Anomalous magnetotransport in $(Y_{1-x}Gd_x)Co_2$ alloys: Interplay of disorder and itinerant metamagnetism

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New mechanism of magnetoresistivity in itinerant metamagnets with a structural disorder is introduced basing on analysis of experimental results on magnetoresistivity, susceptibility, and magnetization of structurally disordered alloys $(Y_{1-x}Gd_x)Co_2$. In this series, YCo_2 is an enhanced Pauli paramagnet, whereas $GdCo_2$ is a ferrimagnet ($T_c = 400$ K) with Gd sublattice coupled antiferromagnetically to the itinerant Co-3d electrons. The alloys are paramagnetic for $x < 0.12$. Large positive magnetoresistivity has been observed in the alloys with magnetic ground state at temperatures $T < T_c$. We show that this unusual feature is linked to a combination of structural disorder and metamagnetic instability of itinerant Co-3d electrons. This new mechanism of the magnetoresistivity is common for a broad class of materials featuring a static magnetic disorder and itinerant metamagnetism.

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I. INTRODUCTION

Interplay of structural disorder and magnetic interactions opens a rich field of new physical phenomena. Among them are the actively discussed possibility of disorder-induced non-Fermi liquid (NFL) behavior near a magnetic quantum critical point (QCP) as well as a broader scope of effects of disorder on magnetotransport.[1–3] Structurally disordered alloys $Y_{1-x}Gd_xCo_2$ are quasibinary solid solutions of Laves phase compounds YCo_2 and $GdCo_2$. The compounds belong to a large family of isostructural composites RCo_2. YCo_2 is an enhanced Pauli paramagnet whose itinerant Co-3d electron system is close to magnetic instability. In external magnetic field of about 70 T this system undergoes a metamagnetic transition into ferromagnetic (FM) ground state.[4] $GdCo_2$ is, on the other hand, a ferrimagnet with a Curie temperature of 400 K in which the spontaneous magnetization of 4f moments is antiparallel to the induced magnetization of the Co-3d band. Compounds of this family and their alloys provide a convenient ground for experimental studies of magnetotransport phenomena. The electronic structure in the important for the transport vicinity of the Fermi energy is composed mainly of Co-3d states and is, to the first approximation, the same for all compounds of RCo_2 family. It has been found that the main contribution to the resistivity of RCo_2 comes from the scattering of conduction electrons on magnetic fluctuations due to strong s-d exchange coupling,[5] therefore the transport properties are expected to be especially sensitive to the magnetic state of the sample. $GdCo_2$ occupies a special place in RCo_2 family since Gd 4f magnetic moment has no orbital contribution and therefore crystal-field effects are not important for this compound.

The experimental results on the transport properties of $Y_{1-x}Gd_xCo_2$ alloys have been published partly in our previous article.[6] Here we analyze these and new experimental results in order to reveal the physical mechanism of anomalous megnetotransport properties observed in the alloys. In this paper we will discuss the magnetotransport properties of the FM alloys. The properties of paramagnetic alloys will be published elsewhere.

II. EXPERIMENTAL

Samples of $Y_{1-x}Gd_xCo_2$ were prepared from pure components by melting in an arc furnace under a protective Ar atmosphere and were subsequently annealed in vacuum at 1100 K for about one week. An x-ray analysis showed no traces of impurity phases. A four-probe dc method was used for electrical resistivity measurements. Magnetoresistivity (MR) was measured with longitudinal orientation of electrical current with respect to the magnetic field. The size of the samples was typically about $1 \times 1 \times 10$ mm^3. Magnetization was measured by a superconducting quantum interference device (SQUID) magnetometer for samples from the same ingot as that used for the resistivity and ac susceptibility measurements.

III. EXPERIMENTAL RESULTS

The magnetic phase diagram of the $Y_{1-x}Gd_xCo_2$ system inferred from the transport and magnetic measurements[6] is shown in Fig. 1.

Curie temperature T_c decreases with increasing content of Y and eventually drops to zero. A precise determination of the critical concentration x_c which separates the magnetically ordered ground state and the paramagnetic region is difficult, since on the onset of the long-range order its signatures in the magnetic and transport properties are very weak. The first firm evidence of the long-range order are found for alloy with $x = 0.14$ in ac susceptibility at $T = 27$ K, Fig. 2.

