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Pressure-dependent electrical resistivity of $R\mathrm{Co}_2$ compounds ($R=$ rare earth)

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Studies of the electrical resistivity ρ were carried out under applied hydrostatic pressures up to 20 kbar and in the temperature range from 1.5 K to room temperature. The pressure dependence of the magnetic-ordering temperatures and the suppression of spin fluctuations in the paramagnetic temperature regime have been deduced from the variation of characteristic features in $\rho(T,P)$. Grüneisen parameters of the magnetic-ordering temperature T_C and the spin-fluctuation temperature T_SF have been obtained and discussed with respect to a pressure-induced destabilization of the itinerant d subsystem. Finally, as the measurements are performed under hydrostatic conditions, a possible change in the character of the magnetic phase transition from first towards second order has been investigated. [S0163-1829(98)02405-9]

I. INTRODUCTION

The cubic $R\mathrm{Co}_2$ compounds have attracted much attention due to some peculiar phenomena related to the metamagnetism of the Co sublattice. When R is nonmagnetic ($R=$ Y, Lu, and Sc) the compound is an exchange-enhanced paramagnet exhibiting metamagnetic behavior in externally applied fields larger than about 70 T (at least for YCo_2 and LuCo_2).[1,2] This can be explained in terms of Landau theory, yielding the conditions for the appearance of itinerant electron metamagnetism (IEM) as proposed by Wohlfarth and Rhodes:[3] large and negative values of the derivative of the density of states at the Fermi energy, a situation, e.g., accomplished for YCo_2 as shown by various band-structure calculations.[4–6] Furthermore, these compounds exhibit spin-fluctuation properties such as enhanced values of the Pauli susceptibility χ_0 and the Sommerfeld value γ. Measurements of the susceptibility χ up to 1000 K show broad maxima at temperatures of $T^{\chi}_{\max}$ of about 300 K followed by a Curie-Weiss behavior at considerably higher temperatures,[7] a behavior consistent with Moriya's theory of spin fluctuations.[8] In the electrical resistivity $\rho\propto AT^2$ at low temperatures with an enhanced coefficient A and a tendency towards saturation is observed at temperatures above about 100 K.[9,10] This behavior is in accordance with theory[8,11,12] which also yields a proportionality between the coefficient A and the spin-fluctuation temperature T_SF via $T_\mathrm{SF}\propto 1/\sqrt{A}$. T_SF is here that temperature above which the influence of spin fluctuations with a characteristic energy $k_\mathrm{B}T_\mathrm{SF}$ becomes less important.

In those $R\mathrm{Co}_2$ compounds, where R bears a permanent magnetic moment, induced cobalt moments of about $1\mu_\mathrm{B}$ are observed in the ordered state.[13] As the transition is of first order for ErCo_2, HoCo_2, and DyCo_2 it is assumed that this is due to the metamagnetic behavior of the Co sublattice which experience an exchange field H^{Co}_{fd} from rare-earth spins which exceeds the critical field for IEM, H_C, at T_C. A metamagnetic behavior was, e.g., confirmed by polarized neutron experiments,[14,15] pronounced anomalies in the temperature dependences of the magnetization, the thermal expansion, and the transport properties at the Curie temperature.[13,16,17] In particular, the large discontinuities in the electrical resistivity at T_C of the above $R\mathrm{Co}_2$ compounds were attributed to both the occurrence of IEM and the suppression of spin fluctuations in the itinerant subsystem in the magnetically ordered state below T_C.[10] For the remaining $R\mathrm{Co}_2$ compounds the magnetic transition is of second order for both magnetic subsystems except for TmCo_2 where $H^{\mathrm{Co}}_{fd}<H_c$ and only the Tm subsystem orders below $T_\mathrm{C}\approx 4$ K.[18]

Based on an s-d model, where coexistence of the localized moments of the R atoms and the itinerant d electrons of Co atoms is assumed, Bloch *et al.* explained the first-order phase transition in ErCo_2, HoCo_2, and DyCo_2 within Landau theory.[19] The change towards a second-order phase transition for TbCo_2 and GdCo_2 is attributed to the change in sign of the Landau coefficient $B(T)$ at about 250 K, a value close to $T^{\chi}_{\max}$ of YCo_2. This model was extended later by Inoue and Shimizu, who provided conditions under which the nature of the magnetic phase transitions will alter on substitution or with pressure.[20,21] Although both NdCo_2 and PrCo_2 order at temperatures well below T_B, the transitions observed are of second order. This is ascribed either to the crystal field influence[19] or to a volume effect,[22] with a *critical* lattice constant of $a_\mathrm{c}=7.27$ Å, above which the Co moment becomes more localized due to the larger unit cell volume.

Another approach to account for the influence of pressure on the itinerant d subsystem has been given by Yamada,[23,24] who included the influence of spin fluctuations into Landau theory. Within this theory field-induced IEM takes place only below a certain temperature T_0 which like T_B of the above model is related to $T^{\chi}_{\max}$. In the scope of this model for the magnetic $R\mathrm{Co}_2$ compounds the magnetic transition driven by R is of first order for $T_\mathrm{C}<T_0$ whereas it is of second order when $T_\mathrm{C}>T_0$. In the case that $T_0>T_\mathrm{C}$ this model also predicts a change from a first-order phase transition towards a second-order one as T_0 decreases rapidly with pressure. The same kind of behavior also follows from the predicted increase of H_c with pressure; i.e., $H_\mathrm{c}(P)$ may become larger than H^{Co}_{fd} and the Co subsystem may stay nonmagnetic when the R subsystem orders magnetically. In the

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case of YCo_2 the calculated pressure dependence of the critical field H_c for IEM yields a critical pressure P_c of about 10–50 kbar, depending on the chosen set of parameters.[24]

The ability to exhibit IEM depends sensitively on the Co susceptibility χ_d which is easily affected by substitution as demonstrated, e.g., in the case of $Y(Co_{1-x}Al_x)_2$ (Refs. 25–27) and by pressure. The latter was shown by the measurement of the pressure-dependent susceptibility of YCo_2 at 40 K where Voiron *et al.* deduced $\partial \ln \chi_d / \partial \ln V = 14$ for the volume dependence of χ_d.[28] The pressure dependence of T_C measured for some of the RCo$_2$ compounds by Bloch *et al.*,[29] Voiron and Bloch,[30] and Brouha and Buschow[31] in the early 1970s revealed enhanced values of the respective Grüneisen parameter Ω_{T_C} ($\Omega_{T_C} = -\partial \ln T_C / \partial \ln V$). Based on the s-d model where T_C is given by

$$T_C = N_R \frac{\mu_B^2}{3k_B} G[\mathcal{J}_{RR} + \mathcal{J}_{Rd}^2 \chi_d(T_C)], \qquad (1)$$

where N_R is the number of R atoms in the unit volume, G the de Gennes factor, $\mathcal{J}_{RR}$ and $\mathcal{J}_{Rd}$ are the respective exchange interactions, and $\chi_d(T_C)$ the susceptibility of the itinerant d band at T_C, Voiron and Bloch attributed the large values of Ω_{T_C} to the pronounced pressure response of χ_d.[30] The pronounced nonlinear pressure dependence of T_C of $ErCo_2$ and $HoCo_2$ led Bloch to the assumption that the character of the magnetic transition will change from a first-order phase transistion towards a second-order one at about 20 kbar (see Inoue and Shimizu[21]).

In this paper we report on temperature-dependent resistivity measurements performed on all the RCo$_2$ compounds (with the exception of $GdCo_2$) under hydrostatic pressures up to 20 kbar. These measurements allowed us to study both the pressure dependence of the onset of magnetic order and the influence of pressure on spin fluctuations, particularly in the paramagnetic temperature regime. In both cases we will give Grüneisen parameters of the respective characteristic temperatures T_C and T_{SF}. Then, as the measurements are performed under hydrostatic conditions, they provide information about a possible change in the character of the magnetic phase transition from first towards second order. A change in the nature of the phase transition can be assumed to take place at least for $ErCo_2$ where H_{fd}^{Co} is closest to H_c.[32]

II. EXPERIMENTAL DETAILS

Polycrystalline samples of RCo$_2$ were melted in an induction furnace under a protective argon atmosphere. A ratio of 1:1.93 has been chosen to avoid the presence of magnetic RCo$_3$. Subsequently a heat treatment at 650–800 °C depending on the rare earth during 14 days and under argon atmosphere has been applied.

