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REMINISCENT RARE-EARTH COBALT MAGNETIC COUPLINGS ABOVE THE CURIE TEMPERATURE AND SPIN FLUCTUATIONS IN RCo_2 COMPOUNDS

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Abstract. The complex and interdependent exchange interactions in RCo_2 compounds with heavy rare-earths are analysed. Reminiscent R5d-Co3d magnetic couplings are still present above the Curie temperatures in a fairly large temperature range $T_c < T < T_1$ with $T_1 \cong T_c$ +300 K. In this temperature range paramagnetic entities of R moments couple antiparallely to those of neighbouring cobalt atoms. The ratio of rare-earth and cobalt moments projected along the field direction (M_R / M_{Co}) is not dependent on the external applied fields nor on temperature. At temperatures higher than $T > T_1$ the rare-earths and cobalt moments decouple, and the cobalt magnetic susceptibilities increase with temperature as predicted by the spin fluctuations model. The saturated effective cobalt moments decrease when the internal mean-field at the cobalt positions increases, a behaviour connected with partial quenching of spin fluctuations.

Key words: rare-earth compounds, paramagnetic behavior, spin fluctuations.

1. INTRODUCTION

The RCo_2 compounds, where R is a rare-earth or yttrium crystallize, at ambient conditions, in a cubic Laves phase type structure, space group $Fd\bar{3}m$. The magnetic ordering induces a finite spontaneous strain, lowering the symmetry to a rhombohedral, $R\bar{3}m$ space group, when R = Tb, Er or to a tetragonal one, $I4_1/amd$ space group, as R = Dy, Ho [1].

The RCo_2 compounds are model systems in analysing the cobalt magnetism. When R = Lu or Y, these are paramagnets. A cobalt moment is induced, when R is a magnetic rare-earth. These systems show ferromagnetic type ordering when R is a light rare-earth and are ferrimagnets for heavy rare-earths. For the heavy rare-earths RCo_2 (R = Gd, Tb, Dy, Ho, Er) the cobalt moment is of $\cong 1\ \mu_B$/atom, and does not dependent on the R element. It was shown that the reciprocal susceptibilities follow non-linear temperature dependences [2–4]. In the early studies the experimental

data were analysed assuming that cobalt has an exchange enhanced magnetic susceptibility [3] or an effective moment [4]. Later on, it was suggested that the cobalt in RCo_2 (R = Y, Lu) shows a spin fluctuations type behaviour [5, 6].

The polarized neutron diffraction studies on RCo_2 compounds with R = Tb [7], Ho [8, 9] and Tm [9, 10] showed that in the temperature range $T_c < T < T_1$, where T_c is the Curie temperature and $T_1 = T_c + \Delta T$, with ΔT is in the range of 200–300 K, the magnetic moments of rare-earths M_R are antiparallely oriented to those of cobalt, M_{Co}, in the presence of external field. The cobalt, in these compounds, was assumed to have an exchange enhanced magnetic susceptibility [7–10]. A similar behaviour was shown in $ErCo_2$ compound [11–13], but in this case the results were interpreted considering the formation of Griffiths- type phases, *i.e.* clusters of magnetic ordered cobalt atoms [14]. Later on, no magnetic, ordered phase was shown at $T > T_c$ [15]. By using the 4f-5d-3d exchange interactions model [16], the magnetic properties of RCo_2 (R = Gd to Tm) compounds were also analysed [17]. It was concluded that R5d band polarizations has an essential role in mediating the exchange interactions at the level of the unit cell.

As an ongoing work on the magnetic properties of RCo_2 compounds with heavy rare-earths, we analyse the exchange interactions, at $T < T_c$, in correlation with cobalt magnetic behaviour. Then, the presence of reminiscent R5d-Co3d magnetic coupling, at $T > T_c$, is discussed in relations with the significant overlap of R5d and Co3d orbitals. We show that the temperature dependences of the effective cobalt moments follow the typical behaviour of systems with spin fluctuations. We stress also that internal fields gradually quench the spin fluctuations.