Quantum critical scaling theory predicts that when Curie temperature of a FM system continuously depends on an external parameter x, this dependence is expressed as[2]

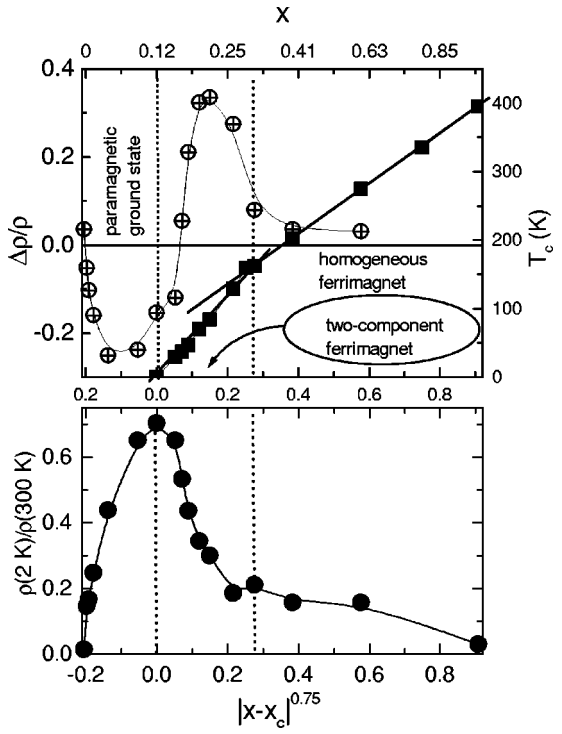

FIG. 1. The upper panel shows the ordering temperature T_c ■ (right y axis), and the MR ⊕ (left axis) of the $Y_{1-x}Gd_xCo_2$ system (Ref. 6). The MR was measured at $T=2$ K in magnetic field of 15 T. The dotted vertical lines indicate phase boundaries at zero temperature. The lower panel displays normalized resistivity $\rho(2\ \mathrm{K})/\rho(300\ \mathrm{K})$.

$$T_c \propto |x - x_c|^{z/d+z-2},$$

with critical index $z=3$ for a FM system of spatial dimension $d=3$. The experimental T_c vs x dependency does follow this relation, but with additional kink at $x=x_t$. A possible origin of this kink will be discussed later. Linear extrapolation of the phase separation line on the phase diagram, Fig. 1 to $T_c=0$ gives as the critical concentration $x_c=0.12$. We do not claim however that QCP exists in this alloy system. Direct experimental verification that $T_c \to 0$ as x approaches x_c from magnetically ordered state is difficult for a disordered alloy system.

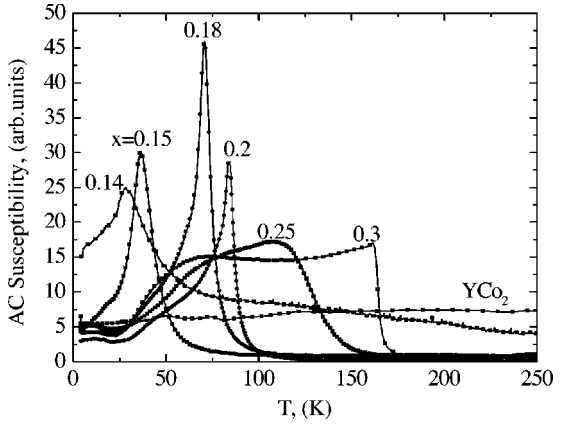

FIG. 2. The ac susceptibility of the $Y_{1-x}Gd_xCo_2$ alloys. Note, the experimental data for YCo_2 and for the alloy with $x=0.14$ are multiplied by factor 20.

