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Magnetism in RCo_2 (R = Dy, Ho, Er, Tm) Under Hydrostatic Pressure

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The short-range parimagnetic configurations, observed in the heavy rare-earth RCo_2 compounds in paramagnetic range far above the Curie temperature T_C, consist in formation of ferromagnetic cobalt clusters antiferromagnetically coupled to the neighboring R magnetic moments. The characteristic temperature of the onset of parimagnetism T_f is very sensitive to changes of external conditions like pressure or composition. The pressure coefficients $\partial T_f/\partial p$ and $\partial T_C/\partial p$ are comparable, indicating a close connection of underlying mechanisms. Our recent measurements indicate systematic evolution of the pressure coefficients with spanning the rare-earth series from Dy through Tm in RCo_2 with a specific case $TmCo_2$.

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1. Introduction

The so called parimagnetism, which was discovered few years ago [1], appearing in heavy rare-earth RCo_2 compounds (R = Dy, Ho, Er, Tm) makes the study of compounds from this family attractive again [1, 2]. In the RCo_2 (R = Dy, Ho, Er, Tm) compounds crystallizing in the cubic Laves phase structure the localized rare-earth $4f$-electron magnetic moments coexist together with the itinerant Co $3d$ moments, which appear on the verge of magnetism. Below T_C, the large exchange field due to the ferromagnetically ordered R moments polarizes the Co $3d$-electron states and the emerged moments in the Co sublattice orient antiparallel to the R sublattice. Recent reports propose that weak Co moments survive rather far above T_C in Co ferromagnetic clusters, remaining coupled antiparallel to paramagnetic R moments forming a parimagnetic configuration in the paramagnetic phase. The onset of this phenomenon causes an anomaly in the AC-susceptibility at T_f – the "characteristic temperature of the onset of parimagnetism".

We have performed a systematic study of pressure influence on both the Curie temperature T_C and the parimagnetic temperature T_f in several $RECo_2$ compounds with R = Dy, Ho, Er and Tm. Whereas the detailed results of these high-pressure experiments will be presented elsewhere, here we present summary of the performed measurements.

2. Experimental

Polycrystalline samples were prepared by arc-melting of the stoichiometric ratio of the constituent elements. Each sample was turned and remelted several times for better homogeneity. After annealing and tempering the samples were checked for their quality by X-ray diffraction and the real composition was determined by SEM with the EDX analyzer. Small amount (less than 3%) of RCo_3 impurity phase was found in the samples, except for $TmCo_2$. For measurements of bulk properties (AC-susceptibility, electric resistivity, specific heat) a commercial instruments PPMS, MPMS (both Quantum Design) and Closed Cycle Cryocooler (CCR; Janis Research) were used. Experiments under hydrostatic pressure up to 3 GPa were performed using double-layered clamped pressure cell made of Cu-Be bronze and Ni-Cr-Al alloy with special design to fit into both – PPMS and CCR. Daphne 7373 oil [3] was used as a pressure-transmitting medium and a pressure dependence of resistivity of a manganin wire was used to determine the pressure inside the pressure cell. For the experiment of magnetic susceptibility under pressure a homemade miniature detection coil set was used, which fits inside the bore of the pressure cell [4].

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3. Results and discussion

In AC-susceptibility data for all the studied samples, both the Curie temperature anomaly and the much weaker anomaly at the parimagnetic temperature T_f are clearly visible (see Table). The determination of T_C was supported by the corresponding phase-transition effects present in specific heat and electrical resistivity data, collected at ambient pressure. The estimated Curie temperature values are in agreement with those published before [4–8].

TABLE

Curie- (T_C) and parimagnetic (T_f) temperatures of the studied RCo_2 compounds at ambient pressure.

	T_C (K)	T_f (K)
$DyCo_2$	137	200
$HoCo_2$	80	125
$ErCo_2$	33	98
$TmCo_2$	3.7	35

Apparently the ordering temperature appears to be very sensitive to applied hydrostatic pressure. Comparable values of pressure coefficient has been found also for T_f for corresponding compounds, which indicates a common underlying mechanism of ferrimagnetism and parimagnetism (see Fig. 1). Both, T_C and T_f, shift to lower temperatures with pressure, exhibiting a monotonous evolution of the pressure coefficients across the rare-earth series. The last compound $TmCo_2$ seems then to be a rather exceptional case with the pressure coefficient being almost zero. The pressure coefficients approaching the zero with the increase of atomic number of the rare earth can be connected with the decrease of compressibility of the respective compound, which unfortunately could not be measured experimentally, but it can be expected with respect to lanthanide contraction causing a reduction of the interatomic space. Thus also the strength of exchange interaction will be less influenced by the external pressure, which is exactly what we observe.

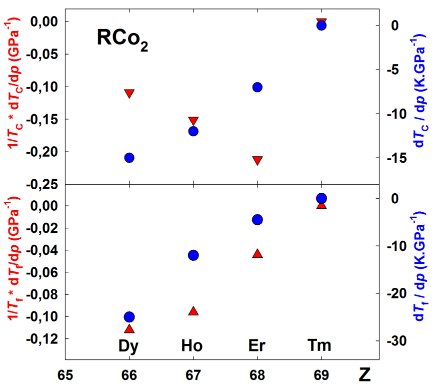

Fig. 1. Evolution of the pressure coefficients and normalized pressure coefficients of the characteristic temperatures T_C and T_f for RCo_2 with R = Dy, Ho, Er, Tm. The values for $HoCo_2$ and $ErCo_2$ were taken from refs. [8] and [4], respectively.

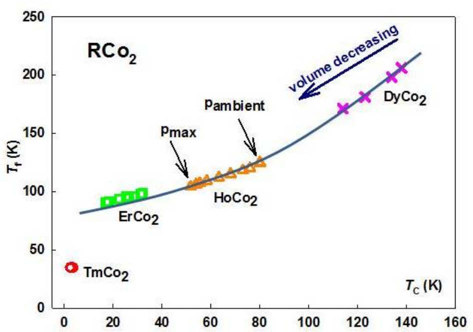

Fig. 2. Empirically estimated relation between the characteristic temperatures T_C and T_f in the studied RCo_2 compounds under various pressures. P_{max} = 1.5 GPa for R = Dy, whereas P_{max} = 3 GPa for R = Tm, Er and Ho.

When searching for a more general relation between T_C and T_f, we have estimated a monotonous and rather smooth evolution, as can be seen in Fig. 2. Here we can see a clearly deviating position of all the pressure points of $TmCo_2$ from the intimated dependence. Although we cannot fully explain the origin of the mentioned behavior yet, it is already clear that the T_C vs. T_f dependence is systematic with the decrease of the unit-cell volume, and that $TmCo_2$ is excess compound with practically zero dependence of T_C and T_f on the applied pressure.

4. Conclusions

We may conclude that our findings of evolution of the Curie temperatures and of the parimagnetic temperatures with the application of external pressure are always characterized by a negative pressure coefficient, which in most of the studied compounds is relatively high in absolute values and systematically evolves when spanning the rare-earth series from Dy through Tm. The exception is $TmCo_2$ which exhibits almost no change of T_C and T_f with pressure up to 3 GPa. This also makes $TmCo_2$ an unusual case in the proposed general evolution T_C vs. T_f for other RCo_2 compounds. Further study for explanation of the pressure influence on the magnetism in RCo_2 ($TmCo_2$) is necessary.

Acknowledgments

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