The phase purity of the samples was proved from Debye-Scherrer photographs and x-ray diffraction (CrK$_\alpha$) measurements. The lattice constants obtained are found to be in good agreement with those reported in literature. Additionally, the samples have been checked by ac and dc susceptibility as well as specific heat measurements. Considerable amounts of foreign phases (less than 5%) were only detected for some of the $NdCo_2$ and $TbCo_2$ samples.

The temperature- and pressure-dependent electrical resistivity was measured on bare shaped samples in the ranges 1.5 K $< T <$ 300 K and $P <$ 20 kbar by means of the common four-probe dc technique. Hydrostatic pressure was generated by a liquid pressure cell, using a 1:4 ethanol-methanol mixture as the pressure-transmitting medium.

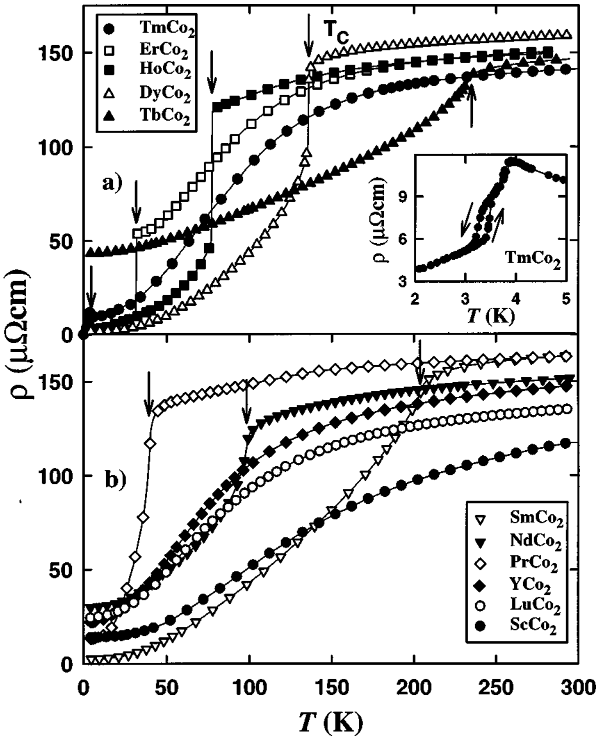

FIG. 1. Temperature-dependent resistivity $\rho(T)$ of RCo$_2$ compounds for $R=$ Tm, Er, Ho, Dy, and Tb (a) and $R=$ Sm, Nd, Pr, Y, Lu, and Sc (b) $ErCo_2$. The inset shows T_C and T_{SR} of $TmCo_2$.

III. EXPERIMENTAL RESULTS

The temperature-dependent resitivity $\rho(T)$ of all the RCo$_2$ compounds studied is given in Figs. 1(a) and 1(b) at ambient pressure. The data are in good agreement with those reported earlier in Refs. 9, 17, 33, and 34. Note that here and in most of the other figures presented only a fraction of the measured data points is given for the sake of clarity. The first-order transitions in $ErCo_2$, $HoCo_2$, and $DyCo_2$ can clearly be deduced from steplike anomalies in $\rho(T)$. For all the other compounds the kinks in ρ vs T at T_C are typically for second-order phase transitions (see the respective arrows in Fig. 1). All the magnetic-ordering temperatures deduced are in good agreement with those obtained by specific heat or ac-susceptibility measurements. In case of $TmCo_2$, $HoCo_2$, and $NdCo_2$ a second anomaly in $\rho(T)$ below T_C indicates a spin reorientation at T_{SR}. The hysteretic behavior observed is, e.g., shown for TmCo2 in the inset of Fig. 1(a). The strong saturation tendency which characterizes the ρ-T curves above T_C is a peculiar behavior of the RCo$_2$ compounds not present in the isostructural RT_2 compounds with $T=$ Al, Fe, or Ni and is attributed to the presence of spin fluctuations.[10]

The temperature-dependent resistivity curves $\rho(T)$ of YCo_2, $LuCo_2$, and $ScCo_2$ are included in Fig. 1(b). The over-

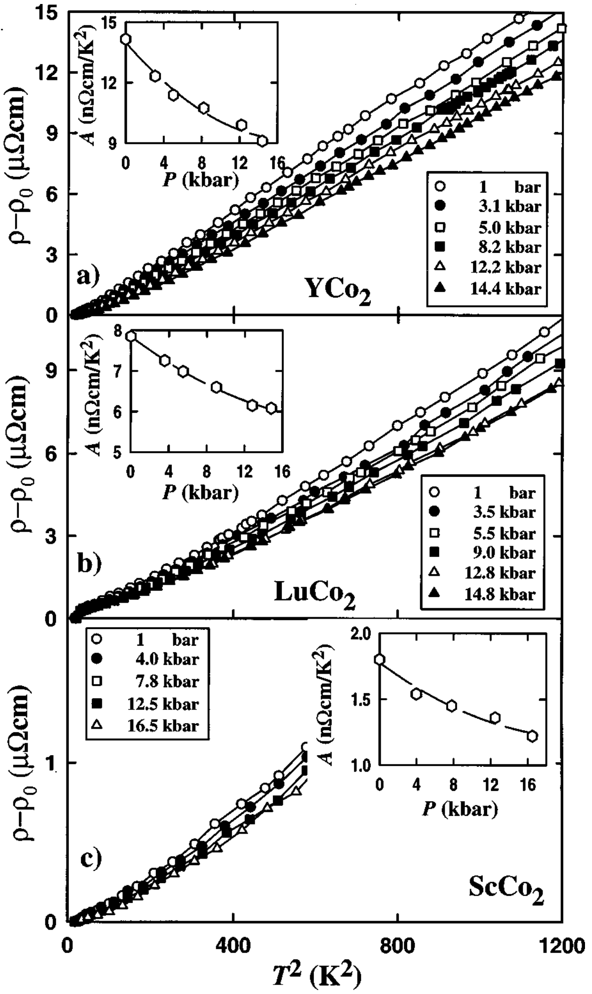

FIG. 2. $[\rho(T)\text{-}\rho_0]$ vs T^2 of YCo_2, $LuCo_2$, and $ScCo_2$ for various values of pressure. The insets show A vs P.

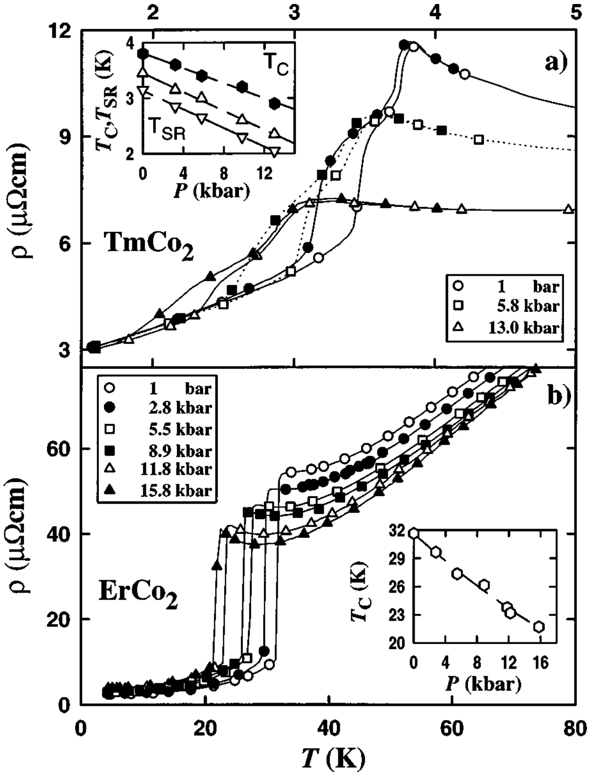

FIG. 3. Temperature-dependent resistivity $\rho(T)$ of (a) $TmCo_2$ and (b) $ErCo_2$ for various values of pressure. The insets show T_C, and also T_{SR} in the case of $TmCo_2$, as function of pressure P. Data for T_{SR} obtained from both the cooling and the heating curves as represented by the respective triangles (△ up and ▽ down).

all temperature dependence of these three compounds resembles the behavior of well-known spin-fluctuation systems such as UAl_2 or UPt_3.[10] The low-temperature regime is characterized by a T^2 behavior, valid over an extended temperature range. The pronounced curvatures in ρ vs T are typical for spin-fluctuation systems having moderate values of T_{SF}.

