

## Positive magnetoresistivity in a localized-moment ferromagnet with itinerant spin fluctuations: $\text{TmCo}_2$

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Results on electrical resistivity and magnetization of  $\text{TmCo}_2$  intermetallic compound in magnetic fields up to 15 T and in temperature range from 1.5 K to 300 K are presented. In zero magnetic field two well-separated phase transitions are observed at low temperatures: at 3.7 K and at 3.3 K. Magnetoresistivity at temperatures below 3.3 K is negative. However at higher temperatures, magnetoresistivity is positive in a weak field, showing a maximum in the dependence on the magnetic field at  $H_{\text{max}}$ .  $H_{\text{max}}$  is found to be proportional to  $T^{1.5}$ . The maximum exists also in the temperature dependence of magnetoresistivity, measured at a constant external field  $H^*$ . The results are interpreted in the framework of a model with two different contributions to magnetoresistivity: a negative one, related to the localized  $4f$  magnetic moments of Tm, and a positive contribution associated with the features of the Co  $3d$  band which are responsible for the itinerant metamagnetism of  $R\text{Co}_2$  compounds. The positive magnetoresistivity arises due to the spin polarization of the Co  $3d$  band by the exchange field and enhancement of the spin fluctuations within this band. [S0163-1829(99)01425-3]

### I. INTRODUCTION

Electronic transport properties of highly enhanced metallic materials have attracted considerable attention. Main features of resistivity and thermoelectric power in Kondo alloys and heavy fermion compounds have been successfully explained by the scattering of the conduction electrons on magnetic impurities or on  $4f$  moments of rare earth elements. There is, however, another large class of enhanced materials whose transport properties have not been well understood—these are nearly magnetic compounds with itinerant spin fluctuations. In terms of enhancement these materials occupy an intermediate position between the normal metals and the heavy fermion compounds. The group of  $R\text{Co}_2$  Laves phase compounds (where  $R$  stays for rare earth elements and Y, Sc) belongs to this class (see for a review for example Refs. 1–4). Compounds with nonmagnetic elements: Y, Lu, and Sc, are enhanced Pauli paramagnets. They exhibit metamagnetic transition, i.e., a field-induced magnetic phase transition from paramagnetic to ferromagnetic state under an external magnetic field exceeding a critical value  $H_c$ . Magnetic  $R\text{Co}_2$  have two magnetic subsystems with distinctly different properties. The first is the  $4f$  localized magnetic moments of the rare earth elements. The second is formed of the enhanced itinerant Co  $3d$  electrons. Collective spin excitations in this  $d$ -electron system are the spin fluctuations. The  $3d$  electron band and the spin fluctuations are also present in the paramagnetic  $R\text{Co}_2$ . Long-range magnetic order in the itinerant  $d$ -electron subsystem of magnetic  $R\text{Co}_2$  compounds is induced by the exchange field of ordered localized magnetic moments of the rare earth ions. Concerning the transport properties it has been shown that the main contribution to the

resistivity of  $R\text{Co}_2$  comes from the spin fluctuation scattering.<sup>4</sup> At low temperatures the resistivity increases with temperature as  $\rho_{sf} = RT^2$  and shows a pronounced saturation at high temperatures. These features are common for systems with strong electron-electron scattering and have been interpreted within self consistent renormalization (SCR) theory of spin fluctuations by Moriya.<sup>5</sup> However, other important features of the electronic transport in these compounds have not been understood. Among them: (1) Complicated low-temperature behavior of the thermopower; (2) dramatic suppression of the temperature-dependent part of the resistivity in  $R\text{Co}_2$  based alloys; (3) positive magnetoresistivity, recently observed in  $\text{YCo}_2$ .<sup>6,7</sup> The latter observation is especially important, for the SCR theory unambiguously predicts that the spin-fluctuation contribution to the magnetoresistivity of a nearly magnetic metal should be negative.<sup>8</sup> Therefore the discrepancy is of a qualitative character. Positive magnetoresistivity has been found also in paramagnetic and even in ferromagnetic  $\text{Y}(\text{Co}_{1-x}\text{Al}_x)_2$  alloys.<sup>9</sup>

In this paper we present detailed experimental results on the magnetoresistivity of  $\text{TmCo}_2$ , which belongs to the family of  $R\text{Co}_2$  compounds. We will restrict our discussion to the paramagnetic temperature range where the positive magnetoresistivity was observed.  $\text{TmCo}_2$  was chosen because of the following reasons.

Among the magnetic  $R\text{Co}_2$  compounds  $\text{TmCo}_2$  occupies a special place. Recently it was shown that the molecular field acting on  $3d$  subsystem of  $\text{TmCo}_2$  is about 60 T, very close to  $H_c = 67$  T of  $\text{YCo}_2$ .<sup>10</sup> However, thermal expansion results<sup>11</sup> and neutron diffraction data<sup>12</sup> revealed that in contrast to other magnetic  $R\text{Co}_2$  compounds, in  $\text{TmCo}_2$  there is no long-range magnetic order in the itinerant Co  $3d$  sub-

system below  $T_c$  since the intersublattice molecular field acting on this subsystem is less than the critical field for the metamagnetic transition. The effective molecular magnetic field which acts on  $3d$  electrons in a magnetic  $R\text{Co}_2$  can be expressed as  $B_{\text{eff}} = nM + B$ , where  $n$  is the intersublattice exchange coefficient,  $M$ , is the magnetization of the  $4f$  localized moments, and  $B = M + \mu_0 H$ ,  $\mu_0 H$  is the external field. The exchange coefficient  $n$  of  $\text{TmCo}_2$  was estimated to be about  $-13 \text{ T f.u.}/\mu_B$  ( $n \approx -50$  in SI units),<sup>10</sup> whereas  $M \approx 6 \mu_B/\text{f.u.}$  at 4.2 K in field of about 4 T. Therefore, a comparatively weak external magnetic field induces much larger molecular effective magnetic field acting on Co  $3d$  subsystem. Moreover, since the exchange field  $B_{\text{ex}} = nM$  is not a real magnetic field, it does not induce classical, Lorentz-force-driven magnetoresistivity which arises due to an orbital motion of the conduction electrons in magnetic field. This permits us to separate the contribution of the Lorentz-force-driven magnetoresistivity to the total magnetoresistivity. Important also is that the spin disorder contribution to electrical resistivity coming from the scattering on magnetic excitations within Tm moments, is comparatively small in  $\text{TmCo}_2$ .<sup>4</sup>

Throughout this paper the sign of  $B_{\text{eff}}$  is not important, therefore, to avoid a confusion, from now on we will assume that  $n$  is positive and express  $B_{\text{eff}}$  as  $B_{\text{eff}} = |nM - B|$ .

## II. EXPERIMENTAL PROCEDURES

A sample of  $\text{TmCo}_2$  was prepared from pure components by melting in an induction furnace under a protective Ar atmosphere and was subsequently annealed in vacuum at 1100 K for about one week. The x-ray analysis showed no traces of impurity phases. A four-probe dc method was used for electrical resistivity measurements. Resistivity was measured with a longitudinal orientation of electrical current with respect to the magnetic field. The size of the sample was about  $1 \times 1 \times 10 \text{ mm}^3$ .

The precision of the determination of the absolute value of resistivity in  $R\text{Co}_2$  compounds is rather low, mainly due to an uncertainty in sample geometry which is closely related to the mechanical quality of the sample. We have found that the density of polycrystalline samples of the high purity  $R\text{Co}_2$  compounds, prepared by induction or arc melting, is generally lower than the density of less pure ones. Therefore the apparent resistivity of the higher purity samples is higher owing to a smaller effective cross-section. To a some extent the uncertainty of the determination of the absolute resistivity value can be reduced by introducing a correction on the density of the sample. The estimated error in the absolute value of electrical resistivity with such corrections is about  $\pm 20\%$ . However, reproducibility of the resistivity results for the same sample is much better, and uncertainty is about  $\pm 0.5\%$ .

Magnetization was measured by a SQUID magnetometer on a sample from the same ingot as that used for the resistivity measurements. The sample weight was 2.3 mg.

## III. EXPERIMENTAL RESULTS AND DISCUSSION

Temperature dependences of the resistivity of  $\text{TmCo}_2$  in zero magnetic field and in the field of 15 T are shown in Fig.

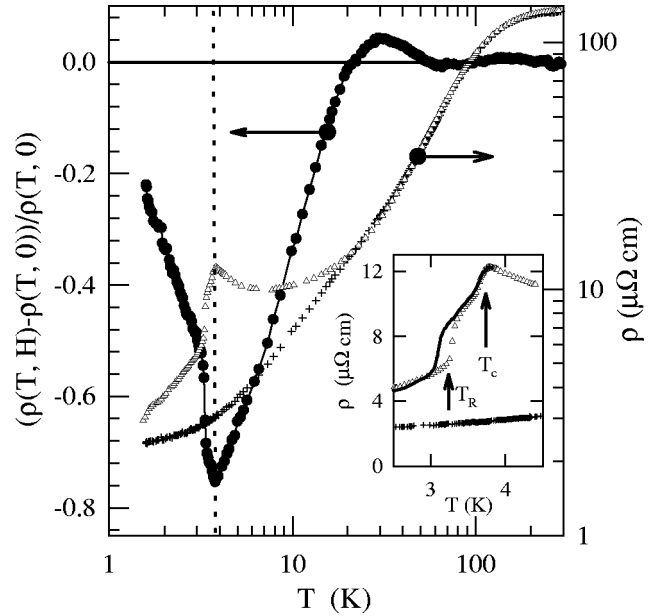


FIG. 1. Temperature dependency of the resistivity of  $\text{TmCo}_2$ .  $\Delta$ : zero field, heating;  $+$ : in field of 15 T; solid line (inset): cooling in zero field;  $\bullet$ : magnetoresistivity,  $[\rho(T, H) - \rho(T, 0)]/\rho(T, 0)$ . Vertical broken line indicates Curie temperature. The inset shows the resistivity in a vicinity of the ordering temperature.