2. EXCHANGE INTERACTIONS

The exchange interactions in RCo_2 compounds, with magnetic rare-earths, are rather complex and interdependent. Despite the tremendous progress in the computation of exchange interactions using *ab-initio* theories [18, 19] it is still a challenge to apply these theories to complex systems with several magnetic sublattices in multi-orbital setups. For this reasons our analysis is limited to a semi-quantitative description, however it includes almost all possible aspects and results provided by the most recent experimental measurements and takes into account theoretical results as far as possible and relevant.

The exchange interactions between R and Co atoms are of 4f-5d-3d type [15–17]. Through 4f-5d local interactions the 5d band becomes spin-polarized. The 5d-bands spin-polarization was denoted by $M_{5d}(f)$ and using band structure calculations was shown to depend linearly on the De Gennes factor $G = (g_J - 1)^2 J(J + 1)$ [17]. Short-range 5d-3d interactions contributes in building up the cobalt moment, M_{Co}, which is stabilized by Co3d-Co3d interactions. The magnetic dilution of cobalt sublattice has a strong effect on the cobalt magnetism [20]. As result of hybridization of R5d-Co3d orbitals, an additional polarization, $M_{5d}(d)$ is induced on R5d band, proportional with the magnetic moments M_i of their z_i atoms, situated in the first

coordination shell of the R atom. We have shown that for RM_2 (M = Fe, Co, Ni) series, the ratio $a = |M_{5d}| / \Sigma z_i M_i \cong 2.1 \cdot 10^{-2}$ remains constant and does not dependent on the transition metal partner [17]. Because of the larger extension of R5d orbitals, r_{5d} = 5.33 Å [21], there are direct interactions between rare-earths [22], in addition to those mediated by the conduction electrons. The spatial separation between the R and Co atoms, at T = 10 K, was shown to be 3.065 Å in $ErCo_2$ [23] and 3.073 Å in $HoCo_2$ [25]. As these distances are significantly smaller than the rare-earth 5d-orbitals extension, a strong overlap of R5d orbitals of nearest neighbour R atoms exists.

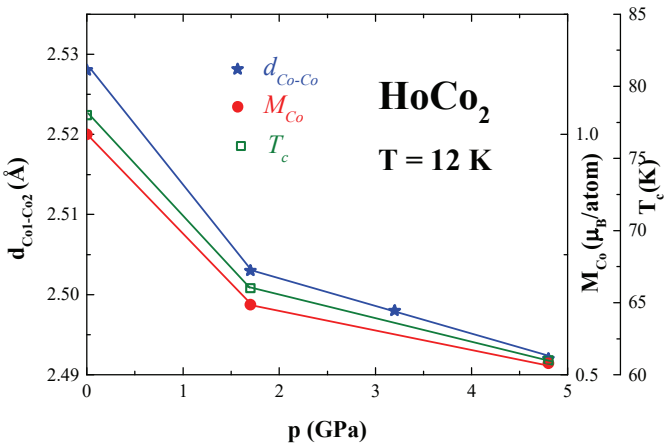

Fig. 1 – Pressure dependences of the distances between cobalt atoms, cobalt moments and Curie temperatures, in $HoCo_2$ compound.

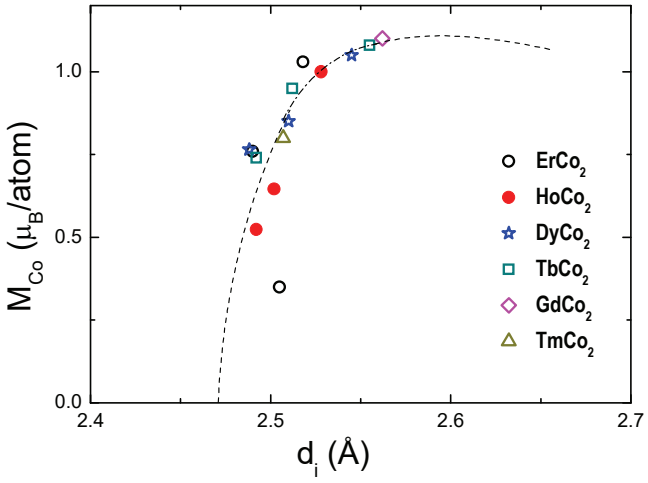

Fig. 2 – The dependence of cobalt moments on the distances d_i between the cobalt atoms as determined from pressure studies. By broken lines is denoted the Néel-Slater curve.