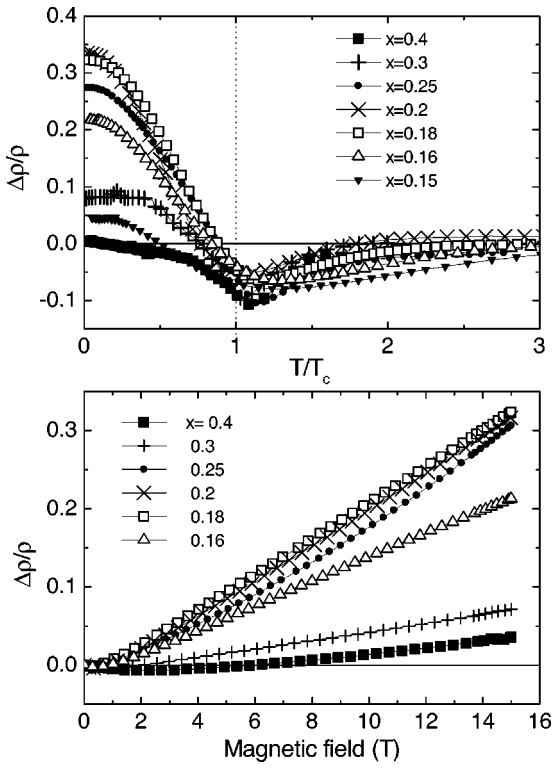

FIG. 3. The upper panel shows the temperature dependence of the MR of the $Y_{1-x}Gd_xCo_2$ alloys, measured in field of 15 T. Large positive MR of FM alloys ($x \leqslant 0.3$) is observed at low temperatures. The field dependencies of MR, measured at $T=2$ K, are presented on the lower panel.

The very surprising result is the positive MR in the FM phase at low temperatures, see Figs. 1 and 3. The well-known theoretical result for MR of a localized moment ferromagnet was derived long ago by Kasuya and De Gennes.[7] As it follows from their theory, MR of a metallic ferromagnet should be negative, having a maximum absolute value at Curie temperature and approaching zero as $T \to 0$, and in the limit of high temperatures. Qualitatively this behavior has been supported by experiment, as well as by later more detailed theoretical calculations. The present experimental results are in a qualitative agreement with this theoretical behavior only for alloys with large Gd content ($x \geqslant 0.4$) (Fig. 3). MR of the FM alloys with composition $0.3 > x > 0.14$ fundamentally differs from this theoretical behavior. Let us note that this composition range falls into the region of the phase diagram between the paramagnetic phase and the additional phase boundary indicated by the kink in the T_c vs x dependency, see Fig. 1. MR of these alloys is positive below Curie temperature and is very large.

The known mechanisms of a positive MR cannot explain the experimental data. A rough estimate of Lorenz force-driven MR one can get from a comparison with the MR of pure YCo_2.[8] In the most pure samples of YCo_2 (with residual resistivity of about 2 $\mu\Omega$ cm) the Lorenz force-driven positive contribution to the total MR does not exceed 5%. On

the other hand the resistivity of the FM alloys at low temperatures falls into the region from 30 to 100 $\mu\Omega$ cm, i.e., at least one order of magnitude larger than the resistivity of pure YCo_2. Therefore, according to Koehler's rule, this mechanism can give MR of only about 0.5%, this has to be compared with the experimental MR of almost 40%.

Weak localization effect is known to give positive MR. However our estimates show that this mechanism can give a contribution which is at least two order of magnitude smaller than the observed MR.

IV. DISCUSSION

The key for understanding the mechanism of the positive MR is a combination of strong dependence of the magnetic susceptibility χ of metamagnetic Co-3d subsystem on the effective magnetic field and of the structural disorder in the R sublattice of the alloys. In case of $GdCo_2$ as well as in the case of other magnetic RCo_2 compounds with heavy R elements, the $4f$-$3d$ exchange interaction is described by introducing an effective field which acts on $3d$ electrons as

$$B_{\mathrm{eff}} = n_{\mathrm{fd}} M_{\mathrm{f}} - B,$$

where B is external field, M_f is the uniform magnetization of R sublattice, and n_{fd} is the f-d coupling constant (in case of $GdCo_2$ $n_{\mathrm{fd}} \approx 50$ T/f.u. μ_{B} (Ref. 9). In $Y_{1-x}Gd_xCo_2$ alloys the Gd moments are randomly distributed over the R sites of the crystal lattice. Therefore, the effective field acting on $3d$ electrons depends on the local distribution of Gd moments and is therefore a random function of coordinate. This random field can be characterized by a distribution function $P\{B_{\mathrm{eff}}(r)\}$. The spatially fluctuating effective field induces an inhomogeneous magnetization of Co-3d electron system:

$$m(r) = \chi(B_{\mathrm{eff}}) B_{\mathrm{eff}}(r).$$

Therefore even at zero temperature in the ferromagnetic ground state, there are two kinds of static magnetic fluctuations in the system: (i) $M_{\mathrm{f}}(r)$; (ii) $m(r)$. These fluctuations give an additional contribution to the resistivity. Since at $T = 0$ K in ferromagnetic phase the $4f$ magnetic moments are saturated the corresponding contribution to the resistivity does not depend on external magnetic field. On the other hand, the $3d$ magnetic moment, as we will see later, is not saturated even in the ferromagnetic ground state. Therefore the $3d$ magnetization does depend on the external field. However, as long as $3d$ susceptibility is field independent and uniform, the external magnetic field will change only the mean (nonfluctuating part) value of the magnetization, whereas the magnitude of the fluctuations of m and corresponding contribution to the resistivity remain unchanged. However actually, the $3d$ system is close to the metamagnetic instability. Therefore the $3d$ susceptibility χ is field dependent and gives rise to static magnetic fluctuations which are dependent on magnetic field resulting in nonzero magnetoresistivity.

For a qualitative analysis of this new mechanism of MR we make the following assumptions.

(1) We consider the system only in its ground state.

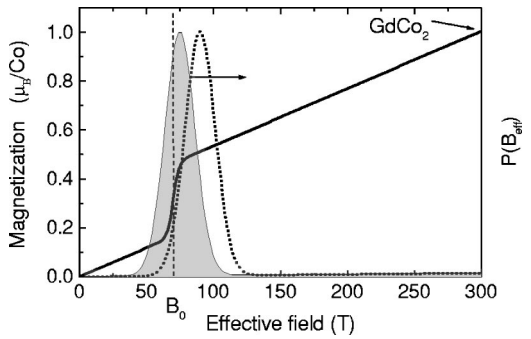

FIG. 4. Dependence of the magnetization of Co-subsystem in RCo_2 compounds vs effective magnetic field (left axis) (Ref. 9) (shown schematically). The dotted line is a schematic representation of the distribution function $P(B_{\mathbf{eff}})$ (right axis), the shaded area indicates the position of $P(B)$ in external field of 15 T.

(2) Correlations between potential and spin-dependent scattering are neglected.

(3) Only the contributions which may be strongly dependent on external magnetic field are retained, e.g., we do not include the effects related to a change of d density of states in magnetic field, and contributions due to potential scattering and scattering on $4f$ moments. In fact only the contribution related to the scattering on fluctuations of Co-3d magnetization will be considered.

Figure 4 shows schematically the dependency of the Co-3d magnetization of RCo_2 compounds on effective magnetic field[9] and the distribution function $P\{B_{\mathrm{eff}}(r)\}$. The metamagnetic transition is indicated by the rapid increase of the magnetization around 70 T.

For further discussion it is important to have an estimate of the magnitude of the fluctuations of the effective field, i.e., the width of the distribution function $P\{B_{\mathrm{eff}}(r)\}$. We can get a hint considering experimental data on field (0–7 T) dependency of the magnetization, Fig. 5.

Two important points follow from these data: (i) there is a nonsaturating para-process in these field dependencies above about 2 T; (ii) the estimated susceptibility of this para-process ($\approx 0.016\ \mu_B$/T) is larger than the susceptibility of $3d$ system below and above metamagnetic transition ($\approx 0.002\ \mu_B$/T), however it is smaller than the susceptibil-

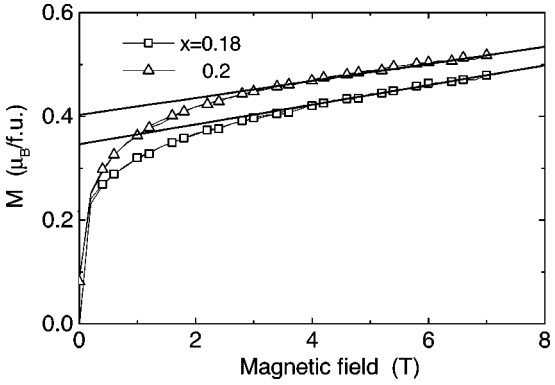

FIG. 5. The magnetization of the $Y_{1-x}Gd_xCo_2$ alloys vs magnetic field.

