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Magnetism in $TmCo_2$

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Recently a new wave of interest in physics of RCo_2 compounds has been boosted by reports on unusual magnetic short range order configurations in the heavy rare-earth RCo_2 ferrimagnets in paramagnetic state. The newly observed phenomenon called parimagnetism consists in a short-range antiferromagnetic coupling between small ferromagnetic Co clusters and the nearest rare-earth moments. This paper is devoted to results of our experimental investigation of carefully prepared and characterized samples of $TmCo_2$, a compound on which controversial results can be found in literature. Detailed magnetization, AC susceptibility, specific heat and electrical resistivity measurements revealed two magnetic phase transitions at 4.6 and 3.6 K, respectively. The anomalies connected with the transitions are strongly dependent on magnetic field and hydrostatic pressure. The AC susceptibility anomaly connected with the onset of parimagnetism has been found at $T_f = 35$ K. Contrary to other heavy rare-earth RCo_2 counterparts T_f of $TmCo_2$ is entirely pressure independent.

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1. Introduction

The RCo_2 compounds (where R denotes rare-earth element) crystallize in the cubic $MgCu_2$-type Laves phase. The R ions occupy the diamond lattice sites whereas the closely packed Co ions are located at the common vertices of pairs of equilateral tetrahedrons forming a complete network. These materials serving an example of coexistence of magnetism of well localized $4f$ electrons of rare-earth ions and strongly delocalized Co $3d$ electrons have been a subject of numerous studies already since the second half of the last century. The commonly accepted scenario of physics of these compounds was based on the band character of the Co $3d$ states which are for the heavy rare earths (R = Dy, . . . , Tm, Lu) appearing on the verge of magnetism. The Co $3d$ electron magnetism here comes from the splitting of the spin-up and spin-down subbands. The $3d$ bands are near the Stoner criterion for band ferromagnetism. The spin-up and spin-down bands can be split by large external magnetic field or due to large molecular field acting by the ferromagnetic R sublattice at temperatures below the Curie temperature T_C. Within this scenario Co $3d$ electrons carry no magnetic moment in paramagnetic range ($T > T_C$).

The RCo_2 compounds have attracted new attention after a novel phenomenon called parimagnetism was reported for $ErCo_2$ compound [1]. The parimagnetism is based on a local anti-parallel coupling between ferromagnetic cobalt clusters and nearest rare-earth magnetic moment. The parimagnetism appears in paramagnetic region of ferrimagnetic compounds, $R \in \{Gd, \ldots, Tm\}$ [1, 2]. It can be detected in the temperature dependence of AC magnetic susceptibility, where it appears as a tiny peak like anomaly. X-ray magnetic circular dichroism (XMCD) measurements revealed that the position of this anomaly corresponds to the flipping of cobalt magnetization with respect to the magnetic moment of R. The respective temperature is denoted as flipping temperature T_f.

We have studied $TmCo_2$ compound not only as a further step in the systematic research of the parimagnetism in the RCo_2 compounds but especially because of interesting properties resulting from its position at the end of ferrimagnetic RCo_2 compounds. Moreover, controversial results concerning behavior in the vicinity of the magnetic ordering temperature can be found in literature [3–5] which may be due to varying sample quality. To clarify the situation with magnetic ordering we have paid special care to sample preparation and characterization. The properly characterized sample was subjected to detailed magnetization, AC susceptibility, specific heat and electrical resistivity measurements.

2. Preparation

The $TmCo_2$ polycrystalline sample was prepared from the nominal $TmCo_{1.83}$ stoichiometry by arc melting, where higher amount of the thulium should compensate its higher evaporation. In order to improve homogeneity, the sample was then annealed at the temperature of 1200 K for 10 days. The annealed sample was characterized by the X-ray powder diffraction and by energy dispersive X-ray (EDX) analysis. In the diffraction pattern, there are not visible any foreign diffraction peaks. The microprobe has shown only tiny impurities of Tm rich phase in the majority of the $TmCo_2$ phase. We should note that despite more problematic operation with Tm because of its higher vapor pressure, the quality of this Tm-based sample surprisingly appears to be better in comparison to other RCo_2 compounds previously prepared (with R = Er, Ho, Dy [2,6]), where there was visible a small amount of foreign phase.

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3. Results

The annealed sample was used to perform bulk measurements. There was observed an anomaly at $T_C = 3.6$ K in the temperature dependence of the electrical resistivity (Fig. 1). The anomaly showed unusual increase of the electric resistivity following by a rapid drop with the decreasing temperature. In the temperature dependence of the specific heat (Fig. 1) we observed at 3.6 K a sharp peak which is accompanied by a side maximum at $T^* = 4.6$ K. Closer inspection of resistivity data revels a shoulder around T^*. Application of a magnetic field of 0.5 T promotes the T^* anomaly on account of the anomaly at T_C. We have observed anomaly at the temperature T^* also in the temperature dependence of AC magnetic susceptibility (Fig. 1). The temperature dependence of magnetization showed two steps at the temperature regions close to the temperatures T_C and T^*.