The separation between cobalt atoms, at $T = 10$ K, is slightly smaller than twice the extension of Co3d orbitals, of ≅ 1.3 Å [21]. These values are $d_{Co\text{-}Co} = 2.517$ Å in $ErCo_2$ and 2.53 Å in $HoCo_2$ [23, 25]. References [23–26] reported a similar pressure dependence of the cobalt moments, Curie temperatures and the distances between the cobalt atoms as illustrated in Fig. 1, for $HoCo_2$. The magnitude of the cobalt moments changes with the $d_{Co\text{-}Co}$ parameter and shows a Néel-Slater type behaviour [27] – Fig. 2. This trend is similar with that evidenced in cobalt solid solutions with transition metals, or with that of ^{57}Fe hyperfine fields in RFe_2 compounds [28]. The exchange interactions between cobalt atoms, as determined by neutron inelastic scattering are $J_{Co\text{-}Co} = 10$ meV in $ErCo_2$ [29] and 40 meV for $HoCo_2$ [30]. The magnetic coupling between cobalt atoms break at the Curie temperatures. We mention that the magnetic transition temperature T_c, of RCo_2 compounds, showing a first order magnetic transition, is related to the decrease of the exchange fields, acting on cobalt, below a critical value, $H_c \cong 75$ T [23, 31].

The exchange interactions parameters, $J_{R\text{-}Co}$, between rare-earths and cobalt, determined by neutron inelastic scattering, are negative, their absolute values being more than two order of magnitude smaller than those between cobalt atoms [29, 30, 32, 33]. The $J_{R\text{-}Co}$ values, as well as $M_{5d}(f)$ band polarizations follow a linear dependence on the De Gennes factor, $G = (g_J - 1)^2 J(J + 1)$ as it is shown in Fig. 3.

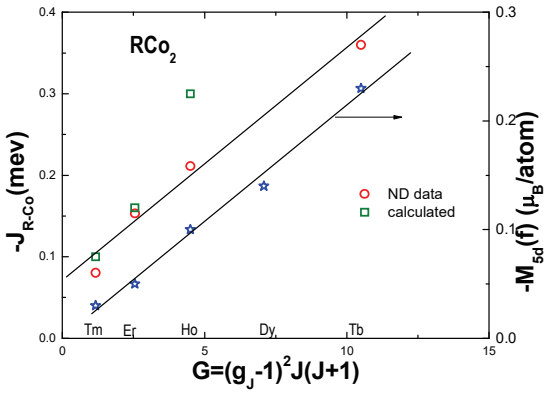

Fig. 3 – The dependences of computed $J_{R\text{-}Co}$ exchange parameters and those experimentally determined as well as of $M_{5d}(f)$ band polarizations as function on De Gennes factor.

The $J_{R–Fe}$ exchange parameters in homologous RM_2 (M = Fe) compounds, were determined from band structure calculations, assuming only the spin contribution to the R magnetic moments [34, 35]:

$$J_{R–M} = (1 / 12)\,(g_J - 1)\,J_{4f–5d}\,(S_{5d}(f) / S_M)\ \text{meV} \qquad (1)$$

with

$$J_{4f–5d} = 94 – 3.4\ (x – 1)\ \text{meV} \quad (2)$$

by S are denoted spin values and x parameter increases by one from the $x = 2$, for $TbFe_2$ up to $x = 6$ in TmM_2, when M = Fe.