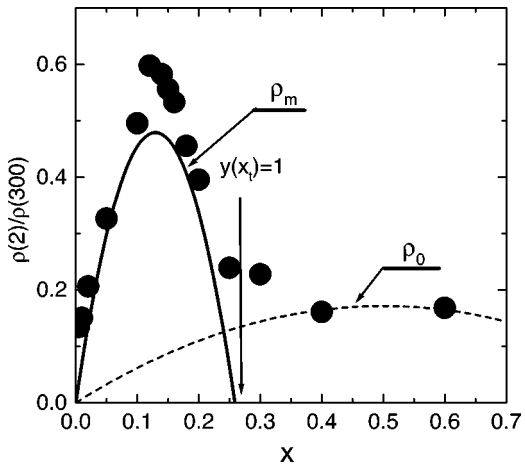

FIG. 6. Schematic of contributions to the low-temperature resistivity of $Y_{1-x}Gd_xCo_2$ alloys. ●—experimental normalized resistivity measured at $T=2$ K in field of 15 T. Solid line represents schematically ρ_m, broken line—ρ_0.

ity in the transition region ($\approx 0.04\ \mu_B$/T).[9] This indicates, that the scale of the fluctuations is larger than the width of the metamagnetic transition. Therefore we cannot treat the fluctuating part of the effective field as a perturbation and have to resort to a phenomenological analysis.

The distribution function $P(B_{\mathbf{eff}})$ describing the fluctuating effective field depends on the alloy composition. For diluted alloys ($x\approx 0$) the most probable value B_{av} of B_{eff} is close to zero. As x increases B_{av} shifts to higher values and in a certain range of the concentrations, the function will have essentially nonzero weight for both $B_{\mathbf{eff}}<B_0$ and $B_{\mathbf{eff}}>B_0$, see Fig. 4. In this case there shall be regions with low and with high $3d$ magnetization in the sample. The resistivity resulting from this static disorder in $3d$ magnetization can be expressed as

$$\rho_m=\rho_{sd}y(1-y),$$

where $y=\int_{B_0}^{\infty}P(B_{\mathrm{eff}})dB$ is the volume fraction of the high magnetization component. Parameter y depends on the alloy composition x and on external magnetic field B. In zero field, point $y=0$ corresponds to $x=0$. As the content of Gd increases B_{av} shifts to larger effective fields, and finally, at some alloy composition x_t, it becomes larger than B_0, i.e., at this composition almost whole volume is occupied by the high magnetization component, i.e., $y\approx 1$. With a further increase of Gd content the mean magnetization of Co should increase with a smaller rate, determined by the slope of the $m(B)$ dependency above B_0, however $y=1$ in this region. According to this scenario ρ_m will increase with x at first, reach a maximum value at x which corresponds to $y=0.5$, and will decrease with further increase of x approaching zero at $x\approx x_t$. We believe that x_t corresponds to the kink on the phase diagram, Fig. 1. The expected variation of ρ_m with the alloy composition is schematically shown in Fig. 6. The total experimental resistivity includes additionally contributions coming from potential scattering and from scattering on $4f$ moments (ρ_0), which both are proportional to $x(1-x)$ and also are depicted in Fig. 6. For comparison the experimental resistivity is shown in this picture too. We present here the resistivity measured at $T=2$ K in the field of 15 T to exclude the contributions related to spin-flip scattering.

The experimental low-temperature resistivity shows the expected variation with x (one needs to keep in mind that the relation between x and y is in general nonlinear, especially around magnetic phase boundary x_c). The resistivity attains the maximum value of about 100 $\mu\Omega$ cm in the region, which corresponds to maximum static magnetic disorder at $y\approx 0.5$ (room-temperature resistivity of the alloys weakly depends on x and is about 150 $\mu\Omega$ cm). About the same value was obtained for the high-temperature limit (maximum magnetic disorder) of magnetic part of the resistivity, arising from scattering on $3d$ temperature-induced magnetic fluctuations in YCo_2.[5] This suggests that the main part of the experimental low-temperature resistivity at $x<x_t$ originates from the scattering on the magnetic fluctuations of $3d$ magnetization, i.e., is identical to ρ_{m}.

Let us show that ρ_m is of order of the spin-fluctuation contribution to resistivity at large temperatures, i.e., is of order of 100 $\mu\Omega$ cm. In case of YCo_2 the spin-fluctuation contribution is the most important and is nearly independent of temperature above about 200 K. Hamiltonian of s-d exchange interaction is given by

$$H_{sd}=G\int d\mathbf{r}\mathbf{s}(\mathbf{r})\mathbf{S}_d(\mathbf{r}),$$

here $\mathbf{s}(\mathbf{r})$ and $\mathbf{S}_d(\mathbf{r})$ are spin density of s- and d- electrons, correspondingly.[17]