1. It is known from specific heat, resistivity, and susceptibility measurements that in high-quality  $\text{TmCo}_2$  samples there exist two transitions: at 3.8 K and at 3.4 K.<sup>4,11,13</sup> In contrast, in the less pure material (characterized by a larger residual resistivity), only one transition was found.<sup>11,14</sup> The higher temperature transition was associated with Curie temperature of the Tm localized moments. Below the second transition temperature a ferromagnetic spiral structure along the  $[111]$  direction was observed.<sup>12</sup> In agreement with published results,<sup>4,11</sup> we have found two well-separated phase transitions, marked as  $T_c$  and  $T_R$  in the inset of Fig. 1. The presence of two phase transitions indicates that the sample is of a high quality. Transition temperatures, as determined from the maxima of  $d\rho/dT$  are  $T_c = 3.7 \text{ K}$ ,  $T_R = 3.3 \text{ K}$  (the latter was obtained on heating). Resistivity at  $T_R$  shows a clear hysteresis with the width of about 0.2 K, whereas at  $T_c$  no hysteresis (within the temperature resolution of our measurements—better than 0.05 K) was detected. Magnetoresistivity  $[\rho(T, H) - \rho(T, 0)]/\rho(T, 0)$  at  $\mu_0 H = 15 \text{ T}$  is negative below 20 K and amounts at its minimum at 4 K to almost 80%. Qualitatively, this negative magnetoresistivity is in agreement with theoretical results for ferromagnetic metals.<sup>15</sup> But details of the magnetic field and temperature dependences of the magnetoresistivity are different from those predicted by theory.<sup>15</sup> The most striking result is that the magnetoresistivity measured at a temperature  $T^*$  above  $T_R$  is positive in an intermediate range of magnetic fields, having a maximum at a magnetic field  $H_m$ , which increases with  $T^*$ , see Fig. 2,  $H_m \approx K_h \cdot T^{*1.5}$ . The maximum is not observable below  $T_R$ , it appears just above this temperature in a very small magnetic field. Note that the magnetoresistivity is positive even in a very close vicinity to the Curie temperature, where a small external field induces a long range order in the  $4f$  local moment system, which should result in a strong decrease of resistivity. The magnitude of

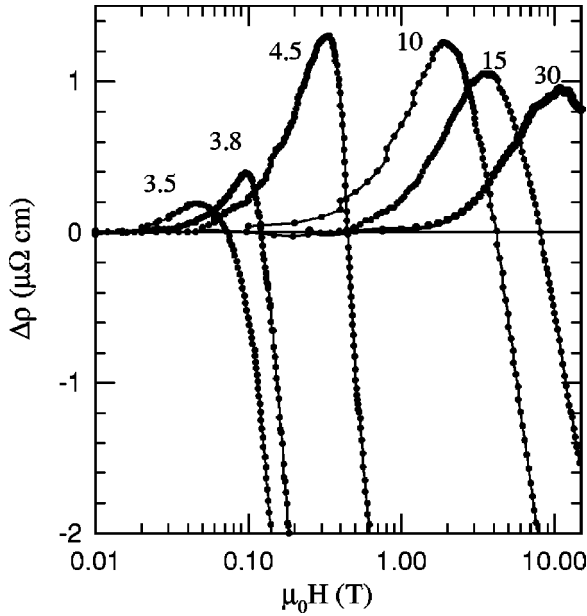


FIG. 2. Magnetoconductivity  $\Delta\rho = \rho(H, T^*) - \rho(0, T^*)$  of TmCo<sub>2</sub> against external magnetic field, measured at different temperatures  $T^*$  from 3.5 K to 30 K. The temperature is indicated by the numbers near to every curve.

the positive magnetoconductivity at its maximum [ $\Delta\rho(H_m)$ ] increases with increasing temperature and reaches the maximum value at around 5 K. At higher temperatures  $\Delta\rho(H_m)$  slowly decreases.

A maximum is also present in the temperature dependence of magnetoconductivity, as it can be seen in Fig. 3. In these measurements the external magnetic field,  $H^*$ , was kept constant. However, as the temperature increases, magnetization of the Tm subsystem, induced by the constant ex-

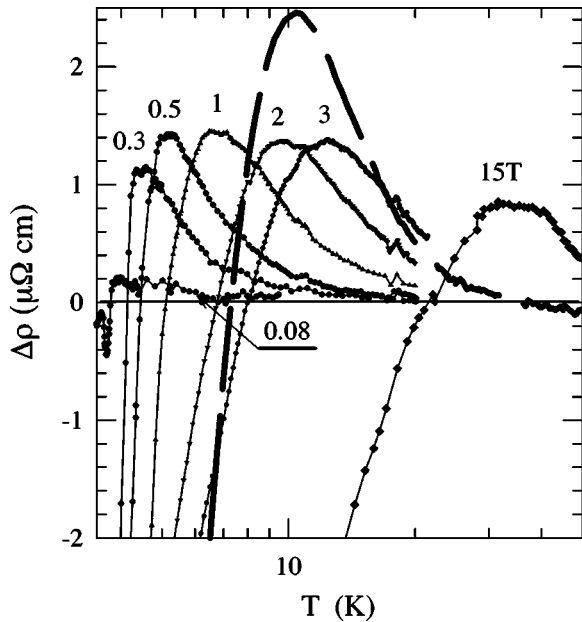


FIG. 3. Magnetoconductivity  $\Delta\rho = \rho(H^*, T) - \rho(0, T)$  of TmCo<sub>2</sub> against temperature  $T$ , measured in magnetic fields  $H^*$  from 0.08 T to 15 T. The field strength is indicated by the numbers near to each curve. The magnetoconductivity of a less pure sample of TmCo<sub>2</sub>, measured at  $H^* = 1.9$  T, is shown by the broken line.

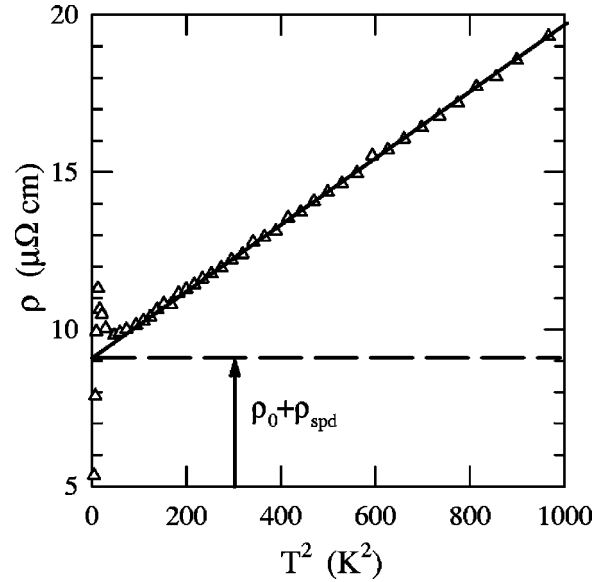


FIG. 4. The resistivity of TmCo<sub>2</sub> at low temperatures, against  $T^2$ , measured in zero magnetic field. The triangles represent the experimental points; the line is obtained by the least squares fit of the function  $\rho = \rho_0 + \rho_{spd} + RT^2$  to the experimental resistivity.

ternal magnetic field, decreases. Therefore both the exchange field acting on the 3d itinerant subsystem  $B_{ex} = nM$  and the magnetic field induced in the sample  $B = \mu_0 H + M$  decrease. This variation of the internal effective magnetic fields should result in a temperature dependence of magnetoconductivity. Moreover, magnetoconductivity may have an intrinsic temperature dependence. The observed temperature variation of the magnetoconductivity is a result of a combination of these two effects. The temperature  $T_{max}$  at which the magnetoconductivity reaches a maximum increases with  $H^*$  showing a similar relation as that between  $H_{max}$  and  $T^*$  in case of the field dependencies:  $K_t T_{max}^{1.5} \approx H^*$ . A strong resemblance between the field dependence and the temperature dependence of magnetoconductivity (see Figs. 2 and 3) implies that the variation of the magnetization of 4f subsystem plays the major role in the magnetoconductivity behavior.