As already reported [17], the induced polarization by 4f–5d local exchange, $M_{5d}(f)$, in RM_2 compounds are not dependent on M = Fe, Co, Ni partner. This suggests that the relations (1) and (2) can be also used for estimating $J_{R–Co}$ exchange parameters in RCo_2 compounds. The computed $J_{R–Co}$ values which turns out to agree with the experimental values for R = Tm, Er, but not for R = Tb, Dy. The determined larger values of $|J_{R–Co}|$ parameters than those experimentally reported in RCo_2 compounds, are apparently due to the fact that in relation (2) a linear dependence on the number of 4*f* electrons is used. This leads to a larger slope in comparisons to the one of the $M_{5d}(f)$ band polarizations [36].

3. MAGNETIC BEHAVIOUR OF FERRIMAGNETIC RCo_2 COMPOUNDS IN THE PARAMAGNETIC RANGE

Although the magnetic ordering of cobalt atoms is lost at the Curie temperatures, the magnetic coupling between R and Co atoms still exist in the range $T_c < T \leq T_1$, where T_1 is a temperature situated above the Curie points by about 300 K. By polarized neutron diffraction studies on RCo_2 (R = Tb, Ho, Tm), in the presence of external field, was shown that R and Co moments, in the above temperature range, are antiparallely oriented [7–10]. A similar behaviour was shown in $ErCo_2$, at $T > T_c$, by XMCD [11–13], or polarized neutron diffraction [13] investigations. The magnitude of the R and Co moments, are determined by their projections along the field direction. With increasing external field, both M_R and M_{Co} increase and remain however antiparallely oriented. The ratio $a = M_R/M_{Co}$, for a given compound (*i.e.* a given rare-earth, R) does not dependent on the intensity of the applied field nor on temperature – Fig. 4. These results, in addition to the linear field dependence of magnetization at $T > T_c$, constitute further evidence that the compounds are in the paramagnetic state. Nevertheless, fluctuating magnetic entities resulting from the coupling of the R moments to those of their 12 Co atoms situated in their neighbour exits [13]. The ratios ($a = M_R/M_{Co}$) are linearly dependent on De Gennes factor, similar as $J_{R\text{-}Co}$ values, suggesting a common physical origin – Fig. 4 inset.

Polarized neutron diffraction [37] demonstrated also the antiparallel magnetic coupling between Lu5d-Co3d in paramagnetic $LuCo_2$ single crystal, at $T = 100$ K, in field $\mu_0 H = 5.72$ T. The induced moment on cobalt has a value $M_{Co} = 0.015\ \mu_B$, while the spin-polarization of Lu5d band is $M_{5d} = –0.007(5)\ \mu_B$. The ratio $|M_{5d}| / \Sigma z_i M_i = (3.5 \pm 2.5)\ 10^{-2}$, in the limit of experimental errors is, the same as that determined by analysing the band structures of RCo_2 compounds with magnetic rare-earths [17].

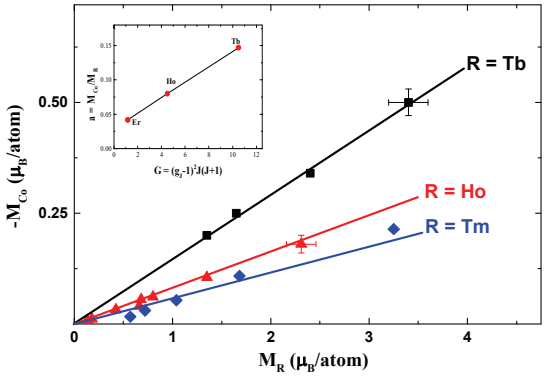

Fig. 4 – The dependences of cobalt moments on the rare-earth ones in RCo_2 compounds with R = Tb, Ho, Tm at $T > T_c$. For each compound, linear trend is evidenced, not dependent on the external field and temperature. In inset is shown the rates of the above dependences as function of De Gennes factor.