For all the RCo_2 compounds the application of hydrostatic pressure P results in a reduction of the absolute value of resistivity in the whole temperature range studied. While the shape of $\rho(T)$ remains not affected by pressure considerable influence is observed for temperatures close to the magnetic-ordering temperatures or in the low-temperature regime of the enhanced paramagnetic RCo_2 compounds. Therefore, the $\rho(T,P)$ curves presented in the following sections will cover only limited temperature ranges in order to show the pressure dependence of the spin-fluctuation properties and the magnetic-ordering temperatures in more detail.

A. YCo_2, $LuCo_2$, and $ScCo_2$

Figure 2 depicts the low-temperature behavior of the pressure-dependent resistivity $\rho(T,P)$ of YCo_2, $LuCo_2$, and $ScCo_2$ plottet as $(\rho(T)-\rho_0)$ vs T^2. For all three compounds an almost T^2 behavior holds in $\rho(T)$ up to maximum pressures. A more accurate description of these data taking into account the electron-phonon interaction as BT^5 and the scattering of the conduction electrons with the spin fluctuations as AT^2 yields reasonable agreement. Results of least squares fits for the coefficient A are shown in the insets of Fig. 2 for various values of pressure. (The coefficient B was not found to be affected substantially by pressure.)

At ambient pressure values of $A=$ 14, 7.8, and 1.7 $n\Omega$ cm/K^2 are deduced for YCo_2, $LuCo_2$, and $ScCo_2$, respectively. These values become reduced by pressure by about 20–30 % for maximum pressure applied which in turn indicates an increase of T_{SF} with pressure as $T_{SF}\propto 1/\sqrt{A}$. Note that the range of validity increases with pressure and holds up to 20–30 K in the case of YCo_2. The pressure response of A is most pronounced for YCo_2 which has the largest value at ambient pressure and decreases in the same manner as the absolute value of A is smaller in the case of $LuCo_2$ and subsequently for $ScCo_2$. The absolute value of the total resistivity is strongly reduced in the low-temperature regime while it stay nearly constant at elevated temperatures.

B. RCo_2 ($R=$ magnetic rare earth)

Figure 3 shows the temperature- and pressure-dependent resistivity $\rho(T,P)$ of $TmCo_2$ and $ErCo_2$ for temperatures near the magnetic-ordering points. As a hysteretic behavior in $\rho(T,P)$ at T_{SR} is observed in the case of $TmCo_2$, both the cooling and the heating curves (solid and open symbols) are shown. Note that only a few ρ vs T curves measured at

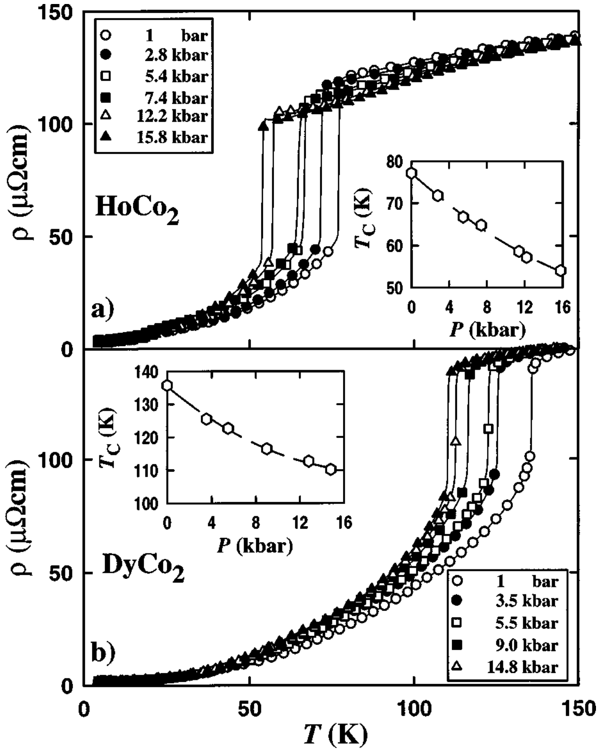

FIG. 4. Temperature-dependent resistivity $\rho(T)$ of (a) $HoCo_2$ and (b) $DyCo_2$ for various values of pressure. The insets show T_C vs P.

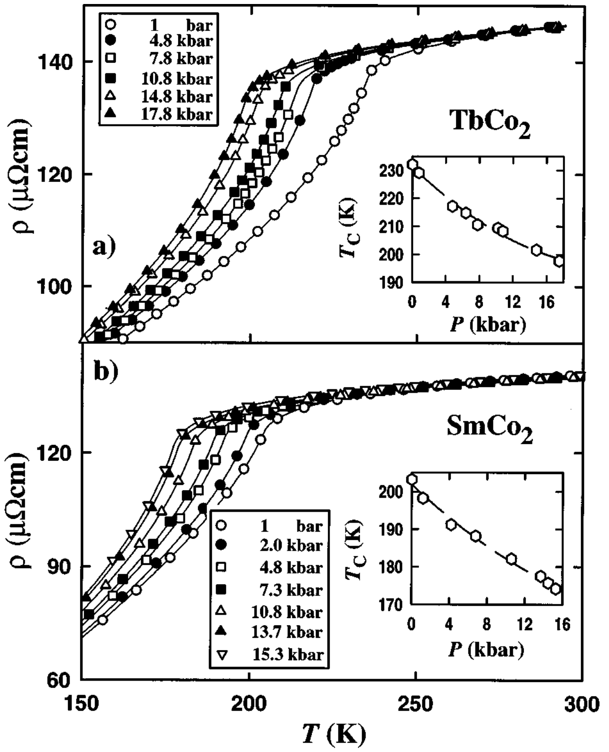

FIG. 5. Temperature-dependent resistivity $\rho(T)$ of (a) $TbCo_2$ and (b) $SmCo_2$ for various values of pressure. The insets show T_C vs P.

various pressures are given. The insets show T_C, and also T_{SR} in the case of $TmCo_2$, as function of applied pressure; the respective values obtained from the cooling and the heating curves are given by triangles (△ up and ▽ down). The applied pressure affects both the ordering temperatures and the absolute value of the resistivity in the paramagnetic temperature range. For $TmCo_2$ a fairly linear pressure dependence of T_C and T_{SR} with $\partial T_C/\partial P = -0.04(4)$ K/kbar and $\partial T_{SR}/\partial P = -0.03(8)$ K/kbar, respectively, is found [see the insets of Fig. 3(a)]. The pressure-dependent variation of the respective ordering temperatures has been deduced from characteristic points in $\partial\rho/\partial T$ vs T. We note that pressure influences $\rho(T,P)$ of $ErCo_2$ in a rather wide temperature range. Above T_C, $\rho(T)$ decreases considerably. Furthermore, the enhancement in resistivity for temperatures just above T_C, which is ascribed to the combined interaction of magnetic correlations within the R sublattice and spin fluctuations in the itinerant d band, becomes more pronounced as pressure increases. For this $\partial T_C/\partial P = -0.8$ K/kbar is deduced. For higher pressures a deviation from linearity is observed [see the inset of Fig. 3(b) and Fig. 11, Sec. IV B]. According to Bloch this change in slope should indicate a change in the order of the magnetic phase transition. However, from our data we could not find a clear hint for a change of the first-order transitions towards second order one for $ErCo_2$.

Figure 4 depicts the temperature-dependent resistivity at various external pressures for the other two RCo_2 compounds which also exhibit a first-order transition at T_C. For both compounds T_C strongly decreases with pressure, with $\partial T_C/\partial P = -1.4$ K/kbar and $\partial T_C/\partial P = -2.1$ K/kbar for the initial slope for $HoCo_2$ and $DyCo_2$, respectively. In the paramagnetic temperature range the response of resistivity to pressure is less pronounced in the case of $DyCo_2$ than compared to $ErCo_2$ or $HoCo_2$ (see also Fig. 11, Sec. IV B). For the latter also a slight upturn in $\rho(T)$ on cooling just above T_C evolves for highest applied pressures. As in the case of $ErCo_2$ the transition at T_C for both compounds seems to be still of first order even for maximum applied pressure. The pressure dependence of the spin-reorientation temperature T_{SR} of $HoCo_2$, which is indicated by a small steplike anomaly at 16 K, will be discussed below (see Fig. 7).