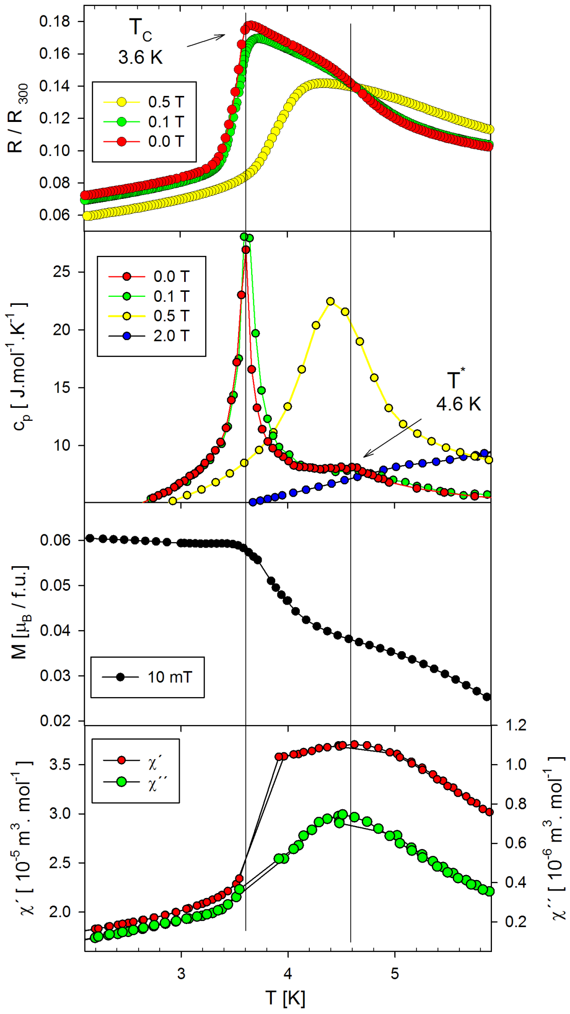

Fig. 1. The comparison of the observed anomalies in bulk measurement data in the ambient pressure (resistivity, heat capacity, magnetization and AC magnetic susceptibility).

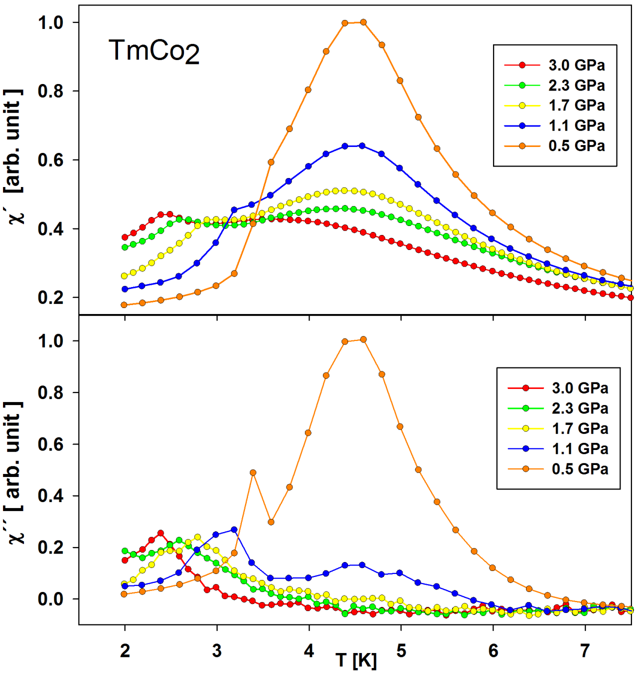

Fig. 2. Temperature dependences of the AC magnetic susceptibility around T_C under the external pressure.