The temperature dependences of reciprocal susceptibilities, χ^{-1} *vs* T, of RCo_2 (R = Gd, Tb, Dy, Ho, Er) series change from a nonlinear trend to a linear one, at temperatures T > 700–800 K – Fig. 5. The χ values determined from neutron diffraction studies, in the range $T_c < T < T_c$+300 K [7–10], taking into account antiparallel arrangement of R and Co moments, $\chi = \chi_R - 2\chi_{Co}$, agree well with those obtained by magnetic measurements. Assuming linear χ^{-1} *vs* T dependences, in the above mentioned temperature range, effective moments has been reported of magnitude M_{eff} = 8.8 μ_B/f.u., in $ErCo_2$ [11] and 10.6 μ_B/f.u. in $HoCo_2$ [13], smaller than the R^{3+} free ion values, in agreement with the antiparallel arrangement of R and Co moments, however no reasons for this behaviour were presented.

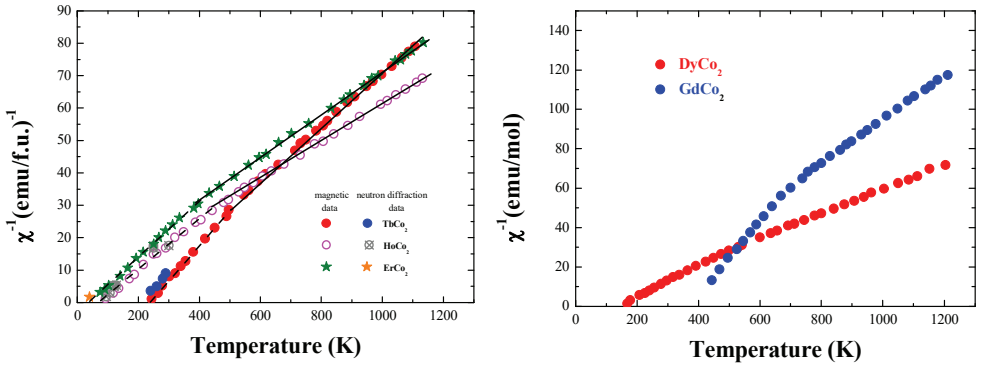

Fig. 5 – Thermal variations of reciprocal susceptibilities for RCo_2 compounds with: a) R = Tb, Ho, Er; b) R = Gd, Dy. The magnetic susceptibilities determined from polarized neutron studies, in the temperature range $T_c < T < 300$ K are as also given.

Starting from the data plotted in Fig. 5, the contributions of cobalt atoms to the magnetic susceptibilities, at $T_c < T < T_1$, $\chi_{Co} = (\chi - \chi_R) / 2$, were determined in different temperature ranges. It was shown that the effective cobalt moments increase from 0.8 μ_B/atom up to 1.7 μ_B/atom in $ErCo_2$ and from 1.36 μ_B/atom to 1.86 μ_B/atom in $DyCo_2$, as temperature increases, in agreement with the predictions of the spin fluctuations model [5]. XMCD studies [14] reported cobalt magnetic moments $M_{Co} = gS_{Co} = 0.21$ μ_B/atom, at $T = 70$ K in $ErCo_2$ [12], and 0.3 μ_B/atom in $HoCo_2$ and 0.4 μ_B/atom in $TmCo_2$ [14]. These correspond to effective cobalt moments $M_{eff}(Co) = g\sqrt{S_{Co}(S_{Co}+1)}$ of 0.7 μ_B/atom, 0.84 μ_B/atom and 1 μ_B/atom, for $ErCo_2$, $HoCo_2$ and $TmCo_2$ respectively, in agreement with data obtained from magnetic susceptibilities. Note however, that in the vicinity of T_1 ($T_1 \pm 30$ K) fluctuations make the orientation of moments less reliable.

At temperatures $T > T_1$ and above the region of critical fluctuations the projections of M_R and M_{Co} magnetic moments, in the presence of external field are parallely oriented, similarly as in classical ferrimagnets. The estimated T_1 temperatures, in RCo_2 compounds with heavy rare-earths, are by 300 ±30 K higher than the Curie points. A somewhat smaller T_1 values were also reported [13]. We expect that thermal energies, corresponding to T_1 temperatures, allows the decoupling of R and Co sublattice magnetizations.