Figure 5 displays $\rho(T,P)$ of $TbCo_2$ and $SmCo_2$. The deduced ratios of the initial pressure dependences of T_C yield values of -2.8 K/kbar and -2.4 K/kbar for $TbCo_2$ and $SmCo_2$, respectively. Although the temperature-dependent resistivity and ordering temperatures of both compounds are close to each other, the pressure-dependent variation of T_C is obviously different. While T_C vs P of $TbCo_2$ shows a clear curvature in the pressure dependence of the ordering temperature only a weak deviation from linearity is deduced for $SmCo_2$.

Because of the high ordering temperature of $GdCo_2$ ($T_C = 395$ K), no measurements have been performed on this compound in this study. However, values of -2.45 K/kbar and -2.7 K/kbar are reported for $\partial T_C/\partial P$ in the literature.[29,31]

The pressure- and temperature-dependent resistivity of the remaining two RCo_2 compounds, $NdCo_2$ and $PrCo_2$, is shown in Fig. 6. The transitions remain of second-order type under applied pressure. The initial rates for the pressure-dependent variation of T_C are -1.1 K/kbar and -0.53 K/kbar for $NdCo_2$ and $PrCo_2$, respectively. The pressure-

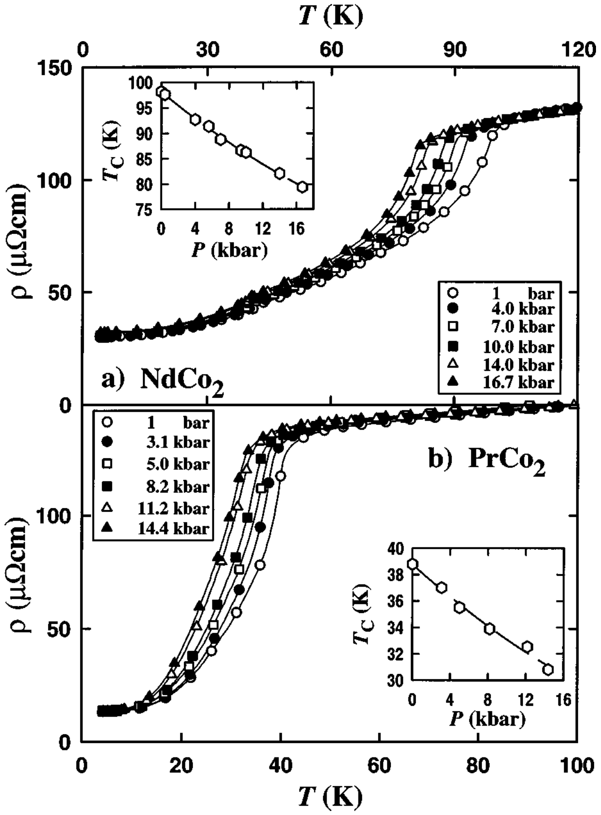

FIG. 6. Temperature-dependent resistivity $\rho(T)$ of (a) $NdCo_2$ and (b) $PrCo_2$ for various values of pressure. The insets show T_C vs P.

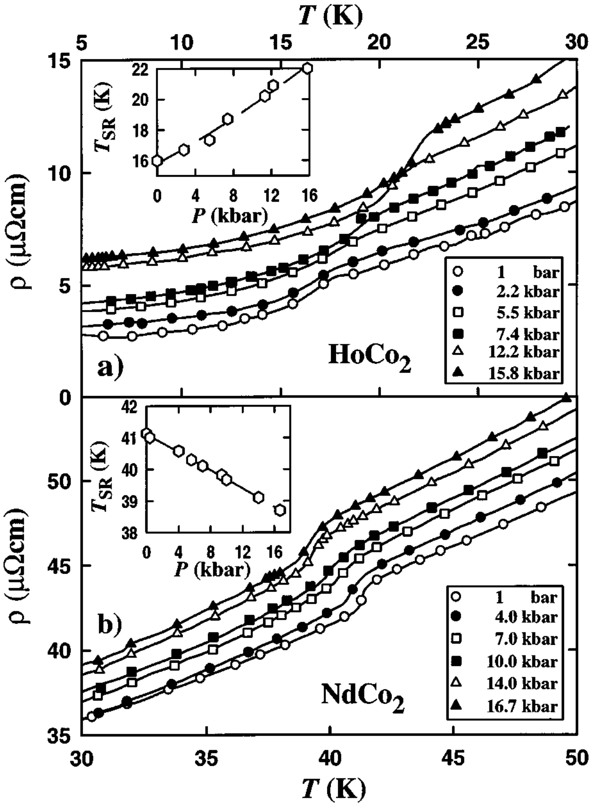

FIG. 7. Pressure-dependent shift of T_{SR} in $\rho(T)$ of (a) $HoCo_2$ and (b) $NdCo_2$ for various values of pressure. The insets show T_{SR} vs P.

dependent variation of T_C, which is depicted in the insets of Fig. 6, shows, as in the case of $SmCo_2$, an almost linear slope.

The pressure dependence of the spin-reorientation temperature T_{SR} of $NdCo_2$ and $HoCo_2$ is shown in Fig. 7. Note that the different $\rho(T,P)$ curves of $HoCo_2$ at various pressures are shifted against each other by $+0.5$ $\mu\Omega$ cm. T_{SR} is indicated by a more or less clear steplike anomaly in ρ vs T. Different signs are observed for the initial pressure dependence of T_{SR}, namely, $\partial T_{SR}/\partial P = +0.21$ K/kbar and -0.14 K/kbar for $HoCo_2$ and $NdCo_2$, respectively. As in the case of the respective T_C vs P dependence some curvature is observed in T_{SR} vs P for $HoCo_2$ in contrast to the linear behavior obtained for $NdCo_2$.

Table I summarizes the ordering temperatures derived from $\partial\rho/\partial T$ vs T and their initial pressure dependence, which have been deduced from a linear fit to the data at low pressures. Data on $\partial T_C/\partial P$ put in brackets are those given in the literature. The observed ordering temperatures are in good agreement with those obtained by specific heat measurements performed on the same samples. The values for the different Grüneisen parameters Ω_{T_C} and Ω_{SR} were deduced from the actual data (except for $GdCo_2$) by using the appropriate compressibility κ_S or, if available, κ_T.[31,36–38]

While for most of the RCo_2 compounds values for Ω_{T_C} of about -11 to -13 have been evaluated an increase up to $\Omega_{T_C} \approx -27$ is found for $ErCo_2$ where H_{fd}^{Co} is assumed to be close to the critical field H_c, indicating that the Co subsystem is easily destabilized by pressure. Note that for $GdCo_2$ a somewhat smaller Grüneisen parameter than for the other compounds is reported. This might be either due to sample quality or to a more stable Co subsystem at elevated temperatures. The latter can be seen in the context that $GdCo_2$ orders magnetically at temperatures well above $T_{\max}^{\chi}$, the temperature above which $\chi(T)$ of YCo_2 exhibits a Curie-Weiss law.

Some discrepancies in $\partial T_C/\partial P$, and consequently in Ω_{T_C}, are observed between our values and those given by Refs. 28 and 29 which are put in brackets in Table I. Especially in the case of certain heavy RCo_2 compounds we deduce a pressure dependence which is almost 2 times larger. But the recent measurements of T_C at 10 kbar on $Dy(Co_{1-x}Al_x)_2$ and $Tb(Co_{1-x}Al_x)_2$ compounds by Leont'ev *et al.*.[35] nicely fit to our data. For an additional test, the Clausius-Clapeyron relation has been employed for $ErCo_2$ and $HoCo_2$, which both show a first-order phase transition at T_C. The values employed for ΔV and ΔS, the change in volume and entropy at T_C, have been taken from measurements on our samples as far as possible or from data given in the literature.[15,39–42] The values thus deduced yield $\partial T_C/\partial P = -0.8$K/kbar and -1.4 to -1.6 K/kbar for $ErCo_2$ and $HoCo_2$, respectively. In the case of $DyCo_2$ no test could be performed due the lack of reliable values for ΔS. However, we want to note that recently Kamarád *et al.*[43] reported $\partial T_C/\partial P = -2.1$ K/kbar of $DyCo_2$ as deduced from thermal expansion measurements which are in good agreement with our data. Therefore, we believe that our results are quite reasonable. The differences to the older results may be related to some extent to advances in sample preparation.