#### A. Evaluation of different contributions to the resistivity

The total resistivity of TmCo<sub>2</sub> includes several contributions. For further analysis of the experimental results it is useful to have an estimate of the magnitude of different contributions. Assuming that the Matthiessen rule is valid we write the resistivity of TmCo<sub>2</sub> as

$$\rho(T) = \rho_0 + \rho_{ph} + \rho_{sf} + \rho_{spd}, \quad (1)$$

where  $\rho_0$  is the impurity resistivity,  $\rho_{ph}$  is the phonon contribution,  $\rho_{sf}$  is the spin fluctuation resistivity due to the scattering of the conduction electrons on spin fluctuations within the Co 3d band, and  $\rho_{spd}$  is spin disorder contribution arising from the scattering on Tm 4f moments.  $\rho_{spd}$  is independent of temperature above Curie temperature  $T_c$ , whereas  $\rho_{spd} = aT^2$  below  $T_c$ . Phonon resistivity  $\rho_{ph}$  at low temperatures varies as  $\rho_{ph} = bT^5$ . It has been shown, that in RCo<sub>2</sub> compounds  $\rho_{ph} < \rho_{sf}$  at all temperatures in the range from 0 K to 1000 K.<sup>4</sup> Therefore in the further analysis we will disregard this contribution. With these assumptions we

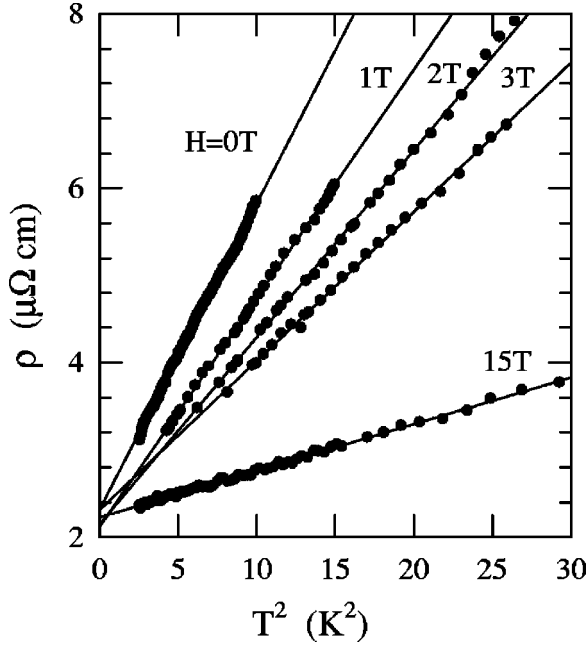


FIG. 5. Temperature dependency of the resistivity of TmCo<sub>2</sub> at low temperatures (below Curie temperature), measured in zero magnetic field and in the magnetic fields from 1 to 15 T. The lines are obtained by the least squares fit of the function  $\rho = \rho_0 + aT^2 + RT^2$  to the experimental resistivity.

estimate the sum  $\rho_0 + \rho_{spd}$  by extrapolating the total resistivity from the temperature region  $T > T_c$  where  $\rho_{sf} = RT^2$  and  $\rho_{spd} = \text{const}(T)$  hold, to  $T = 0$ ; see Fig. 4. The extrapolation yields  $9.2 \mu\Omega \text{ cm}$  for  $\rho_0 + \rho_{spd}$ . The resistivity above Curie temperature shows purely  $T^2$  variation in a broad temperature range from about 5 K to at least 30 K with the gradient  $R = 0.011 \mu\Omega \text{ cm K}^{-2}$ . This indicates that the temperature dependent part of the resistivity in this temperature range is dominated by the spin fluctuation scattering.

Below  $T_c$  there are two temperature dependent contributions:  $\rho_{sf}$  and  $\rho_{spd}$ , both, according to theory should vary as  $T^2$ . The experimental resistivity does follow this law, see Fig. 5. The extrapolation of the  $\rho(T) = RT^2 + aT^2 + \rho_0$  to  $T = 0$ , yields  $\rho_0 \cong 2.2 \mu\Omega \text{ cm}$ , independent, within the limits of the experimental uncertainty, of the magnetic field. As the result, for the  $\rho_{spd}$  at temperatures above  $T_c$  we get about  $7 \mu\Omega \text{ cm}$ . The spin fluctuation resistivity  $\rho_{sf}$  is very small at  $T_c$ , however, since  $\rho_{sf}$  increases with the temperature, already at  $T > 26 \text{ K}$  it exceeds  $\rho_{spd}$ . Therefore in the paramagnetic temperature range the main contributions to the total resistivity are  $\rho_{sf}$  and  $\rho_{spd}$ :  $\rho(T, H) = \rho_{sf}(T, H) + \rho_{spd}(H)$ .

### B. Field dependence of magnetoresistivity

The existence of a maximum in the field dependence of magnetoresistivity implies that there are at least two competing contributions of opposite sign and with different field dependencies. To understand an origin of these contributions one has to take into account the specific features of the electronic band structure of RCo<sub>2</sub> compounds. From the band structure calculations<sup>16,17</sup> it is known that the Co 3d states give a major contribution to the total density of states (DOS) at the Fermi energy  $N(\epsilon_F)$ , so that  $N_d(\epsilon_F) \gg N_s(\epsilon_F)$ . There-

fore the well known  $s$ - $d$  scattering model of Mott has been used for interpretation of the transport properties.<sup>4,18</sup> It is assumed within this model that  $s$  electrons carry the bulk of the current. Impurities, phonons, disordered local moments, and spin fluctuations cause scattering of the  $s$  electrons into vacant  $s$  or  $d$  states, so that  $\rho_{sf} = \rho_{sf}^{s-s} + \rho_{sf}^{s-d}$  and  $\rho_{spd} = \rho_{spd}^{s-s} + \rho_{spd}^{s-d}$ . Since the scattering probability is proportional to the density of states into which the electrons are scattered,<sup>19</sup> the  $s$ - $d$  scattering occurs much more frequently than the  $s$ - $s$  scattering. As the result of this,  $\rho_{spd} \cong \rho_{spd}^{s-d} \sim N_d(\epsilon_F)$ . The spin fluctuation scattering is in essence the electron-electron scattering of the conduction  $s$  electrons on 3d electrons. Since the number of 3d electrons available for the scattering is proportional to  $N_d(\epsilon_F)$ , the spin fluctuation resistivity  $\rho_{sf} \cong \rho_{sf}^{s-d} \sim N_d^2(\epsilon_F)$ .

External magnetic field can affect the electrical resistivity in a number of ways.

First, the internal field  $B = M + \mu_0 H$  can give rise to the classical, Lorentz-force-driven positive magnetoresistivity  $\Delta\rho_L$ . Both the transversal and the longitudinal Lorentz-force-driven magnetoresistivity  $\Delta\rho_L \sim B^2$  in a weak field. The transversal magnetoresistivity is saturated in the limit of high fields in the case of a metal with closed Fermi surface, longitudinal magnetoresistivity is saturated always.<sup>20</sup>

Secondly, the external field suppresses a disorder within the localized magnetic moments, giving rise to an uniform magnetization  $M$  and to a negative magnetoresistivity  $\Delta\rho_{spd}$ . It has been shown that for a system of localized spins  $S$ , the spin disorder resistivity  $\rho_{spd}$  can be expressed as:<sup>21-23</sup>

$$\rho_{spd} = \rho_\infty \left( 1 - \frac{\langle S \rangle^2}{S(S+1)} \right), \quad (2)$$

where  $\rho_\infty$  is a constant, proportional to  $S(S+1)$ . In the paramagnetic temperature range, if external field is zero,  $\langle S \rangle = 0$ , and  $\rho_{spd} = \rho_\infty$ . External magnetic field induces the uniform magnetization  $M = \langle S \rangle$  and the magnetoresistivity

$$\Delta\rho_{spd} = \rho_{spd}(H) - \rho_{spd}(0) = -\rho_\infty \frac{\langle S \rangle^2}{S(S+1)} = -CM^2 \quad (3)$$

(where  $C$  is a temperature independent constant).

And thirdly, the external field causes a spin polarization of the 3d band via effective molecular field  $B_{\text{eff}} = |nM - B|$ , where intersublattice exchange constant  $n \gg 1$ . This polarization can change the total 3d-DOS  $N_d(\epsilon_F)$  and the magnitude of the itinerant spin fluctuations. It has been proposed, that external magnetic field suppresses spin fluctuations and leads to a decrease of  $N_d(\epsilon_F)$ , and to a negative magnetoresistivity.<sup>8,5,24</sup> However there are strong arguments that this conclusion is not valid for the itinerant metamagnets, like RCo<sub>2</sub> compounds. It has been shown that the metamagnetism of an itinerant electron system is intimately connected with specific features of DOS near the Fermi energy, namely, the DOS energy dependence should have a positive curvature.<sup>2,25,26</sup> In this case the splitting of DOS in the effective field results in an increase of  $N_d(\epsilon_F)$ , as illustrated by Fig. 6. The increase of DOS under application of a magnetic field is the necessary condition for the existence of the metamagnetic transition. Since the magnetic field brings the system closer to the ferromagnetic instability, the magnitude of

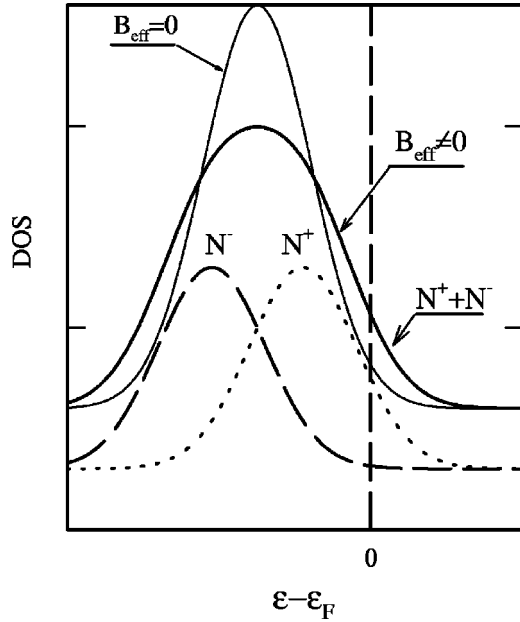


FIG. 6. DOS of  $\text{TmCo}_2$  near to Fermi energy and the effect of magnetic field on  $N_d(\epsilon_F)$  (schematically). The splitting of the  $3d$  DOS due to a polarization in magnetic field results in an increase of the total DOS at  $\epsilon = \epsilon_F$ .

spin fluctuations should also increase. In agreement with this scenario of the increase of  $N_d(\epsilon_F)$  and of the magnitude of the spin fluctuations, it has been experimentally found that the paramagnetic susceptibility of itinerant-electron metamagnets increases under external magnetic field.<sup>2</sup>

Because of the increase of  $N_d(\epsilon_F)$  due to the polarization of the  $3d$  band by a magnetic field acting on this system, the  $s-d$  scattering contributions to resistivity should also increase, giving rise to a positive magnetoresistivity  $\Delta\rho_{s-d}$ . The previous discussion implies that this should be a common feature of itinerant-electron metamagnets.