Spin fluctuation contribution to resistivity has form:[10]

$$\rho=\frac{3m}{4ne^2}G^2N_s\frac{1}{T}\int_0^1 dqq^3\int_{-\infty}^{\infty}\frac{d\omega\omega}{\sinh^2(\omega/2T)}Im\chi\left(q\frac{2k_F}{k_F^*},\omega\right) \tag{1}$$

where N_s is density of states of s-electrons, k_F/k_F^* is ratio of Fermi momentum of s- and d- electrons. Dynamic susceptibility $\chi(q,\omega)$ is given by the equation:

$$\chi^{-1}(q,\omega)=\chi^{-1}(q)(1-i\omega/\Gamma_q).$$

Here Γ_q is damping of the spin-fluctuations, whereas static nonlocal susceptibility $\chi^{-1}(q)$ is given by

$$\chi^{-1}(q)=\chi^{-1}+Aq^2/N_d$$

with $\chi=N_d/\zeta(T)$, where $\zeta(T)$ is inverse Stoner factor, renormalized by spin fluctuations, N_d is density of states of d-electrons, and $A<1$ is a dimensionless constant.

At large temperatures $T>\Gamma_q$ the expression (1) reduces to:

$$\rho=\frac{3\pi m}{ne^2}G^2N_sT\int_0^1 dqq^3\chi\left(q\frac{2k_F}{k_F^*}\right)\simeq\frac{3\pi m}{4ne^2}G^2N_sT\chi. \tag{2}$$

The last equality is valid when $2Ak_F/k_F^*<\zeta(T)<1$ which must be the case for YCo_2. Note that neglecting mo-

mentum dependence of susceptibility means that s-electrons see the d-spin fluctuations as point scatterers.

Scattering of conducting *s* electrons by the static random distribution of spin density of *d* electrons we can estimate in the following way.

Due to random distribution of magnetic moments of Gd, *s*-electrons experience scattering by

$$\frac{G}{2}\langle S_d(\mathbf{r})\rangle = \frac{G}{2}\int \chi_m(\mathbf{r}-\mathbf{r}')\,\delta B_{eff}(\mathbf{r}')$$

random potential. Here $\chi_m(\mathbf{r}-\mathbf{r}')$ is nonlocal susceptibility near metamagnetic transition. We estimate correlation function of fluctuating effective field as

$$\langle \delta B_{eff}(\mathbf{r})\,\delta B_{eff}(\mathbf{r}')\rangle = (2S_{\mathrm{Gd}}n_{\mathrm{fd}})^2\delta(\mathbf{r}-\mathbf{r}')x(1-x)a^3.$$

Here a^3 is volume of the formula unit. In Born approximation the corresponding contribution to the resistivity is given by the expression

$$\rho_m = \frac{m}{ne^2}G^2N_s(2S_{\mathrm{Gd}}n_{\mathrm{fd}})^2x(1-x)a^3\int_0^1 dq\,q^3\chi_m^2\left(q\frac{2k_F}{k_F^*}\right). \tag{3}$$

Both expressions (1) and (3) give the contributions to the resistivity due to scattering on spin fluctuations, however in the first case they are of thermal origin, whereas in the second case the fluctuations are due to randomness of the effective field.

Assuming that nonlocality of the susceptibility is not important we obtain from Eqs. (2) and (3)

$$\frac{\rho_m}{\rho} = \frac{(2S_{\mathrm{Gd}}n_{\mathrm{fd}})^2x(1-x)a^3\chi_m^2}{3\pi T\chi}.$$

Using experimental results for χ_{m} and χ (Refs. 4,9,11) we find ρ_m/ρ in the range from 0.5 to 3, i.e., the resistivity caused by the static magnetic fluctuations is of the same order as the temperature-induced spin-fluctuation resistivity. The uncertainty is mainly due to determination of χ_{m} near the metamagnetic transition. Taking χ_{m} as the susceptibility of YCo_2 at $B=70$ T (at the field of metamagnetic transition) (Ref. 4) gives the upper bound for ρ_m/ρ. Whereas χ_{m} for the disordered alloy of $x=0.18$, estimated from our results on $M(B)$, Fig. 5 gives the lower bound.