TABLE I. The magnetic-ordering temperatures of the $R\mathrm{Co}_2$ compounds as deduced from resistivity measurements. The initial slope of the pressure dependence of the magnetic-ordering temperature T_C and the spin-reorientation temperature T_SR is deduced from a linear fit to the first data points. Data put in brackets denote those given in the literature. The Grüneisen parameters Ω_{T_C} and Ω_{T_SR} are deduced using the values for the compressibility κ_S given in the literature.

	T_C (K)	$\partial T_\mathrm{C}/\partial P$ (K/kbar)	Ω_{T_C}	T_SR (K)	$\partial T_\mathrm{SR}/\partial P$ (K/kbar)	Ω_{T_SR}
Pr	39	−0.53 [−0.75[a]]	−12.4 [−15[a]]			
Nd	99	−1.1 [−0.8[a]]	−10.8 [−9[a]]	41	−0.14	−3.2
Sm	203	−2.4	−11.3			
Gd	395	[−2.45,[a] −2.7[b]]	[−6,[a] −6.9[b]]			
Tb	232	−2.8 [−1.1,[a] −1.6[b]]	−11.9 [−5,[a] −7.2[b]]			
Dy	135	−2.1 [−0.8,[a] −2.1[c]]	−15.6 [−6[a]]			
Ho	78	−1.4 [−0.65[a]]	19.2 [−9[a]]	16	+0.21	+14.1
Er	32	−0.8 [−0.4[a]]	-27 [−13[a]]			
Tm	3.8	−0.04(4)	−12.9	3.45	−0.03(8)	−12.2

[a] Reference 29.

[b] Reference 31.

[c] Reference 43.

IV. DISCUSSION

A. YCo_2, LuCo_2, and ScCo_2

In the discussion of the physical properties of the magnetic $R\mathrm{Co}_2$ compounds their paramagnetic behavior is commonly approximated by YCo_2. The electronic structure of d electrons in $R\mathrm{Co}_2$ is considered to be very similar to that of YCo_2 and consequently the temperature dependence of $\chi_d(T)$ of $R\mathrm{Co}_2$ is assumed to be similar to that of YCo_2.[44] The occurrence of exchange-enhanced paramagnetism in these compounds is referred to the particular position of the Fermi energy in the density of states curve. In case of the $R\mathrm{Co}_2$ compounds it is assumed that the Fermi energy ϵ_F is near to a steep decrease of a narrow band of hybridized $3d$ and $5d$ states, thus giving rise to the presence of spin fluctuations. The effect of pressure is a broadening of this narrow band, resulting in a decrease of spin fluctuations. Therefore, an analysis of the pressure- and temperature-dependent resistivity in those $R\mathrm{Co}_2$ compounds where R is nonmagnetic ($R=$ Y, Lu, and Sc) allows us to account for the pressure response of the itinerant d-electron subsystem of the $R\mathrm{Co}_2$ compounds.

As shown in Fig. 2 the application of pressure leads to a decrease in the absolute value of the resistivity and the coefficient A. Both observations indicate that spin-fluctuations are suppressed by pressure, a finding which is consistent with an expected reduction of the electron density of states at $\epsilon=\epsilon_\mathrm{F}$ as the d band becomes broader with pressure. Note that at ambient pressure the coefficient A also decreases from YCo_2 via LuCo_2 to ScCo_2 which is consistent with the shrinking unit cell volume along this series.

In a description in terms of a model given by Coqblin *et al.*[12] the spin fluctuation contribution to the total resistivity is a function of the Stoner enhancement factor S and the parameter $\xi=k_{\mathrm{F}_\mathrm{c}}/k_{\mathrm{F}_\mathrm{d}}$, which gives the size difference of the Fermi surfaces for the conduction and the narrow fluctuating band. The change in resistivity observed can be ascribed to the pressure dependence of both parameters. It should be mentioned that within this model scaling of $\rho(T)$ for different values of pressure can only occur for small values of ξ.

In order to account for the pressure dependence of the characteristic temperature T_SF one may look for the pressure variation of the coefficient A. Employing a Grüneisen parameter for A given by

$$\Omega_A=-\frac{\partial \ln 1/\sqrt{A}}{\partial \ln V}=-\frac{\partial \ln T_\mathrm{SF}}{\partial \ln V} \tag{2}$$

reveals a value of $\Omega_A=17$ for YCo_2. According to Wire *et al.*[45] the pressure dependence of $\chi(T)$ is also related to T_SF ($\partial\ln\chi/\partial\ln V=-\partial\ln T_\mathrm{SF}/\partial\ln V$) and can thus be compared to Ω_A, which was proved to be valid for the spin-fluctuation compounds UAl_2 and TiBe_2. In the case of of YCo_2 the pressure dependence of the susceptibility reveals $\partial\ln\chi_d/\partial\ln V=14$, in reasonable agreement with Ω_A.[28] For LuCo_2 and ScCo_2 values of $\Omega_A=11.8$ and 13.7, respectively, are found. (For the latter, κ_T was assumed to be the same as that of YCo_2.)

In the literature[45,46] T_SF is also assumed to be proportional to the inflection point T_inf in ρ vs T, leading to the following relation:

$$\Omega_\mathrm{inf}=-\frac{\partial \ln T_\mathrm{inf}}{\partial \ln V}=-\frac{\partial \ln T_\mathrm{SF}}{\partial \ln V}. \tag{3}$$

However, Eq. (3) is valid only if spin fluctuations dominate $\rho(T)$, i.e., when $\rho_\mathrm{ph}(T)$ can be neglected [this seems to be valid since, e.g., $\rho(300\ \mathrm{K})\approx 30\ \mu\Omega$ cm for isostructural YNi_2 or YAl_2 where no spin fluctuations are present while $\rho(300\ \mathrm{K})\approx 150\ \mu\Omega$ cm for the $R\mathrm{Co}_2$ compounds].[10] Here we assume that the Matthiessen rule is applicable. Employing Eq. (3) and $T_\mathrm{inf}=52$ K, 56 K, and 64 K for YCo_2, LuCo_2, and ScCo_2, respectively, the following values are obtained: $\Omega_\mathrm{inf}=6.6$, 10.5, and 3.9 ($\partial T_\mathrm{inf}/\partial P=+0.33$, $+0.47$, and $+0.24$ K/kbar), respectively. Note that only in the case of LuCo_2, $\Omega_A\approx\Omega_\mathrm{inf}$ while there is a considerable difference for the other compounds. However, referring to Wire *et al.*[45]

TABLE II. The pressure-dependent characteristics of the exchange-enhanced paramagnetic $R\mathrm{Co}_2$ compounds. A and T_{inf} as well as the respective Grüneisen parameters are deduced from pressure- and temperature-dependent resistivity measurements. Values for the Stoner enhancement factor S, the susceptibility χ_0 measured at $T=4.2$ K, the Sommerfeld coefficient γ, and the lattice parameter a are taken from the literature.

	A (nΩ cm/K^2)	Ω_A	T_{inf} (K)	Ω_{inf}	S	χ_0 (emu/mol)	γ (mJ/mol K^2)	a (Å)
YCo_2	14.2	17	52	6.6	9.6[a]	1.8×10^{-3} [b]	30[a]	7.22[b]
LuCo_2	7.8	11.8	56	10.5	8.5[a]	1.2×10^{-3} [b]	26.6[a]	7.11[b]
ScCo_2	1.8	13.7[c]	64	3.9[c]	6.3[b]	1.1×10^{-3} [b]	18.4[a]	6.93[b,c]

[a] Reference 48.

[b] Reference 49.