$\Delta\rho_{s-d}$  includes, in principle, contributions from all the scattering processes which can cause  $s-d$  transitions. In particular, in the case of high-purity  $\text{TmCo}_2$  these are the itinerant spin fluctuation scattering and the localized spin disorder scattering. As we discussed above, disorder in the Tm localized moment subsystem is suppressed by the external field, therefore  $s-d$  scattering, induced by the localized moments, also decreases. In contrast, we expect that the magnitude of the itinerant spin fluctuations increases in the field. Therefore, the increase of  $N_d(\epsilon_F)$  due to the polarization of the  $3d$  band in the effective field  $B_{\text{eff}}$  affects most strongly the spin fluctuation contribution to the resistivity, giving rise to a positive  $\Delta\rho_{sf}$ . However, the impurity scattering and the scattering on the disordered localized magnetic moments can give a substantial contribution to  $\Delta\rho_{s-d}$  too. Indeed, the experimental magnetoresistivity of a  $\text{TmCo}_2$  sample with a larger residual resistivity ( $\rho_0 = 20 \mu\Omega \text{ cm}$ ) exceeds considerably the magnetoresistivity of the high purity  $\text{TmCo}_2$ ; see Fig. 3.

Both  $\Delta\rho_L$  and  $\Delta\rho_{s-d}$  are positive, but there is an important difference between them: the  $s-d$  scattering contribution to the magnetoresistivity is a function of the effective molecular field  $B_{\text{eff}}$ :  $\Delta\rho_{s-d} = f(|nM - B|)$ ; whereas the Lorentz force magnetoresistivity  $\Delta\rho_L$  arises due to an orbital

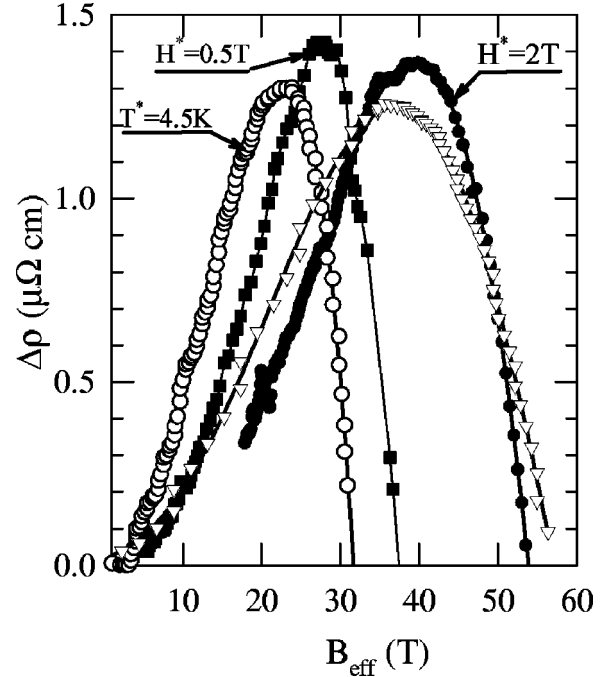


FIG. 7. Magnetoresistivity in dependence on effective molecular field  $B_{\text{eff}} = nM - B$ . The dependence of the magnetoresistivity on magnetic field  $\Delta\rho(H, T^*)$  is shown by the open symbols for  $T^* = 4.5 \text{ K}$  and  $T^* = 10 \text{ K}$ . The filled symbols display the temperature dependencies of the magnetoresistivity  $\Delta\rho(H^*, T)$ , measured at  $H^* = 0.5 \text{ T}$  and  $H^* = 2 \text{ T}$ .

motion of the conduction electrons in magnetic field  $B = \mu_0 H + M$ , and therefore  $\Delta\rho_L = f_L(B)$ .

Summing up, we expect that in  $\text{TmCo}_2$  three mechanisms may give the most important contributions to magnetoresistivity: the negative magnetoresistivity due to the suppression of disorder within the localized Tm-moment subsystem  $\Delta\rho_{spd} = -CM^2$ ; the positive, Lorentz-force-driven magnetoresistivity  $\Delta\rho_L = f_L(B)$ ; and, the positive magnetoresistivity  $\Delta\rho_{s-d}$  arising due to the polarization of the Co  $3d$  band and enhancement of the itinerant spin fluctuations  $\Delta\rho_{s-d} = f(|nM - B|)$ .

To estimate the relative importance of  $\Delta\rho_L$  and  $\Delta\rho_{s-d}$  for the observed dependencies of  $\Delta\rho$  on magnetic field and on temperature, we note, that the variation of the magnetic field  $B$  is very different in the measurements of the magnetoresistivity dependence on magnetic field at a constant temperature as compared with the measurements of the temperature dependence of the magnetoresistivity at a fixed external magnetic field. In the former experiment the temperature is fixed and the internal magnetic field varies from zero to a maximum value  $B_{\text{max}} = \mu_0 H_{\text{max}} + M(H_{\text{max}}, T)$ . In the latter, however, the external magnetic field is fixed and the temperature is varied from a  $T_{\text{max}}$  to a  $T_{\text{min}}$ . In this case  $B$  varies at most from  $B_{\text{min}} \approx \mu_0 H$  (high temperature limit) to  $B_{\text{max}} = \mu_0 H + M(H, T_{\text{min}})$ ,  $M$  being  $\ll \mu_0 H$  in the paramagnetic temperature region, i.e., variation of  $B$  is in this case much smaller than in the field dependence measurements. However, the magnitude of the experimental magnetoresistivity variation is the same in these two experiments, as it can be seen in Figs. 2 and 3. Therefore, only the variation of  $B$  in the temperature dependent measurements cannot cause the magne-

toresistivity temperature dependence. This variation could be explained by an intrinsic temperature variation of the magnetoresistivity, but this is in contradiction to the experimental results for  $\text{YCo}_2$ , where the magnetoresistivity is nearly independent of the temperature in the temperature range from about 2 K to at least 30 K. On the other hand, the effective molecular field  $B_{\text{eff}} = |nM - B|$  has in both these experiments almost the same variation (note,  $n \approx 50$ , when  $M$  is expressed in tesla). Magnetoresistivity, when plotted versus  $B_{\text{eff}}$  reveals nearly the same dependence in both the field dependent measurements and the temperature dependent measurements; see Fig. 7. Therefore the driving force for the positive magnetoresistivity is not  $B$ , but  $B_{\text{eff}}$ , whose main contribution is the exchange field  $nM$ . Since the exchange field is actually not magnetic field, it cannot induce the Lorentz-force magnetoresistivity and we conclude that the existence of maxima in both the magnetic field and the temperature dependences of the magnetoresistivity indicates that the effect of  $\Delta\rho_L$  on the nonmonotonic behavior of the magnetoresistivity should be small. In the further analysis we will not take  $\Delta\rho_L$  into account.

Concerning  $\Delta\rho_{s-d}(B_{\text{eff}})$  there is no theory which can give an actual form of the dependence. Therefore we suggest this dependence based only on the experimental facts.

(1) The magnetoresistivity of  $\text{TmCo}_2$  is positive at small field and negative at larger fields. The negative contribution  $\Delta\rho_{spd} \sim B^2$  according to theory. Therefore the positive contribution should have a weaker field dependence.

(2) The magnetoresistivity of the paramagnetic  $\text{YCo}_2$  (where Co 3d subsystem is essentially the same as that in  $\text{TmCo}_2$ , but in  $\text{YCo}_2$  there are no localized moments) was found to be positive.<sup>7</sup> The best fit of the magnetoresistivity of  $\text{YCo}_2$  is given by  $\Delta\rho = b_1 H + b_2 H^2$ , both the linear and  $H^2$  terms being positive. The result implies that the  $H^2$  contribution is due to the classical, Lorentz-force-driven magnetoresistivity. The positive  $H^2$  term cannot result in the non-monotonic variation of  $\Delta\rho(H)$  in  $\text{TmCo}_2$ , furthermore, in  $\text{YCo}_2$ , it is about 5 times smaller than the linear term. Therefore, we assume for the time being, that  $\Delta\rho_{s-d}$  is a nearly linear function of  $B_{\text{eff}}$ :  $\Delta\rho_{s-d} \sim B_{\text{eff}}^\alpha$ , and represent the total magnetoresistivity as

$$\begin{aligned} \Delta\rho(T, H) &= \rho(T, H) - \rho(T, 0) \\ &= \Delta\rho_{s-d} + \Delta\rho_{spd} = AB_{\text{eff}}^\alpha - CM^2. \end{aligned} \quad (4)$$

When  $B \ll nM$  and  $n(dM/dH) \gg 1$ , we can use a simplified expression for  $B_{\text{eff}}$ :  $B_{\text{eff}} = nM$ . These conditions are not satisfied in high magnetic field when  $M$  approaches to saturation magnetization  $M_s$ ; or at high temperatures. We will consider such a case later.

