\rho_{\text{sf}}(T)$. For temperatures far above T_C the latter may be best approximated by the spin-fluctuation contribution to the resistivity in YCo_2 or LuCo_2 . Additionally, in the vicinity of T_C the interaction between the 4f- and d-electron subsystems provides an increase and progressive strengthening of the scattering of the conduction electrons due to the enhanced critical spin fluctuations.

The sharp drop in ρ versus T for $x = 0.1, 0.15$ and 0.2 indicates the corresponding Curie temperatures at 12 K, 25 K and 42 K, respectively. This anomaly in the resistivity is intimately related to the onset of magnetic order in the Co sublattice caused by the f–d exchange interaction for $x \geq 0.1$. Due to this metamagnetic transition within the itinerant sublattice the spin fluctuations are suppressed below T_C , and thus their huge contribution to the resistivity vanishes [7, 20]. The appearance of the minima in $\rho(T)$ above T_C for $x = 0.05$ ($T_C = 4.5$ K) and 0.1 ($T_C = 12$ K) can be understood by assuming that short-range correlations within the 4f sublattice give rise to an enhancement of the d-band spin-fluctuation scattering beyond the ‘normal value’ as e.g. given for YCo_2 at the corresponding temperature. This enhancement and the following suppression are most pronounced if the magnetic order occurs in the temperature region where the value of $\rho(T)$ is still not saturated, i.e. at low temperatures up to about 50 K. Neutron diffraction experiments recently performed on HoCo_2 confirm the existence of enhanced spin fluctuations in a limited temperature region above T_C [24].

According to Boltzmann's transport theory, an external magnetic field acts on the resistivity (magnetoresistance) in a twofold way. In the presence of a magnetic field the conduction electron movement, driven by the electric field, is additionally influenced by the Lorentz force. This gives rise to the so-called *classical magnetoresistance*, which is always positive and limited mainly to the low-temperature region in sample materials with small residual resistivities. The second mechanism is evoked by the influence of the external field on the scattering processes described by the scattering operator in the Boltzmann equation [22] and is related to the field-induced spin arrangement of the magnetic atoms, with the tendency to line up the moments along the field direction. In ferromagnetic materials this second mechanism provides a negative contribution to the total magnetoresistance and is normally much larger than the classical magnetoresistance near to the breakdown of long-range magnetic order [23]. This concept has been used to describe $\Delta\rho/\rho$ for simple ferromagnets, such as RNi_2 and RAI_2 [7]. In the case of ferrimagnetic materials the total effective magnetic field acting on a magnetic sublattice can either increase or decrease when applying an external magnetic field. Therefore, depending on the partial sublattice contributions, the magnetoresistance can show different behaviour as regards its value and sign.

The magnetoresistance of the $Tm_{1-x}Gd_xCo_2$ system does not follow such simple rules and can only be understood when considering the presence of two magnetic sublattices. Above the Curie temperature, $\Delta\rho/\rho$ is negative in this system. The magnetoresistance of the ferromagnetic $Tm_{0.95}Gd_{0.05}Co_2$ compound is negative at any temperature measured in accordance with the above arguments. However, a comparison with the magnetoresistance of simple ferromagnetic compounds (RNi_2 or RAI_2) shows that the negative $\Delta\rho/\rho$ values above T_C are much larger in this compound where spin fluctuations are present, i.e. the resistivity is much more strongly influenced by the external field. This can be understood when we take into account the fact that the applied magnetic field suppresses the short-range correlations within the 4f-electron subsystem near to T_C . As a consequence the enhanced spin-fluctuation scattering of the conduction electrons vanishes.

In the samples with $x \geq 0.1$ (where the induced Co moments are antiparallel to the R moments) the magnetoresistance shows different temperature variation depending on the concentration. With decreasing temperature, $\Delta\rho/\rho$ changes its sign slightly below T_C . The positive values of $\Delta\rho/\rho$ cannot solely be ascribed to the classical magnetoresistance (which is always positive, see above). The $\Delta\rho/\rho$ values are largest at the critical concentration. Note that at 10 T and 4 K for $Tm_{0.9}Gd_{0.1}Co_2$, $\Delta\rho/\rho$ is about twice as large as for $Tm_{0.85}Gd_{0.15}Co_2$ (see the figures 2(b) and 2(c)) and about five times larger for the sample with $x = 0.2$ (not shown here). This extremely strong increase of the positive magnetoresistance can be explained by assuming that the external field reduces the field necessary to induce the full Co moment. Due to the antiparallel orientation of the induced Co moments and the external field H_{ext} the effective magnetic field ($H_{eff} = H_{f-d}^{Co} - H_{ext}$) acting on the Co sublattice (and giving rise to the induced d moments) decreases. Therefore a substantial positive contribution to $\Delta\rho/\rho$ arises with increasing H_{ext} . Hence, external magnetic fields drive the itinerant subsystem closer to the magnetic instability as $H_{f-d}^{Co} \approx H_{met}$ at T_C . For temperatures close to T_C the spin-fluctuation scattering contribution to the total resistivity becomes stronger than it is for ambient conditions.