[c] For κ_T the same value as that of YCo_2 has been employed.

who have shown that $\Omega_A \approx \Omega_\chi$ for UAl_2 and UPt_3 we identify Ω_A with Ω_{SF} in the further discussion.

Table II summarizes the pressure-dependent characteristics of the exchange-enhanced paramagnetic $R\mathrm{Co}_2$ compounds. Values for the Stoner enhancement factor S, the susceptibility χ_0, the Sommerfeld coefficient γ, and the lattice parameter a are taken from the literature. Note that the coefficient A and T_{inf} seem to vary with S and that all gathered data change with volume. Employing both the values of A and of γ all three compounds can be included in the Kadowaki-Woods plot as they nearly obey the relation

$$A/\gamma^2 = 1\times10^{-5}\ \ \mu\Omega\ \mathrm{cm}\,(\mathrm{mol\ K/mJ})^2. \tag{4}$$

This proportionality which, e.g., holds for UAl_2 or UPt_3 is valid for a wide class of strongly correlated electron systems where the ratio A/γ^2 is equally enhanced compared to simple metals.[50]

Figure 8 depicts the temperature- and pressure-dependent resistivity $\rho(T,P)$ of YCo_2, LuCo_2, and ScCo_2 divided by $T_{\mathrm{inf}}(P)$. For each compound a scaling behavior is obviously obtained which holds over the whole temperature range. Such a scaling was also observed for UAl_2 and YMn_2 for pressures up to 20 kbar.[45,47] In Coqblin's model scaling takes only place for small values of ξ. Due to the fact that scaling holds up to maximum applied pressure, it can be assumed that ξ does not vary much with pressure. Therefore the main response of resistivity to pressure seems to be primarily related to a change of the Stoner enhancement factor S rather than to an anticipated broadening of the d band. These findings point to a weak influence of the unknown phonon contribution in this limited pressure range. This leaves the puzzling question of why the Grüneisen parameters deduced in different ways differ substantially for YCo_2 and ScCo_2 whereas good agreement was found for LuCo_2 such as in the case of UAl_2 or YMn_2.[47]

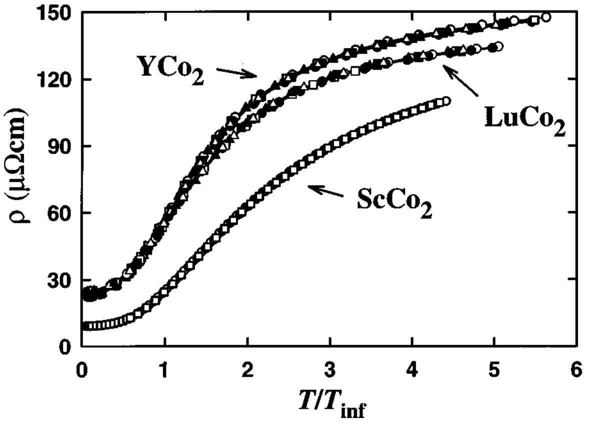

FIG. 8. Temperature- and pressure-dependent resistivity $\rho(P,T)$ of YCo_2, LuCo_2, and ScCo_2 divided by $T_{\mathrm{inf}}(P)$.

Here we want to mention that there is also a clear difference between YCo_2 and LuCo_2 on doping, e.g., with Al: While the maximum T_{C} observed for $\mathrm{Y(Co,Al)}_2$ is of about 27 K it reaches a value of about 145 K at nearly the same Al concentration in the case of $\mathrm{Lu(Co,Al)}_2$.[25,51] Furthermore, the difference in volume between YCo_2 and LuCo_2 corresponds to an applied pressure of roughly 40–50 kbar. According to the calculations given by Yamada[24] for the pressure dependence for the occurrence of IEM at this pressure either no metamagnetism should be observed in LuCo_2 or only at considerably higher fields than $H_{\mathrm{c}}=72$ T, a value which is almost the same as that of YCo_2 ($H_{\mathrm{c}}=68$ T). However, based on the same arguments it is understandable why no metamagnetism for fields up to 120 T is reported for ScCo_2 as in this case the difference in volume relative to YCo_2 amounts to about 100 kbar.

B. $R\mathrm{Co}_2$ ($R=$ magnetic rare earth)

The magnetic behavior of the $R\mathrm{Co}_2$ depends sensitively on the itinerant d subsystem and the internal field generated by the rare earth. In the case of magnetic R a ferrimagnetic coupling among both magnetic subsystems is observed for the heavy $R\mathrm{Co}_2$ compounds whereas it is ferromagnetic for the light ones. Long-range magnetic order is induced in the itinerant d subsystem by the localized R moments.[29] The magnetic-ordering temperatures of the $R\mathrm{Co}_2$ compounds are found to be enhanced compared to those of the isostructural $R\mathrm{Ni}_2$ or $R\mathrm{Al}_2$ compounds where only the R sublattice exhibits magnetic behavior. However, the fact that T_{C} of the heavy $R\mathrm{Co}_2$ roughly scales with the de Gennes factor, which also holds for the $R\mathrm{Al}_2$ or $R\mathrm{Ni}_2$ compounds, indicates that magnetic order is mainly driven by the R sublattice. This differs from the isomorphous $R\mathrm{Fe}_2$ compounds where magnetic order is governed by the itinerant subsystem as indicated by the ferromagnetic order observed for YFe_2 and LuFe_2 with ordering temperatures well above 500 K.[10,55]

In the discussion of the pressure- and temperature-dependent resistivity of the $R\text{Co}_2$ compounds where R is a magnetic rare earth the validity of the Matthiessen rule is assumed. At high temperatures, i.e., well above T_C, $\rho_{\text{mag}}(T)$ consists of two spin-dependent contributions. One is due to scattering of the conduction electrons by spin fluctuations, as in, e.g., LuCo_2, and the other one is due to scattering processes on the localized $4f$ moments, usually denoted as ρ_{spd}. However, the straightforward application of the Matthiessen rule is lacking for several reasons: No appropriate reference for the phonon contribution $\rho_{\text{ph}}(T)$ is available and (ii) no model exists up to now which can be used to account properly for the contribution due to scattering on spin fluctuations which are enhanced by correlation effects due to the presence of a matrix of local moments. The latter leads to a pronounced increase in resistivity for temperatures just above T_C. However, if one assumes that the high-temperature part of $\rho(T)$ of, e.g., LuCo_2 reflects both the phonon contribution and the spin-fluctuation contribution to the resistivity, not enhanced by short-range correlations, a reasonable estimation of ρ_{spd} can be given for the heavy $R\text{Co}_2$ compounds.[52] This in turn provides a possibility to account for that certain magnetic contribution in $\rho_{\text{mag}}(T)$ which is thought to be caused by an enhancement of spin fluctuations due to magnetic correlations within the R sublattice. Following Ref. 52 we define an excess resistivity $\Delta\rho(T)$ by

$$\Delta\rho(T)=\{\rho(T)-\rho_0\}_{R\text{Co}_2}-\{\rho(T)-\rho_0\}_{\text{LuCo}_2}. \qquad (5)$$

At high temperatures a common feature of all the $\Delta\rho(T)$ curves thus evaluated for the $R\text{Co}_2$ compounds is an almost logarithmic decrease in temperature.[34,52] At T_C the spin-fluctuation scattering becomes suppressed when a moment is induced on the Co sites due to the presence of an effective field. The latter is caused by the appearance of a molecular field due to the spontaneous alignment of the R ions. Therefore, spin fluctuations are quenched below T_C and, apart from band-structure effects, the resistivity is reduced and may become even lower than that obtained for LuCo_2. This in turn leads to the negative values of $\Delta\rho(T)$ below T_C occasionally observed.[34,52]

In order to trace the effect of pressure on the temperature-dependent resistivity we have applied this procedure to our measured data, where $\rho(T)$ of LuCo_2 at ambient pressure only has been taken as a reference. Figure 9 depicts $\Delta\rho(T,P)$ of ErCo_2 and HoCo_2. While in the case of HoCo_2 the height in the jump in resistivity at T_C does not change considerably with pressure the same discontinuity becomes progressively smaller in the case of ErCo_2. For maximum applied pressure the discontinuity is only about 60% of that at ambient pressure. For comparison the $\Delta\rho(T,P)$ curves of PrCo_2 and NdCo_2, both of which also order below 100 K, are shown too (see Fig. 10). The latter two also show no essential change in the jump in resistivity at T_C. The same holds also for other compounds ordering at higher temperatures not displayed here.