There are two special points in the dependence of  $\Delta\rho(H)$  on the field: the field  $H_{\text{max}}$  at which the magnetoresistivity reaches a maximum value  $\Delta\rho(H_{\text{max}}) \equiv \Delta\rho_{\text{max}}$ ; and the field  $H_0$ , where  $\Delta\rho$  crosses the zero line,  $\Delta\rho(H_0) = 0$ . The position of the maximum of  $\Delta\rho(H)$  can be found from the condition

$$\frac{d\Delta\rho}{dH} = \frac{dM}{dH} (An\alpha M^{\alpha-1} - 2CM) = 0. \quad (5)$$

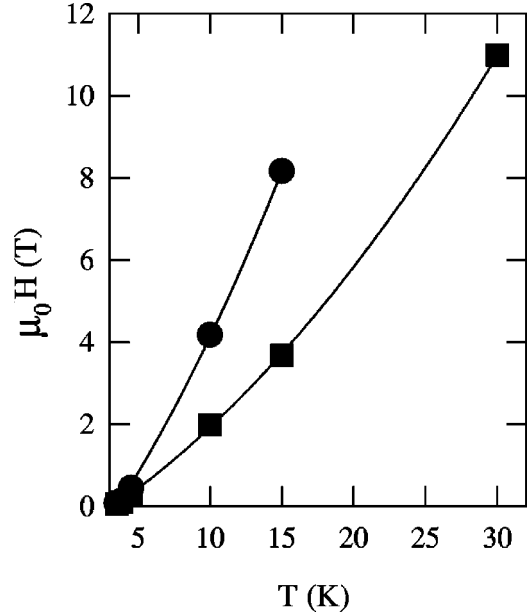


FIG. 8. The plot of the position of the magnetoresistivity maxima and zeros on the H-T plane in the linear scale. ■: the maxima of the dependence of  $\Delta\rho$  on the magnetic field at a constant temperature:  $H_{\text{max}}(T^*)$ ; ●: the zeros of  $\Delta\rho(H)$  at a constant temperature:  $H_0(T^*)$ .

In the paramagnetic range,  $dM/dH \neq 0$ , therefore from Eq. (5) follows

$$M_{\text{max}} = \left( \frac{An\alpha}{2C} \right)^{1/(2-\alpha)}. \quad (6)$$

Magnetoresistivity  $\Delta\rho = 0$  at  $H = 0$  and at

$$M_0 = \left( \frac{An}{C} \right)^{1/(2-\alpha)}. \quad (7)$$

Note that both  $M_{\text{max}}$  and  $M_0$  depend neither on the magnetic field nor on the temperature if we assume that both  $A$  and  $C$  are temperature independent constants. This assumption seems to be well justified in case of  $\Delta\rho_{spd}$  (the constant  $C$ ). The negative magnetoresistivity  $\Delta\rho_{spd}$  arises due to suppression of a disorder within the localized Tm moments. Since in the paramagnetic temperature region  $\rho_{spd}$  is independent of temperature, its decrease in an external field depends only on the induced magnetization  $M$ . However the coefficient  $A$  can be in general temperature dependent. Our guess on its temperature independence is based on the experimental fact<sup>7</sup> that the magnetoresistivity of  $\text{YCo}_2$  is almost independent of the temperature in the range from 2 K to about 40 K.

To compare theoretical results (6) and (7) with the experimental data we need to know the dependence of the magnetization on magnetic field and on temperature. In the paramagnetic temperature range, when  $T \gg T_c$ , the susceptibility  $\chi$  of  $R\text{Co}_2$  compounds was found to follow, in a first approximation, to Curie-Weiss law.<sup>5,27-29</sup> Therefore, when the magnetization  $M$  is considerably smaller than the saturation magnetization  $M_s$  so that  $M \propto \chi \cdot H$ , it can be approximately expressed as  $M = DH/(T - T_c)$ . This results in

$$H_{\text{max}} = \left( \frac{An\alpha}{2C} \right)^{1/(2-\alpha)} D^{-1} (T^* - T_c), \quad (8)$$

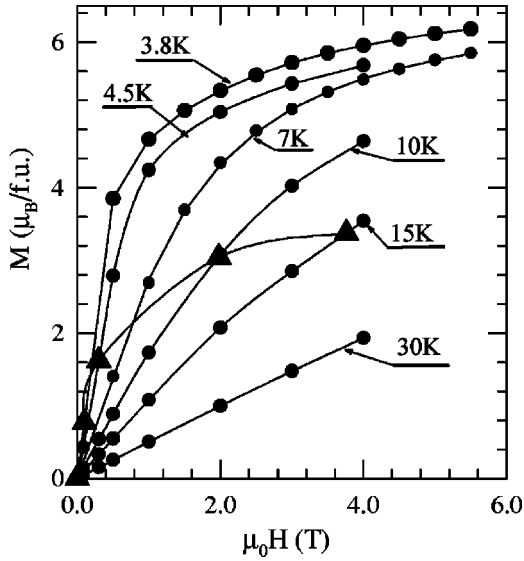


FIG. 9. The dependence of magnetization on the external magnetic field.  $\bullet$ : experimental results at temperatures from 3.8 K to 30 K. The temperatures are indicated by numbers near to each curve. Solid triangles show the position of  $M_{\max}$ . The lines are guides for the eye.

$$H_0 = \left( \frac{An}{C} \right)^{1/(2-\alpha)} D^{-1} (T^* - T_c), \quad (9)$$

where  $T^*$  is the temperature at which the field dependence of the magnetoresistivity is measured. Equations (8) and (9) show that both  $H_{\max}$  and  $H_0$  linearly increase with temperature, however with different gradients. This is in rough agreement with the experimental results, however the experimental temperature dependencies  $H_{\max}(T^*)$  and  $H_0(T^*)$ , shown in Fig. 8 are not linear.

The theoretical ratio of the gradients of the  $H_{\max}(T^*)$  and  $H_0(T^*)$  dependencies,  $K = K_0/K_m$ , is equal (in the approximation of the linear relation between  $M$  and  $H$ )  $K = (2/\alpha)^{1/(2-\alpha)}$ . It depends only on value of  $\alpha$ . The experi-

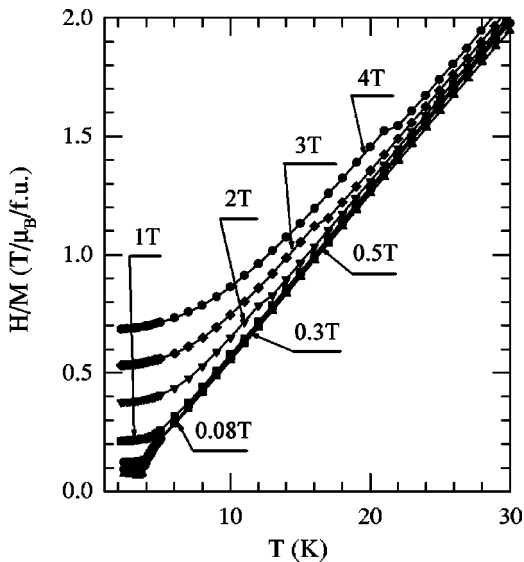


FIG. 10. Temperature dependence of the inverse susceptibility  $1/\chi = H/M$  at different external magnetic fields from 0.08 T to 4 T.

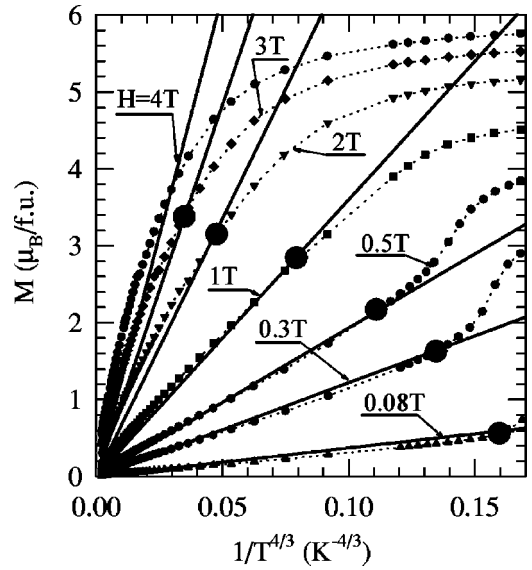


FIG. 11. The dependence of magnetization on  $1/T^{4/3}$  at different external magnetic fields from 0.08 T to 4 T. Small points with the broken lines as the guides for the eye represent the experimental results. Solid lines show the function  $M = 36 (H^{0.9}/T^{4/3})$ . Big solid circles:  $M_{\max}$ .