The magnetic field and the external pressure modify the temperature dependences of the resistivities in quite different ways as seen from the figures 1 and 3. While a magnetic field mainly acts on the exchange-enhanced spin fluctuations close to T_C , pressure influences the spin-fluctuation contribution to the resistivity also at more elevated temperatures. Following the sequence $x = 0.2$ to 0.10 one observes that the ρ versus T

curves become more affected by pressure as x approaches x_c . Of great interest is the measurement for the residual resistivity ρ_0 taken at 1.5 K, which changes drastically. Comparing ρ versus T for $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$ with applied pressures above about 8 kbar and ρ versus T for $\text{Tm}_{0.95}\text{Gd}_{0.05}\text{Co}_2$, it is obvious that there is no longer a magnetic order within the itinerant subsystem in the sample with $x = 0.1$ in this higher-pressure range. Assuming that in $\text{Tm}_{0.9}\text{Gd}_{0.1}\text{Co}_2$ the total molecular field acting on the Co sublattice ($H_{f-d}^{\text{Co}} = 0.9\lambda_{\text{TmCo}}M_{\text{Tm}} + 0.1\lambda_{\text{GdCo}}M_{\text{Gd}}$) is about 97 T [12], one gets for the critical field for demagnetization of the Co sublattice $H'_{\text{met}} = H_{f-d}^{\text{Co}} - H_{\text{met}}(\text{YCo}_2) \approx 27$ T. Hence, the pressure response of H'_{met} , when neglecting the pressure dependence of H_{f-d}^{Co} , can be estimated as $\partial H'_{\text{met}}/\partial P \approx 3.4$ T kbar $^{-1}$, which is comparable with the value of 1 T kbar $^{-1}$ found experimentally for $\text{Lu}(\text{Co},\text{Ga})_2$ [25]. Note that, as H_{f-d}^{Co} will also decrease with increasing pressure, the value 3.4 T kbar $^{-1}$ is the upper limit for $\partial H'_{\text{met}}/\partial P$. For these reasons one can understand the pronounced changes in ρ_0 with pressure, as the itinerant d subsystem may stay at least partly non-magnetic at T_C as $H_{f-d}^{\text{Co}}(P)$ approaches H_{met} , and spin fluctuation will persist even in the ordered state (see the sequence $x = 0.2$ to 0.1 with increasing pressure). Above a certain pressure the Co sublattice will show no long-range magnetic order even when the R sublattice is ordered ($H_{f-d}^{\text{Co}}(P) \leq H_{\text{met}}$ at T_C), and the enhanced residual resistivity becomes a maximum ($x = 0.1$ for maximum applied pressure). Finally, ρ_0 will decrease with further increasing pressure as the spin fluctuations will become progressively suppressed (see figure 3(b) where $\partial\rho_0/\partial P > 0$ and figure 3(a) where $\partial\rho_0/\partial P < 0$).

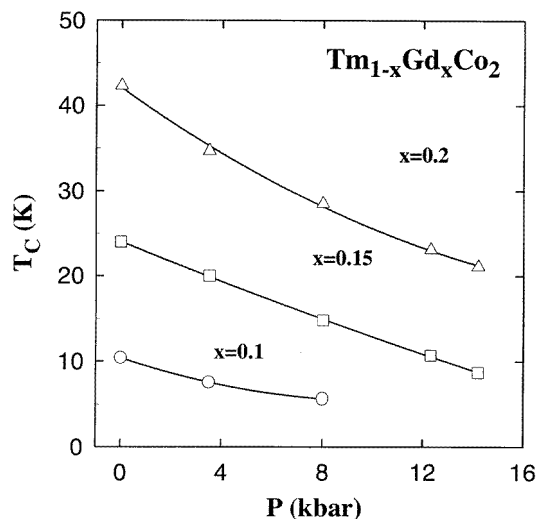


Figure 4. Pressure dependences of the Curie temperatures for the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ system at around the critical concentration $x_c = 0.1$.