Starting from the data plotted in Fig. 5, we analysed the temperature dependences of the effective cobalt moments, at $T > T_1$, according to the addition law of magnetic susceptibilities, those of Curie constants, respectively. For this aim, different temperature ranges, of ≅ 100 K interval, above T_1 were analysed assuming the linear χ^{-1} *vs* T dependences; the determined effective cobalt moments so obtained are plotted in Figs. 6 at a temperature corresponding to the center of the considered temperature range. The computed effective cobalt moments increase with temperature and saturate at temperatures, depending on R partner, above 700–800 K.

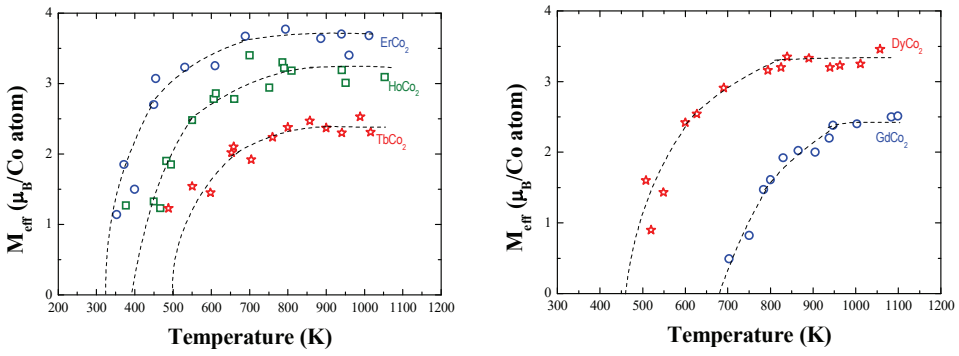

Fig. 6 – Thermal variations of the effective cobalt moments, at $T > T_1$, for RCo_2 compounds with: a) R = Tb, Ho, Er; b) R = Gd, Dy.

The magnetic behaviour of cobalt at $T > T_1$ in ferrimagnetic RCo_2 compounds, can be analysed in comparison with that of YCo_2. At low temperature, T < 10 K, the magnetic susceptibility of YCo_2 follows a T^2 dependence, then increasing up to $T_m \cong 250$ K – Fig. 7 inset [6, 38]. Above a characteristic temperature, $T^* = 560$ K, the magnetic susceptibilities follow a Curie-Weiss type dependence, with an effective cobalt moment of 3.87 μ_B/ Co atom. By using the calculated density of states and taking into account the effect of spin fluctuations, for the general model of itinerant electron paramagnetism, in the classical statistics [39–41], the temperature dependence of the magnetic susceptibility has been analysed. The mean square amplitude of spin fluctuations $<S^2>$, thus obtained, increases with temperature and shows a tendency for saturation, at high temperatures, at a value of 3.48 μ_B/atom, little smaller than that experimentally obtained value from the temperature dependence of χ^{-1} – Fig. 7. The saturation value is little smaller than that of cobalt in ferrimagnetic RCo_2 compounds – Fig. 6. The magnetic behaviour of YCo_2 and the magnetism of cobalt in RCo_2 compounds are characteristic for spin fluctuations system. The wave number dependent susceptibilities, χ_q, of cobalt, has a large enhancement due to electron-electron interactions, for small q values. The average amplitude of spin fluctuations $<S^2> = 3\, k_B T \Sigma \chi_q$, increases with temperature and reaches an upper limit at a characteristic temperature T^* [5].

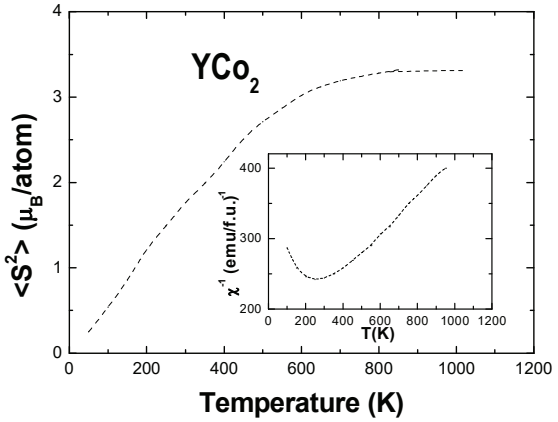

Fig. 7 – Temperature dependence of the mean square amplitude of spin fluctuations in YCo_2. In inset is shown the thermal variation of reciprocal susceptibility.