In external magnetic field, the effective field $B_{\mathbf{eff}}$ decreases,[18] therefore y also decreases. Depending on the value of y_0—the volume fraction in zero field ρ_{m} will either increase or decrease, resulting in positive or negative magnetoresistivity: for $0.5<y_0<1$ it will be positive, whereas for $0<y_0<0.5$ we will have a negative MR. In agreement with the model, the experimental MR is positive at $0.15<x<x_t$ and quickly decreases at $x>x_t=0.3$ where $y\approx 1$. Nearly linear field dependencies of MR, observed for $x<0.3$, see Fig. 3, implies that the width of $P(B_{\mathrm{eff}})$ in this composition range is larger than our experimental field limit of 15 T. The region $y<0.5$ almost coincides with the paramagnetic region of the phase diagram. The model predicts negative MR for this region, and this prediction agrees with the experimental result. The model also gives a satisfactory description of the residual resistivity behavior in this region, see Fig. 6. This agreement suggests that at $x<x_c$ the system is actually in spin-glass state. In this region additional essential contributions to MR are present. First, in the paramagnetic region there is a negative MR due to suppression of magnetic disorder in $4f$ magnetic-moment system by external magnetic field. This negative contribution may be the reason why the cross over point from the positive to negative magnetoresistivity does not coincide with the maximum of resistivity, see Fig. 1. Second, there can be additional contributions, both positive and negative, near to zero-temperature magnetic phase boundary due to closeness to QCP.

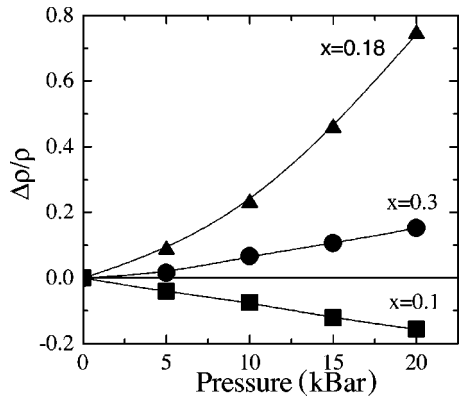

FIG. 7. The resistivity of the $Y_{1-x}Gd_xCo_2$ alloys vs external hydrostatic pressure at $T=2$ K.

An independent test of the model is based on the observation that the critical magnetic field B_0 of the metamagnetic transition in $3d$ subsystem increases under external hydrostatic pressure (P).[12,13] Therefore, basing on the model, we expect that the resistivity of the alloys with the composition left of the resistivity maximum (see Fig. 1) will decrease with pressure, whereas for the alloys right of the maximum it will increase with the increasing pressure. The experimental results for three alloy compositions are shown in Fig. 7.

The sign of the pressure effect is in agreement with the model prediction: the resistivity decreases with pressure for $x=0.1$, whereas it increases with P for $x=0.18$ and $x=0.3$. Moreover, there is a good scaling of pressure and magnetic-field dependencies of the resistivity, Figs. 3 and 7 $(P\rightarrow\alpha B)$ with scaling parameter α which is close to literature data on the pressure dependence of B_0: $dB_0/dP \approx 1.5$ T/kBar.[12,13]

V. CONCLUSION

It has been found that at low temperatures there is a large contribution to the resistivity related to scattering on magnetic fluctuations in metamagnetic itinerant $3d$ system, induced by fluctuating effective field of $4f$ moments. Large positive MR, found in the FM $Y_{1-x}Gd_xCo_2$ alloys and strong pressure dependence of the resistivity are explained as arising from a combination of static magnetic disorder and strong magnetic-field dependence of magnetic susceptibility.

We want to emphasize that this mechanism of resistivity

(and of MR) is not material specific, rather it should be common for a broad class of disordered itinerant metamagnets with strong coupling of conduction electrons to the magnetic fluctuations. In $Y_{1-x}Gd_xCo_2$ the relevant disorder originates from random distribution of *d-f* exchange fields, however a similar effect should arise when there is a random distribution of local susceptibilities (a corresponding treatment for Kondo systems was recently developed in Ref. 14). Positive MR observed in FM alloys $Y(Co_{1-x}Al_x)_2$ (Ref. 15) and in $Er_xY_{1-x}Co_2$ (Ref. 16) may be explained by this mechanism.

ACKNOWLEDGMENTS

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[17] Hereafter we take $\mu_B \equiv 1$.
[18] The effective field decreases for antiferromagnetic $4f$-$3d$ exchange, for ferromagnetic exchange (like that in RCo_2 compounds with light *R*-elements) the effective field increases with the external field.