The extraordinary behavior of the $\Delta\rho(T,P)$ curves of ErCo_2 may be related to the fact that for this compound the evaluated value of H_{fd}^{Co} is closest to H_c. As given in the theoretical considerations by Yamada $H_c(P)$ increases with pressure.[24] Therefore pressure will bring ErCo_2 closer to the metamagnetic transition. Consequently, the moment on the Co sites below T_C thus decreases progressively with pressure. As for the other heavy $R\text{Co}_2$ compounds their deduced effective exchange fields are much higher than H_c and the influence of pressure on the stability of the Co moments is less pronounced. The different behavior observed for light $R\text{Co}_2$ compounds might be related to the assumption that due to the larger unit cell the more localized Co moment is more stable against pressure.

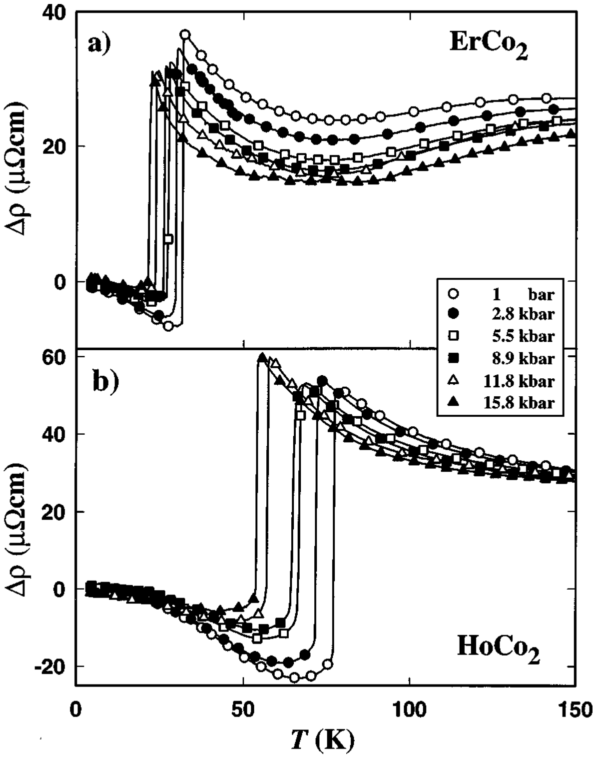

FIG. 9. Temperature-dependent resistivity $\Delta\rho(T)$ of (a) ErCo_2 and (b) HoCo_2 for various values of pressure.

Of certain interest is also the evolution of $\Delta\rho(T,P)$ just above T_C. With increasing pressure the observed cusp at the magnetic transition point becomes more enhanced for ErCo_2 and HoCo_2 than for DyCo_2. Here we want to mention that also, e.g., for $\text{Er}_{1-x}\text{Y}_x\text{Co}_2$ a strong increase in resistivity on cooling towards T_C occurs when $x\approx x_c$, the critical concentration for the breakdown of long-range magnetic order. Neutron diffraction measurements show that ordering of the d subsystem and subsequently long-range order of the R subsystem vanish for higher concentrations of Y.[53] A more detailed study confirmed the presence of both long-range and short-range magnetic order above the ordering temperature for compounds with $x\approx x_c$.[54] The rise in spin fluctuations when approaching the IEM may serve as a source for short-range correlations. In the present case, i.e., for applied pressure, a closely related situation occurs. Therefore we conclude that at the maximum applied pressure ErCo_2 is close to the critical pressure necessary to transform the type of magnetic phase transition towards second order. For pressures above P_c one may consider the case of TmCo_2 where no stable Co moment is present as shown by thermal expansion measurements.[18] For this compound $H_{fd}^{\text{Co}}\approx 60$ T and there-

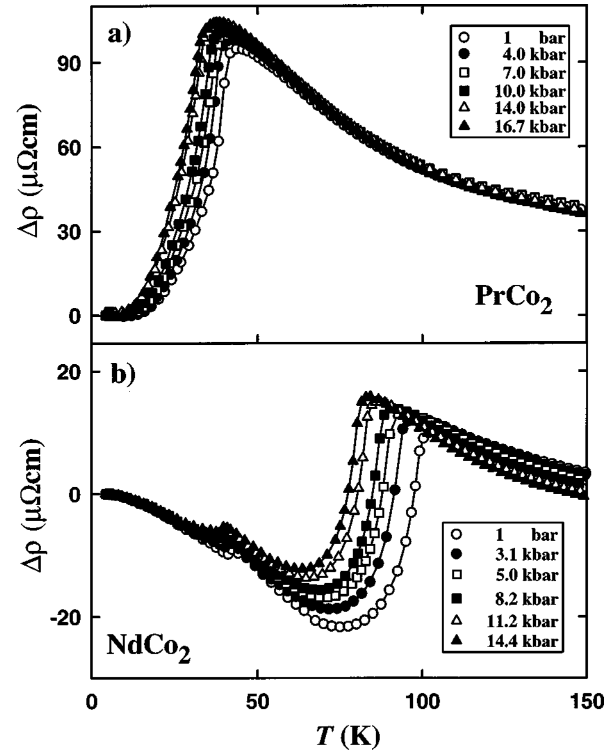

FIG. 10. Temperature-dependent resistivity $\Delta\rho(T)$ of (a) $PrCo_2$ and (b) $NdCo_2$ for various values of pressure.

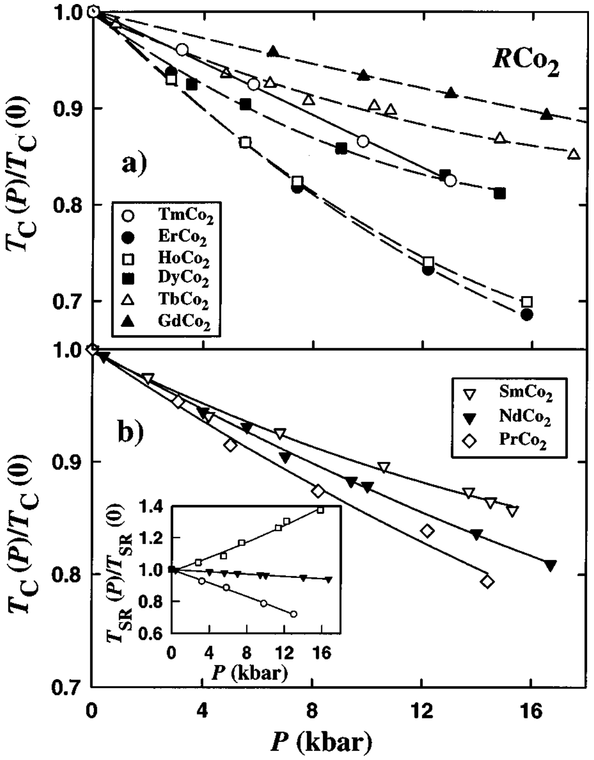

FIG. 11. Pressure dependence of the magnetic ordering temperature T_C for (a) heavy and (b) light RCo$_2$ compounds normalized to ambient pressure. Data for $GdCo_2$ are taken from Ref. 31. The inset shows the pressure dependence of the spin-reorientation temperature T_{SR}.

fore only the R subsystem orders magnetically as the condition for IEM is not fulfilled at T_C. Here, the gradual smearing out of the cusplike resistivity anomaly around T_C with pressure can be understood as a consequence of the suppression of enhanced spin fluctuations above the magnetic-ordering point (see Fig. 3).

The normalized pressure variation of the magnetic ordering temperature T_C is given in Fig. 11. An almost linear behavior is obvious for the light RCo$_2$, $GdCo_2$, and $TmCo_2$ where either a stable or no itinerant Co moment is present. On the other hand, pronounced curvatures in T_C vs P are observed for the remaining compounds which are most prominent in the case of $HoCo_2$ and $ErCo_2$. The change from a linear pressure dependence of T_C, as observed for $PrCo_2$, to a clear curvature for the heavy RCo$_2$ and back to linearity in the case of $TmCo_2$ may reflect the destabilization of the itinerant subsystem. This assumption is in agreement with a fairly linear slope of T_C vs P observed for RAl$_2$,[56] where only a local moment is present, $Y(Co_{1-x}Al_x)_2$,[57] with an itinerant moment only, or RFe$_2$,[58] where the magnetic behavior is governed by the itinerant subsystem. On the other side, the pronounced pressure dependence of the magnetic-ordering temperatures of the RMn$_2$ compounds is due to the destabilization of the itinerant subsystem.[47] Therefore, it is likely to attribute the nonlinear pressure variation of T_C of some of the RCo$_2$ compounds primarily to the pressure response of the itinerant d subsystem.