mental value of  $K$  is about 2.2. The nearest integer solution of the equation  $(2/\alpha)^{1/(2-\alpha)} = 2.2$  is  $\alpha = 1$  (this is the exact solution for  $K = 2$ ) in agreement with the experimental results for  $\text{YCo}_2$ , as it was discussed above. The difference between the experimental  $K \approx 2.2$  and the  $K = 2$  corresponding to  $\alpha = 1$  originates from the fact that in the region of  $M_{\max}$  and  $M_0$ , the magnetization is not precisely a linear function of the external field as it can be seen in Fig. 9, where the magnetization of  $\text{TmCo}_2$  is displayed in dependence on the applied magnetic field. Knowing the  $H_{\max}$  at a given temperature, it is easy to find also the experimental values of  $M_{\max}$  from the  $M(H)$  dependence, measured at the same temperature. These  $M_{\max}$  are shown in that figure too. As it can be seen from Fig. 9,  $M_{\max}$  falls in the region where  $M(H)$  has an appreciable curvature. Somewhat better approximation for that region would be  $M \sim H^i$  with  $i < 1$ . Evaluation of  $i$  from relation  $2^{1/i} = 2.2$ , gives  $1/i \approx 1.1$  ( $i = 0.9$ ). This also accounts for, in a part, the nonlinearity of the  $H_{\max}(T^*)$  and  $H_0(T^*)$  dependencies. Another, and more important contribution to the nonlinearity of the  $H_{\max}(T^*)$  and  $H_0(T^*)$  dependencies relates to the observation that, although at low magnetic fields  $\chi$  closely follows to Curie-Weiss dependence, see Fig. 10; there are considerable deviations from this dependence at higher magnetic fields in the temperature range, in which the maximum of the magnetoresistivity is observed. Therefore, in the case of the present problem, the Curie-Weiss law is not a sufficiently good approximation for  $\chi$ . As it is well known, a more precise theoretical result for the susceptibility of a ferromagnetic metal is<sup>29</sup>  $\chi = D/T^{4/3} - T_c^{4/3}$ . In Fig. 11 we show the magnetization of  $\text{TmCo}_2$  in dependence on  $T^{-4/3}$ . At low magnetic fields the experimental data display almost  $T^{-4/3}$  dependence on the temperature, however again there is a noticeable deviation at high fields. But, what is important for the present consideration, the expression  $M = DH^{0.9}/T^{4/3}$  accurately pre-

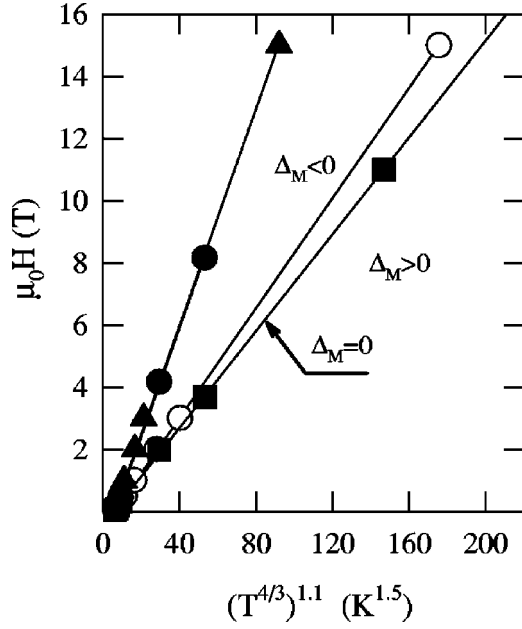


FIG. 12. The plot of the position of the magnetoresistivity maxima and zeros on the  $H - (T^{4/3})^{1.1}$  plane. ■: the maxima of the dependence of  $\Delta\rho$  on the magnetic field at a constant temperature,  $H_{\max}(T^*)$ ; ○: the maxima of the dependence of  $\Delta\rho$  on the temperature at a constant magnetic field,  $T_{\max}(H^*)$ ; ▲: the magnetic field  $H_0(T^*)$  at which  $\Delta\rho(H, T^*) = 0$ ; and ●: the temperature  $T_0(H^*, T)$  at which  $\Delta\rho(H^*, T) = 0$ .

dicts the values of  $M_{\max}$  with a single value of parameter  $D$ , equal to  $36\mu_B/\text{f.u.}T^{-0.9}K^{4/3}$ . Therefore a more accurate empirical expression for  $H_{\max}$  is

$$H_{\max} = \left[ \left( \frac{An}{2C} \right) \frac{T^{*4/3}}{D} \right]^{1.1}. \quad (10)$$

In agreement with this formula, the experimental  $H_{\max}$  and  $H_0$  reveal a linear variation when plotted against  $(T^{*4/3})^{1.1}$ ; see Fig. 12 (note, the product  $4/3 \times 1.1 = 1.47 \approx 1.5$ ). We would like to emphasize, that we do not claim that the magnetization of the paramagnetic  $\text{TmCo}_2$  can be expressed as  $M = DH^{0.9}/T^{4/3}$  in a broad range of temperatures and magnetic fields. As we mentioned, the experimental  $\chi$  closely follows to Curie-Weiss dependence at low magnetic field, see Fig. 10, it however deviates from this dependence at higher magnetic fields. We have found that, because of these deviations, the dependence of  $M_{\max}$  on the magnetic field and on temperature cannot be described by the Curie-like law:  $M_{\max} = DH_{\max}/T^*$ , rather it is well approximated by the modified expression:  $M_{\max} = DH_{\max}^{0.9}/T^{*4/3}$ , from which the relation (10) follows.

Another discrepancy between the theory and the experiment relates to  $M_{\max}$ . According to the previous discussion,  $M_{\max}$  should be a constant, which, in a first approximation depends neither on magnetic field nor on temperature. Figure 9 however clearly shows that  $M_{\max}$  depends strongly on temperature. This result has important implications. The simplest explanation of this strong dependence would be a temperature dependence of coefficient  $A$  in the expression for magnetoresistivity. However, as we already discussed, the experimental results on the magnetoresistivity of  $\text{YCo}_2$  suggest

that  $A$  is almost independent of temperature. In our later discussion we will show that the present results indicate that  $A$  does have a temperature dependence, slowly decreasing with the increasing temperature. However the temperature dependence of  $A$  is too weak, moreover, it has the wrong sign, to explain the variation of  $M_{\max}$ . We think that the explanation of the strong dependence of  $M_{\max}$  on temperature lies in the different time scales and the symmetry of resistivity and magnetization experiments. The resistivity is a very ‘‘fast’’ experiment with the characteristic time scale of order of  $10^{-10} - 10^{-14}$  sec. Therefore even comparatively high-frequency fluctuations of the magnetization of the  $4f$  Tm moments are seen by the conduction electrons as regions of a static magnetization. We understand here as the fluctuating magnetization the long-range magnetic correlations of Tm moments. Where ‘‘long-range’’ means that the spatial extension of the ordered region (correlation length) is much larger than the mean free path of conduction electrons. The fluctuating magnetization of the  $4f$  Tm moments induces the fluctuating molecular field, acting on the Co  $3d$  electrons. On the other hand, the magnetization was measured in the essentially static experiment. Additionally, the magnetoresistivity, in contrast to magnetization, is an even function of the magnetic field in the sense that it does not change the sign if the direction of the magnetic field is reversed. The time- and the space-average of the fluctuating part of the magnetization in the magnetization measurement is equal to zero and gives no contribution to the measured magnetization. In the resistivity analysis however, one has to take into account the total magnetization, which is a sum of the static magnetization  $M^s$  and of the fluctuating magnetization  $M^f$ :  $M = M^s + M^f$ . The magnitude and spatial extension of the fluctuating component of the magnetization should increase as temperature approaches to the Curie temperature. Now the sum  $(M^s + M^f)_{\max}$  should be constant, therefore, the static part  $M_{\max}^s$  decreases near to the Curie temperature. The fluctuating magnetization exists also when no external magnetic field is applied. We suppose that the sharp increase of the zero field resistivity of  $\text{TmCo}_2$  near the Curie point arises due to the positive magnetoresistivity in this fluctuating molecular magnetic field. Similar mechanism should be effective also for other  $R\text{Co}_2$  compounds. We will turn to this subject later, in discussion of the temperature dependence of the magnetoresistivity.

### C. Temperature dependences of the magnetoresistivity

The temperature dependence of the magnetoresistivity, measured at a constant external magnetic field  $H^*$ ,  $\Delta\rho_T$ , reveals also a nonmonotonic variation. We will show that the mechanism of this nonmonotonic behavior is essentially the same as in the case of the field dependencies of the magnetoresistivity,  $\Delta\rho_H$ , measured at a constant temperature.

The magnetoresistivity is defined as follows:

$$\Delta\rho = \rho_{spd}(M(H, T)) - \rho_{spd}(0) + \rho_{s-d}(M(H, T), T) - \rho_{s-d}(0, T). \quad (11)$$

From Eq. (11) the conditions for the maximum follow:

$$\frac{d\Delta\rho_H}{dH} = \frac{dM}{dH} \Delta_M = 0, \quad (12)$$



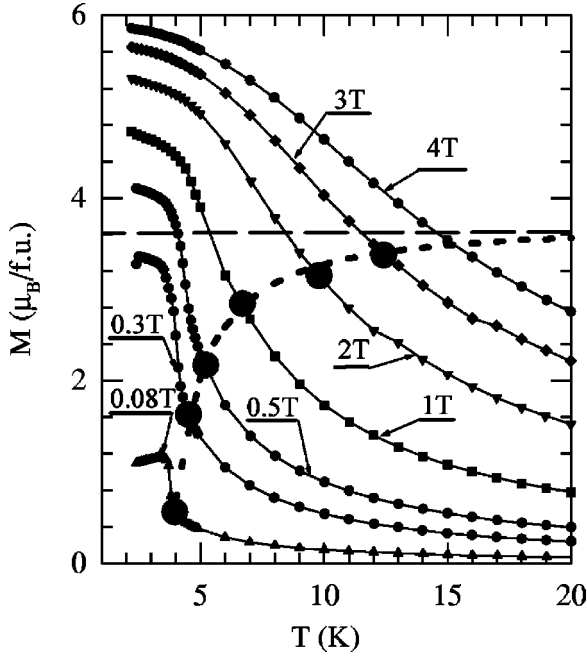


FIG. 13. The temperature dependence of magnetization in different external magnetic fields. The big solid circles indicate  $M_{\max}^s$ . At high temperatures  $M_{\max}^s$  saturates and approaches a constant value, shown by the horizontal broken line.