The saturated effective cobalt moments, are smaller in RCo_2 compounds with larger Curie temperatures, exchange interactions, respectively. This behaviour can be attributed to a gradual quenching of spin fluctuations, when the internal fields increase. A partial quenching of spin fluctuations has been reported in RCo_2 (R = Y, Lu) compounds in the presence of external field [42]. The specific heat decrease by 0.4% in YCo_2 and 1% in $LuCo_2$ when a field of $\mu_0 H = 1$ T is applied. As the magnetic field is sufficiently large, so that the Zeeman splitting energy of

opposite spin states is comparable to, or larger than the spin fluctuations energy, then the paramagnet no longer has sufficiently energy to flip spins and therefore the inelastic spin-flips scattering is quenched. The matter of spin fluctuations quenching by the external/internal fields has been theoretically investigated [43–45]. It was suggested that an effective field, $H_{effc} = k_B T_s / \mu_B s^{1/2}$, where T_s is spin fluctuation temperature and s the Stoner factor, is necessary for quenching spin fluctuations [44]. By using reliable values estimated from the magnetic behaviour of RCo_2 (R = Y, Lu), $T_s \cong 240$ K and $s = 10$, a field $\mu_0 H_{effc} = 112$ T was obtained.

The cobalt spin values, S, determined from effective moments are plotted in Fig. 8 as function of internal field H_{eff} or their square values H_{eff}^2 acting on cobalt in heavy rare earth RCo_2 compounds. Because of data scattering, there is difficult to decide if S values decrease linearly [42] or with a square [43] of internal fields, starting from the estimated critical field H_{effc}. When assuming a linear dependence, there is a decrease of spin values by 0.6% when the exchange field increases by 1 T, value close to those already reported in RCo_2 (R = Y, Lu) compounds [42].

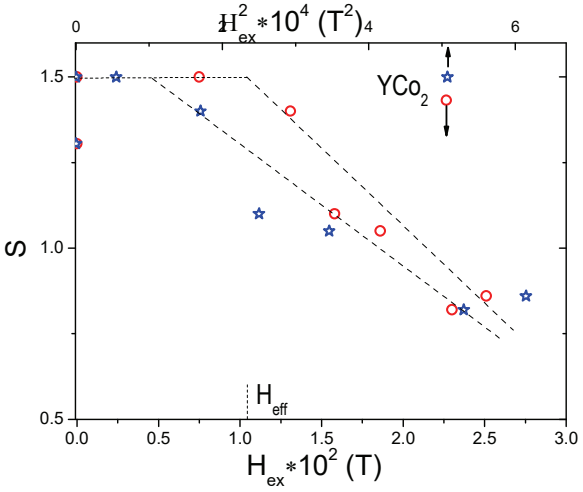

Fig. 8 – The cobalt spin values determined from saturated effective moments in RCo_2 compounds on exchange field, H_{ex} (○) or square of exchange field H_{ex}^2 (★) acting on cobalt in RCo_2 compounds, at $T = 4$ K.

4. CONCLUSIONS

In the present work the Co3d-Co3d short range exchange interactions, as well as those of 4f-5d-3d type are analysed in RCo_2 compounds. Since of high degree of overlap of R5d-Co3d orbitals, the corresponding magnetic couplings are little influenced by temperature, being present also in the paramagnetic range up to a characteristic temperature T_1. In the temperature range $T_c < T < T_1$ paramagnetic

entities of R moments coupled antiparallely to those cobalt neighbours are present. At $T > T_1$ the R and Co moments are magnetically decoupled, the effective cobalt moments increase with temperature reaching finally their saturated values, behaviour predicted by the spin fluctuations model. A gradual quenching of spin fluctuations, is shown as the internal field acting on cobalt increases.

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