Taking the point of inflection (which corresponds to a maximum in $\partial\rho/\partial T$) as an indication of the onset of the magnetic order, one can determine the pressure dependence of T_C . The T_C -values of the $\text{Tm}_{1-x}\text{Gd}_x\text{Co}_2$ compounds decrease almost linearly with increasing pressure as depicted in figure 4. The initial pressure dependences of T_C , $\partial T_C/\partial P$, as deduced from a linear fit to the data in the lower-pressure region, are -0.6 , -1.2 and -2.1 K kbar $^{-1}$ for $x = 0.1$, 0.15 and 0.2, respectively. The normalized values $(1/T_C)\partial T_C/\partial P$ exhibit an essential increase when the critical concentration for the onset of Co magnetism is

approached. The corresponding Grüneisen parameter for the $Tm_{1-x}Gd_xCo_2$ compounds, defined as

$$\Omega_{T_C} = -\frac{\partial \ln T_C}{\partial \ln V} \approx \frac{1}{\kappa_S T_C} \frac{\partial T_C}{\partial P}$$

can be calculated using a value of 0.9 Mbar^{-1} for the adiabatic compressibility κ_S [26]. The values thus obtained are: $\Omega_{T_C} = -61$ (for $x = 0.1$), $\Omega_{T_C} = -54$ (for $x = 0.15$) and $\Omega_{T_C} = -53$ (for $x = 0.2$). Note that for the RCO_2 compounds the values of Ω_{T_C} increase from -5 for $GdCo_2$ towards -27 for $ErCo_2$ [27, 28]. The value for the Grüneisen parameter increases substantially as the itinerant subsystem becomes closer to the magnetic instability. This result represents a further confirmation that the influence of the spin fluctuations on the physical properties is strongest when the Co sublattice is closest to the magnetic instability, when long-range order within the localized subsystem sets in. Such a condition can be achieved by substitution, or by applying a magnetic field or pressure.

5. Conclusion

The influence of both a magnetic field and external pressure on the resistivity has been studied in the $Tm_{1-x}Gd_xCo_2$ cubic Laves phase system near to the critical concentration of Gd ($x_c = 0.1$) for which long-range magnetic order in the itinerant d-electron subsystem below T_C appears. The results can be summarized as follows.

(i) The pronounced curvature in $\rho(T)$ at elevated temperatures in zero field and under ambient pressure is due to the spin-fluctuation scattering of the conduction electrons.

(ii) The substitution of Gd for Tm increases the exchange field acting on the d-electron subsystem and thus increases the Curie temperature from about 4 K for $TmCo_2$ to 42 K for the sample with $x = 0.2$. Only in the concentration range where $x \geq 0.1$ is the Co sublattice magnetically ordered.

(iii) The very large negative magnetoresistance near to T_C in the paramagnetic region is due to an enhanced spin-fluctuation scattering mechanism. This enhancement is attributable to short-range correlations among the 4f moments in combination with a strong f-d interaction. Therefore the resistivity reveals a pronounced increase of $\rho(T)$ above the Curie temperature giving rise to a characteristic minimum. This effect is most pronounced in the vicinity of the critical concentration for the onset of long-range magnetic order in the itinerant sublattice.

(iv) The magnetic field mainly acts on the short-range correlations within the 4f subsystem at around T_C , resulting in a reduction of the enhanced spin-fluctuation contribution to the total resistivity in this temperature range.

(v) With pressure the long-range magnetic order in the itinerant-electron subsystem becomes progressively destabilized. Above 8 kbar the Co sublattice in $Tm_{0.9}Gd_{0.1}Co_2$ is not ordered below T_C due to the pressure-driven increase of the critical field for the metamagnetic transition.

(vi) The magnetic Grüneisen parameter was found to increase as the critical concentration was approached.

(vii) The increase of the residual resistivity in the ordered state with increasing field for $x = 0.1$ and with pressure for $x \geq 0.1$ indicates the destabilization of the itinerant d moments below T_C .

Acknowledgments

This work was supported by the Russian Foundation for Basic Research, project No 96-02-16373-a, and the Austrian Science Foundation FWF, project No P10269.

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