with

$$\Delta_M \equiv \frac{\partial \rho_{spd}(M(H, T^*))}{\partial M} + \frac{\partial \rho_{s-d}(M(H, T^*), T^*)}{\partial M},$$

and

$$\frac{d\Delta\rho_T}{dT} = \frac{dM}{dT} \Delta_M + \Delta_T = 0, \quad (13)$$

where

$$\Delta_T \equiv \frac{\partial \rho_{s-d}(M(H^*, T), T)}{\partial T} - \frac{\partial \rho_{s-d}(0, T)}{\partial T} = \frac{\partial \Delta\rho_{s-d}}{\partial T}.$$

It is clear from Eqs. (12) and (13) that, if  $\Delta_T \neq 0$ , the position of the maximum ( $H_{\max}, T^*$ ) of the  $\Delta\rho_H(H)$  is not coincide with the position of the maximum ( $H^*, T_{\max}$ ) of  $\Delta\rho_T(T)$ . Experimental results reveal that there is a small but systematic difference between the line of ( $H^*, T_{\max}$ ) and the line of ( $H_{\max}, T^*$ ). The points ( $H^*, T_{\max}$ ) sit in the region where  $\Delta_M < 0$ . Since  $dM/dT < 0$ ,  $\Delta_T < 0$  around the line of the maxima on  $H-T$  plane, Fig. 12. In other words, the positive magnetoresistivity  $\Delta\rho_{s-d}$  decreases with the temperature. The magnitude of  $\Delta_T$  can be estimated from the  $\Delta\rho_T$ . The estimation yields  $\Delta_T \approx -0.01 \mu\Omega \text{ cm K}^{-1}$  at  $T = 34 \text{ K}$ ; and  $\Delta_T \approx -0.02 \mu\Omega \text{ cm K}^{-1}$  at  $T = 10 \text{ K}$ . The total decrease of  $\Delta\rho_{s-d}(B_{\text{eff}} = \text{const})$  in the temperature range from 5 K to 30 K does not exceed  $0.4 \mu\Omega \text{ cm}$ , this is about 30% of  $\Delta\rho_{\max}$ . Therefore the coefficient  $A$  in Eq. (4) is not actually temperature independent, however, because of comparatively small value of  $\Delta_T$ , the temperature dependence of  $A$  cannot account for the observed variation of  $\Delta\rho(H^*, T)$ . This variation is mainly the result of the temperature dependence of magnetization  $M$  of Tm localized moments and of the effective field  $B_{\text{eff}} = |nM - B|$ .

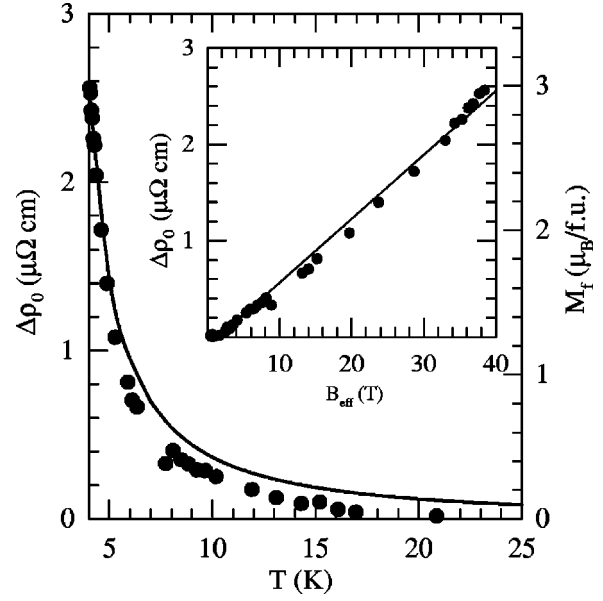


FIG. 14. The temperature dependence of the fluctuating magnetization  $M^f$  (solid line) and of the "zero field magnetoresistivity" (dots). The inset shows the "zero field magnetoresistivity" against the fluctuating effective molecular field. The solid line in the inset is a linear fit to the experimental data.

The decrease of  $\Delta\rho_{\max}$  with temperature or with magnetic field is also partly a consequence of the negative sign of  $\Delta_T$ . But this is not the only mechanism which can cause the decrease  $\Delta\rho_{\max}$  at high temperatures and it certainly cannot explain the sharp decrease of  $\Delta\rho_{\max}$  at low temperatures. At low temperatures we need to take into account the fluctuating part of the magnetization, as it was discussed above. The total magnetoresistivity  $\Delta\rho$  includes two contributions: first, the "zero field magnetoresistivity"  $\Delta\rho^0$  due to the fluctuating magnetization in absence of an external field; and secondly, the static magnetoresistivity  $\Delta\rho^m$  which relates to the static magnetization, induced by the external magnetic field. It is  $\Delta\rho^m$  that we measure experimentally. According to the present model, we have for the magnetoresistivity:  $\Delta\rho = An(M^s + M^f) - C(M^s + M^f)^2$ . The experimentally measured  $\Delta\rho^m$  is equal:

$$\Delta\rho^m = \Delta\rho - \Delta\rho^0 = M^s(An - CM^s - 2CM^f), \quad (14)$$

where  $\Delta\rho^0 = AnM^f - C(M^f)^2$ . It follows from Eq. (14) that  $\Delta\rho^m(H)$  has the maximum at  $M^s + M^f = An/2C$ . The magnitude of  $\Delta\rho^m$  at the maximum is equal:

$$\Delta\rho_{\max}^m = \frac{A^2 n^2}{4C} \left(1 - M_{\max}^f \frac{2C}{An}\right)^2. \quad (15)$$

We assume here that in a first approximation  $M^f$  is dependent only on the temperature but not on the external field. The temperature variation of  $M_{\max}^f$  can be estimated from the temperature variation of  $M_{\max}^s$ ; see Fig. 13. The static magnetization  $M_{\max}^s$  rapidly vanishes, as temperature nears the Curie temperature, and approaches the constant value  $An/2C$  at high temperatures, where  $M^f$  should vanish. The fluctuating part of the magnetization can be obtained as  $M_{\max}^f = An/2C - M_{\max}^s$ . The result is depicted in Fig. 14 by the solid line.

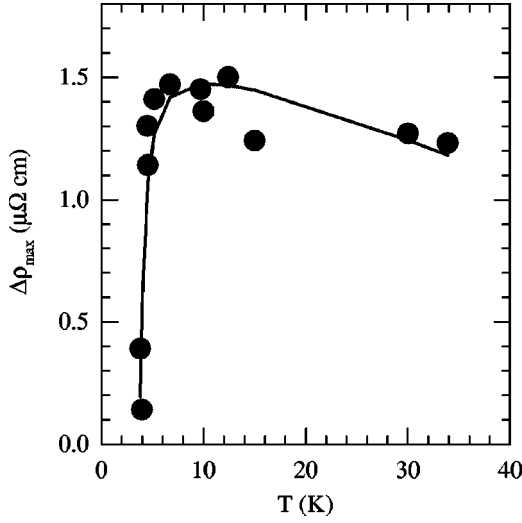


FIG. 15. The magnitude of the magnetoresistivity at its maximum. The solid line shows the fitted function (17). The data points on this plot are the experimental  $\Delta\rho_{\max}$ , corrected on the decrease of the positive contribution of the magnetoresistivity with increasing temperature due to the negative  $\partial\Delta\rho_{s-d}/\partial T$ . The correction was done by addition of  $\langle\partial\Delta\rho_{s-d}/\partial T\rangle(T-5)$  to the experimental  $\Delta\rho_{\max}$  values. The  $\langle\partial\Delta\rho_{s-d}/\partial T\rangle$  is the mean value of  $\partial\Delta\rho_{s-d}/\partial T$  in the temperature interval from 5 K to  $T$ .  $T$  stays for  $T_{\max}$  or  $T^*$  depending on whether  $\Delta\rho_{\max}$  was obtained in the measurement at a constant magnetic field or at a constant temperature.

Figure 14 shows also the positive “zero field magnetoresistivity”  $\Delta\rho_0$ . It was obtained by the following procedure: (1) The  $RT^2$  dependence was fitted to the experimental zero field resistivity of  $\text{TmCo}_2$  in temperature range from 15 K to about 40 K and extrapolated to the low-temperature region; see Fig. (4). (2). This dependence was subtracted from the experimental zero field resistivity of  $\text{TmCo}_2$ . In the inset to Fig. 14,  $\Delta\rho_0$  is plotted against the effective molecular field  $B_{\text{eff}}=nM^f$ . The resistivity increases almost linearly with the effective magnetic field with the rate of  $0.07 \mu\Omega \text{ cm}/T$ . The value is comparable with  $0.09 \mu\Omega \text{ cm}/T$  found for the magnetoresistivity of paramagnetic  $\text{YCo}_2$ .<sup>7</sup> We therefore conclude that the upturn of the zero-field resistivity of  $\text{TmCo}_2$  near to Curie temperature is in essence the positive magnetoresistivity, which arises due to the polarization of the  $3d$  band by the fluctuating molecular magnetic field. Similar behavior of the resistivity of  $\text{ErCo}_2$  and an enhancement of the resistivity of  $\text{HoCo}_2$  and, probably of  $\text{DyCo}_2$ ,<sup>4</sup> can be explained by this mechanism.