As the Co moment becomes progressively supressed it can be assumed that once again a linear pressure dependence of T_C will be observed for pressures high enough, i.e., for pressures higher than P_c. Here we want to mention the fairly linear slope of T_C vs P reported for $ErCo_2$, $HoCo_2$, and $DyCo_2$ for pressures above 20–40 kbar (Ref. 30) and recent resistivity measurements up to 60 kbar performed on $ErCo_2$ indicating a nonmagnetic d subsystem above about 20 kbar.[59] Note here also the values for P_c for IEM in YCo_2 given by Yamada which range from about 10 to 50 kbar.

The Grüneisen parameters Ω_{T_C} of most of the RCo$_2$ compounds ($R=$ Pr, Nd, Sm, Tb, and Tm) are found to range from -11 to -13 (see Table I). These values are larger than those obtained for the RAl$_2$ ($\Omega_{T_C}\approx+4$) (Ref. 60) and the RFe$_2$ compounds ($\Omega_{T_C}\approx+1$) (Ref. 61) and may be related to the pressure response of the less stable itinerant subsystem. Apparently, the itinerant subsystem is less stable as H_{fd}^{Co} is closer to H_c which can be clearly traced when proceeding from $DyCo_2$ to $ErCo_2$ where a maximum value of -27 is observed. Note that an enhanced Grüneisen parameter is also obtained for $TmCo_2$ where no induced moment on the Co sites is deduced, but where the d-electron subsystem is close to IEM.

Starting from the directly measured value for $\partial\ln\chi_d/\partial\ln V=14$ and estimates for $\mathcal{J}_{Rd}$ and $\mathcal{J}_{RR}$ proposed by Bloch and Lemaire,[44] Voiron *et al.*[28] calculated Ω_{T_C} within the s-d model [see Eq. (1)] for several of the RCo$_2$ compounds. We want to remind that this model was successfully employed to calculate T_C of the heavy RCo$_2$ compounds from experimentally deduced values for the exchange interactions and $\chi_d T_C$.[20,63] For both $GdCo_2$ and $TbCo_2$ they obtained $\Omega_{T_C}^{calc}=-5.1$ in good agreement with their experimental values $\Omega_{T_C}=-6$ and -5, respectively. In this calculation the pressure dependence of $\mathcal{J}_{RR}$ and $\mathcal{J}_{Rd}$ was neglected, the latter by virtue of the result $\partial T_C/\partial P=0$ for GdNi$_2$.[29] Jaakola, Parviainen, and Penttilä, however, could show that this may not be generalized and deduced nonzero values for $\partial \ln \mathcal{J}_{Rd}/\partial \ln V$ and $\partial \ln \mathcal{J}_{RR}/\partial \ln V$.[62] Therefore, when employing these estimates and taking the more recent data for $\mathcal{J}_{Rd}$, $\mathcal{J}_{RR}$, and $\chi_d(T_C)$ of Ballou *et al.*[63] and Dubenko *et al.*[64] we recalculated the Grüneisen parameters for the heavy RCo$_2$ compounds. In the case of GdCo$_2$ ($\Omega_{T_C}^{\mathrm{calc}}=-5.9$) and TmCo$_2$ ($\Omega_{T_C}^{\mathrm{calc}}=-10$) we obtain good agreement with the experimental data reported in Table I. However, for TbCo$_2$ we estimate $\Omega_{T_C}^{\mathrm{calc}}=-6.2$, only half of the experimental value $\Omega_{T_C}-12$. In the case of DyCo$_2$, HoCo$_2$, and ErCo$_2$ we obtain $\Omega_{T_C}^{\mathrm{calc}}=-5,-4.2$, and -4.2, respectively. However, due to the first-order type of the magnetic phase transition, these data can only serve as rough estimates. Here one should note that Inoue and Shimizu[21] made an attempt to take the influence of the first-order transition into account, but this model is not easy to handle due to a large number of unknown parameters.

Besides the possible influence of a first-order phase transition on the pressure dependence of T_C estimates of Ω_{T_C} within the s-d model appear only to be valid for GdCo$_2$ and TmCo$_2$. However, we want to remind the reader that within this model the measured $\chi(T)$ curve of YCo$_2$ serves as a measure for the Co susceptibility of the magnetic RCo$_2$ compounds and its pressure dependence. Note that from neutron measurements performed on TbCo$_2$ and HoCo$_2$ a more enhanced d susceptibility at T_C was obtained[65] when compared to that of YCo$_2$. It is likely that also $\partial \ln \chi_d(T_C)/\partial \ln V$ will behave different for the various RCo$_2$ compounds, especially when $H_c(P)$ approaches H_{fd}^{Co}. Therefore, the difference observed between the calculated and experimental Grüneisen parameters may be, at least partly, related to uncertainities in the pressure dependence of the itinerant subsystem. The above assumptions are in line with an easily destabilized d subsystem of ErCo$_2$, thus resulting in a large Grüneisen pararmeter.

V. SUMMARY

The measurement of the pressure-dependent resistivity of the RCo$_2$ compounds provides essential information about the pressure response of an itinerant subsystem. Both the spin-fluctuation properties in the paramagnetic temperature region and the magnetic-ordering temperatures are found to be strongly affected by pressure. The extraordinary $\rho(T,P)$ curves observed above T_C are related to the presence of both short-range and long-range correlations close to the magnetic-ordering temperature. The pressure response of the itinerant subsystem primarily explains the peculiar resistivity curves observed, in particular, when the d subsystem is almost destabilized by pressure.

The pressure dependence of the magnetic-ordering temperatures of the magnetic RCo$_2$ compounds reflects well the destabilization of the itinerant Co subsystem. In the case of some of the heavy RCo$_2$ a nonlinear variation of T_C with pressure is observed when the effective field acting on the Co subsystem is close to the critical field for metamagnetism. This is in contrast to $R=$ Pr, Nd, Sm, and Tm where T_C vs P is more or less linear in the pressure range up to 20 kbar. Deviations may occur for higher values of pressures when the itinerant subsystem becomes destabilized. Thus the linearity observed goes along either with stable itinerant moments (light RCo$_2$, GdCo$_2$) or a disordered Co subsystem (TmCo$_2$).

In case of those RCo$_2$ compounds where $R=$ Y, Lu, or Sc two different Grüneisen parameters Ω_A and Ω_{inf} have been evaluated to get information on the pressure dependence of the spin-fluctuation properties in these exchange-enhanced paramagnets. Values obtained are of the order of 10. However, only in the case of LuCo$_2$ are both parameters close to each other and only Ω_A was taken as a measure for $\Omega_{T_{\mathrm{SF}}}$. Large Grüneisen parameters for T_C were deduced with a maximum value of $\Omega_{T_C}=-27$ for ErCo$_2$ where the itinerant d subsystem is closest to instability. The large values of Ω_{T_C} are reasonably explained in terms of the s-d model for TmCo$_2$ and GdCo$_2$, where the Co subsystem is either stable or disordered. In the case of the other heavy RCo$_2$ compounds especially when a first-order phase transition takes place an analysis can be only of a qualitative nature. However, the large values observed appear to be connected with a pronounced pressure dependence of χ_d gradually increasing when H_{fd}^{Co} approaches H_c, consistent with the instability of the Co subsystem against pressure.

No clear evidence for a change of the first-order phase transition towards a second-order one could be observed, but at least for ErCo$_2$ the itinerant d subsystem appears to be almost destabilized at 15 kbar as inferred from the temperature dependence of the resistivity, the nonlinear $T_C(P)$ curve, and the large Grüneisen parameter. This behavior is found to be consistent with predictions on the pressure dependence of the itinerant d subsystem by Yamada.

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