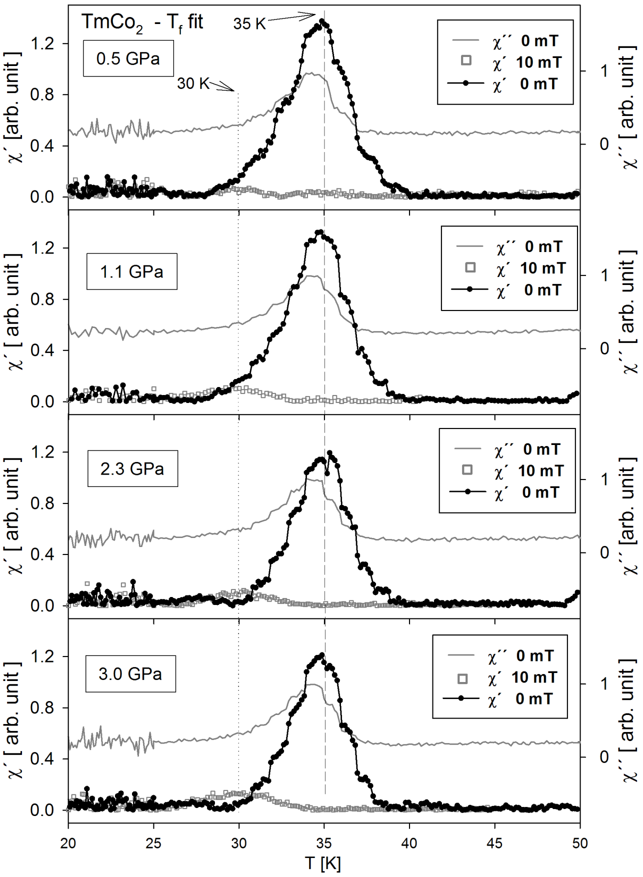

Fig. 3. The subtracted contribution in the temperature dependence of the AC susceptibility under external pressure.

We have performed measurements of the temperature dependence of AC magnetic susceptibility under hydrostatic pressure up to 3 GPa. At the temperatures around T_C and T^* we have observed the splitting of the peak observed at the ambient pressure into the two anomalies (Fig. 2). The anomaly at the lower temperature (T_C) showed slight decrease in the temperature with the increasing external pressure. On the other hand, the second anomaly (T^*) remains unchanged in the position with the pressure.

At higher temperatures we have observed another AC susceptibility anomaly at 35 K (Fig. 3), which is appearing namely under the application of hydrostatic pressure, which should correspond to flipping temperature T_f. In Fig. 3 there is shown contribution to the AC magnetic susceptibility after subtracting a Curie–Weiss law from the measured susceptibility data. The anomaly remains pinned at 35 K irrespective to the applied pressure.

4. Discussion

The anomaly observed at low temperatures (Fig. 1) corresponds well to the anomalies present in the data of other bulk measurements (resistivity, specific heat). From the strong peak in specific heat data corresponding to the Curie temperature of $TmCo_2$, sharpness of the step in the resistivity data and saturation of magnetization below this temperature, we assume this temperature Curie, corresponding being of first order magnetic phase transition (FOMPT). The origin of the second anomaly denoted by the temperature T^* is unknown at the moment, and it is a subject of further studies. In the comparison with the literature we see that the position of the observed anomalies denoted T_C and T^* disagree with some published data [3–5]. Also among these papers there exist discrepancies in the position of Curie temperature and of the other observed anomalies [3, 4]. We suppose that these discrepancies come from different quality of the measured samples. Based on the characterization of our sample by XRD and EDX, we trust in a good quality of our sample — showing better purity in comparison with our samples from the RCo_2 family, showing more consistent results with literature [2, 6, 7]. The position of anomaly denoted by T_f is in the agreement with the position of flipping temperature published in [7] obtained from the XMCD measurement.

5. Conclusions

We have observed the first order phase magnetic phase transition of $TmCo_2$ at $T_C = 3.6$ K. Close to this temperature we have observed another anomaly at $T^* = 4.6$ K, whose origin is unknown at the moment. Although there is a discrepancy with some literature data, we trust in a better quality of our sample, based on results of our careful sample characterization. With respect to these findings, growing of a good quality single crystal and its proper characterization is strongly deserved. Beside this we observed signs of the parimagnetic behavior in the AC susceptibility data at the temperature $T_f = 35$ K. The temperatures of the observed anomalies were found almost independent on external pressure.

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References

[1] J. Herrero-Albilos, F. Bartolomé, L. Garcia, A. Young, T. Funk, J. Campo, G. Cuello, *Phys. Rev. B* **76**, 094409 (2007).

[2] M. Míšek, J. Prokleška, V. Sechovský, D. Turčinková, J. Prchal, A.F. Kusmartseva, K.V. Kamenev, J. Kamarád, *J. Appl. Phys.* **111**, 07E132 (2012).

[3] E. Gratz, R. Hauser, A. Lindbaum, M. Maikis, R. Resel, G. Shaudy, R.Z. Levitin, A.S. Markosyan, I.S. Dubenko, A.Yu. Sokolov, S.W. Zochowski, *J. Phys. Condens. Matter* **7**, 597 (1995).

[4] R. Resel, M. Hedo, T. Nakama, K. Yagasaki, *Solid State Commun.* **95**, 735 (1995).

[5] I.S. Dubenko, I.V. Golosovsky, E. Gratz, R.Z. Levitin, A.S. Markosyan, I. Mirebeau, S.V. Sharigin, *J. Magn. Magn. Mater.* **150**, 304 (1995).

[6] J. Prchal, J. Šebesta, J. Valenta, M. Míšek, D. Turčinková, L. Lapčák, J. Prokleška, M. Kratochvílová, V. Sechovský, *Acta Phys. Pol. A* **126**, 288 (2014).

[7] C.M. Bonnila, J. Herrero-Albillos, A.I. Figueroa, C. Castán-Guerrero, J. Bartolomé, I. Calvo-Almazán, D. Schmitz, E. Weschke, L.M. García, *J. Phys. Condens. Matter* **26**, 156001 (2014).