At high temperatures, or at high magnetic fields the condition  $Mn \gg H$  cannot be satisfied and for the effective field, which acts on Co  $3d$  subsystem we should use  $|nM - B|$  instead of  $nM$ . In this temperature region we disregard  $M^f$  since it decreases with increasing temperature rapidly. Then  $\Delta\rho^m = A(nM^s - B) - C(M^s)^2$ . Using again for the magnetization,  $M^s = DH^{0.9}/T^{4/3}$ , we can obtain as the approximate solution

$$\Delta\rho_{\max}^m \approx \frac{A^2 n^2}{4C} \left(1 - \frac{T_{\max}^{4/3}}{Dn}\right)^2. \quad (16)$$

Combining the results of Eqs. (15) and (16), the overall temperature variation of  $\Delta\rho_{\max}^m$  can be represented as

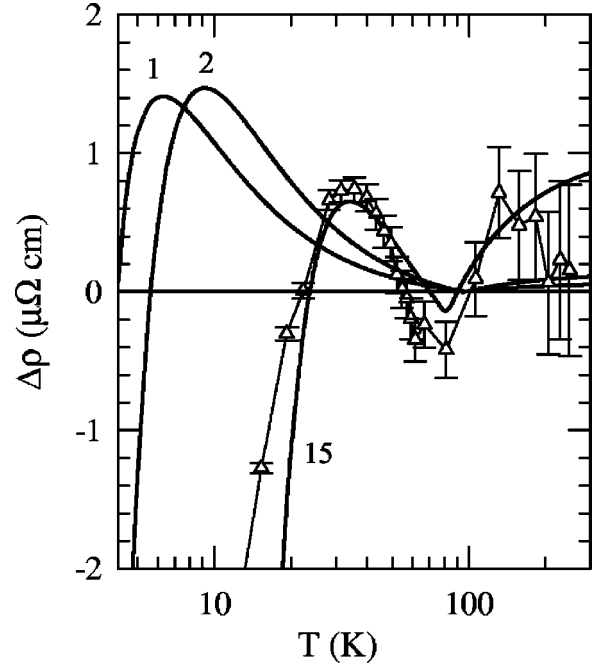


FIG. 16. Temperature dependence of the magnetoresistivity at magnetic fields of 1 T, 2 T, and 15 T. The solid lines show the magnetoresistivity calculated according to expression (18). The triangles with the error bars represent the experimental data for  $\mu_0 H = 15$  T.

$$\Delta\rho_{\max}^m(T) \approx \frac{A^2 n^2}{4C} \left(1 - M_{\max}^f \frac{2C}{An}\right)^2 \left(1 - \frac{T_{\max}^{4/3}}{Dn}\right)^2. \quad (17)$$

Since  $An/2C = M_{\max}$ , which can be estimated to be  $3.7 \mu_B/\text{f.u.}$  (see Fig. 13), we represent  $A^2 n^2/4C = C(An/2C)^2 = C3.7^2$ . Further, we use for  $M_{\max}^f$  an analytical expression:  $M_{\max}^f = An/2C[1 - (T - T_c)/(m_0 + T - T_c)]$  which fits well to the experimentally obtained values, as explained above. This leaves us with three free parameters in the function (17):  $C$ ,  $m_0$ , and  $Dn$ . The experimental  $\Delta\rho_{\max}^m$ , and the function (17) fitted to these data, are shown in Fig. 15.

The following values were obtained for the free parameters from the fit:  $m_0 = 0.2$  K,  $C = 0.12 \pm 0.01 \mu\Omega \text{ cm}(\text{f.u.}/\mu_B)^2$ ,  $Dn = 700 \pm 380 \text{ K}^{4/3}$ . In fact, from these three parameters truly free is only  $m_0$ , the other two can be obtained from independent experimental data. The intersublattice exchange coefficient  $n$  was found to be  $\approx 13 T \text{ f.u.}/\mu_B^{10}$ , whereas  $D$  was estimated from the slope of  $M(T^{-4/3})$  dependence (Fig. 11):  $D \approx 36 \mu_B/\text{f.u. K}^{4/3}/T$ . This yields  $Dn \approx 470 \text{ K}^{4/3}$  in good agreement with the value obtained from the fitting. Coefficient  $C$  can be found from the relation  $An/2C = 3.7 \mu_B/\text{f.u.}$ , if  $A$  is known.  $A$  can be estimated from the field dependence of the magnetoresistivity of  $\text{YCo}_2$  (estimation yields about  $0.09 \mu\Omega \text{ cm}/T$ ), or from the “zero field magnetoresistivity”  $\Delta\rho_0$  of  $\text{TmCo}_2$  (Fig. 14) (about  $0.07 \mu\Omega \text{ cm}/T$ ). With these values of  $A$ ,  $C$  is equal  $0.16 \mu\Omega \text{ cm}(\text{f.u.}/\mu_B)^2$ ; or  $0.13 \mu\Omega \text{ cm}(\text{f.u.}/\mu_B)^2$ , respectively. The agreement with the value of  $0.12 \mu\Omega \text{ cm}(\text{f.u.}/\mu_B)^2$  is rather reasonable.

Summarizing the results of the discussion and making somewhat problematic assumption that  $M^s(T, H)$  and  $M^f(T)$

can be expressed similar to  $M_{\max}^s$  and  $M_{\max}^f$ , we can write down the temperature and the field dependencies of the magnetoresistivity  $\Delta\rho^m$  as

$$\Delta\rho^m = A|nM^s - B| - (CM^s + 2CM^f)M^s$$

or

$$\Delta\rho^m = A \left| n \frac{DH^{0.9}}{T^{4/3}} - B \right| - \left( C \frac{DH^{0.9}}{T^{4/3}} + 2CM^f \right) \frac{DH^{0.9}}{T^{4/3}}. \quad (18)$$

Figure 16 presents the calculated magnetoresistivity as given by formula (18) with the parameters defined above. The theoretical  $\Delta\rho^m(T, H)$  dependencies demonstrate good agreement with the experimental results giving further confirmation that the present model of the magnetoresistivity is basically correct. A feature emerges: on these  $\Delta\rho^m(T, H^*)$  dependencies there is a minimum at high temperatures—about 100 K. The minimum appears at the temperature where internal magnetic field  $B$  just compensates the exchange field  $nM$ :  $nM - B = 0$ . The existence of this feature in the magnetoresistivity opens a new method to determine the intersublattice exchange coefficient  $n$  (for the systems with the antiferromagnetic coupling between sublattices) from the measurements of magnetoresistivity and magnetization. If the position of the minimum ( $T_{com}, H^*$ ) is determined from the magnetoresistivity results, the intersublattice exchange coefficient can be found as  $n = B/M(T_{com}, H^*)$ . Our experimental  $\Delta\rho^m$  measured at highest available in the present measurements magnetic field of 15 T, confirms the existence of the minimum. Unfortunately, the resolution of the resistivity measurements at high temperatures is not good enough to allow a detailed comparison between theoretical and experimental results at these temperatures for lower magnetic fields.

#### IV. CONCLUSION

The electrical resistivity and the magnetization of high quality sample of TmCo<sub>2</sub> compound were measured from 2 K to 300 K in longitudinal magnetic field from 0 T to 15 T.

Nonmonotonic variation of the magnetoresistivity with magnetic field and with temperature has been observed at temperatures from  $T_c$  to about 50 K. Contrary to theoretical predictions for systems with localized magnetic moments

and for spin fluctuation materials, the magnetoresistivity is positive in weak magnetic fields, having a maximum in dependence on magnetic field at  $H_{\max}(T^*)$  or, in dependence on temperature at  $T_{\max}(H^*)$ . There is an universal relation between  $H_{\max}$  and  $T^*$  or between  $T_{\max}$  and  $H^*$ :  $H \sim T^{1.5}$ . This behavior of the magnetoresistivity is explained on the basis of a model which assumes that there are two different contributions to the magnetoresistivity: the positive magnetoresistivity,  $\Delta\rho_{s-d} \sim H$ ; and the negative  $\Delta\rho_{spd} \sim H^2$ . The latter originates from the suppression by the magnetic field of a disorder in the system of localized magnetic moments of Tm. The positive contribution to the magnetoresistivity is found to be proportional to the exchange field acting on the 3d Co electrons. This excludes the possibility that this contribution is due to the orbital motion of conduction electrons in the magnetic field. The positive magnetoresistivity arises due to spin-polarization of the Co 3d band by the exchange field, an increase of DOS magnitude at the Fermi level, and enhancement of spin fluctuations within this band. This mechanism of positive magnetoresistivity should be common for itinerant-electron metamagnets.

A method to determine the intersublattice exchange coefficient for the two magnetic sublattice systems with antiferromagnetic intersublattice coupling is proposed. The method is based on the existence of the tip-like peculiarity in the magnetoresistivity temperature dependences at the temperature at which compensation between the internal magnetic field and the exchange field takes place.

The fluctuating magnetization of the 4f moment subsystem and the corresponding fluctuating exchange field, which acts on the Co 3d subsystem, give important contribution to the magnetoresistivity, as well as to zero field resistivity of TmCo<sub>2</sub>. Particularly, the positive magnetoresistivity in the fluctuating molecular field is responsible for the upturn of the zero field resistivity near to Curie temperature (in the paramagnetic temperature range). We suppose that similar mechanism accounts for the resistivity upturn in ErCo<sub>2</sub> and for an enhancement of the resistivity near to Curie temperature in HoCo<sub>2</sub> and DyCo<sub>2</sub>.

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