Proceedings of the 15th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 17–21 2013

Presence of Parimagnetism in $Ho(Co_{1-x}Si_x)_2$ Under Hydrostatic Pressure

J. Valenta*, J. Prchal, M. Kratochvílová, M. Míšek, V. Sechovský
Charles University in Prague, Faculty of Mathematics and Physics, DCMP, Ke Karlovu 5,
121 16 Prague 2, Czech Republic

A new configuration of cobalt magnetic moments with respect to rare-earth ones on local level, named as *parimagnetism*, surviving above the Curie temperature was determined few years ago for $ErCo_2$. Occurrence of parimagnetism has recently been confirmed also for $HoCo_2$. In this paper we present results manifesting considerable composition and hydrostatic-pressure influence on characteristic temperatures of magnetism in $Ho(Co_{1-x}Si_x)_2$ compounds and draw a preliminary scenario analyzing the exchange interactions involved.

DOI: 10.12693/APhysPolA.126.406
PACS: 75.20.En; 75.50.Gg

1. Introduction

The $RECo_2$ compounds (RE = rare earth metal) which crystallize in the cubic Laves phase structure ($MgCu_2$-type, space group *Fd-3m*) have been a subject of interest for more than fifty years as model materials in which the localized and itinerant electron magnetism coexist. The $4f$ electrons in the RE ions bear a localized magnetic moment while the itinerant Co magnetism originates in the splitting of the Co $3d$ spin-up and spin-down subbands. Below the Curie temperature T_C, the RE and Co magnetic sublattices are ferromagnetic. The antiferromagnetic inter-sublattice exchange, present for RE = Gd ... Tm, leads to ferrimagnetic order [1]. It has been shown recently that small Co magnetic moments survive at temperatures far above T_C and yield to short-range order correlations forming Co magnetic clusters in the paramagnetic state [2–4]. Magnetic clusters (correlation length value $\sim 7 \pm 1$ Å) couple antiparallel to the nearest RE magnetic moment [2, 3]. This configuration was denoted as parimagnetic. Up to now the parimagnetic behavior was observed for $ErCo_2$, $HoCo_2$, $DyCo_2$ and $TmCo_2$ to exist in the temperature range between T_C and T_f (flipping temperature) [2–4]. Here we present results of investigations of influence of alloying and external hydrostatic pressure on the characteristic temperatures of magnetism in $HoCo_2$.

2. Experimental methods

Polycrystalline samples $Ho(Co_{1-x}Si_x)_2$ were prepared by melting in mono-arc furnace using pure elements. Characterization of samples showed small amount of $HoCo_3$ as impurity phase and no change of elementary-unit-cell volume with Si substitution.

Temperature dependences of electrical resistivity, AC susceptibility and specific heat were measured by using PPMS, MPMS instruments (both Quantum Design) and Closed Cycle Refrigerator (CCR; Janis Research). We used a double-layered piston-cylinder pressure cell mounted in the PPMS and/or CCR for experiments under hydrostatic pressures up to 3 GPa. The electrical resistivity was measured by a standard four-point method, whereas a homemade miniature detection coil set, which fits inside the pressure cell, was used for AC susceptibility measurements [4]. The Daphne oil 7373 [6] was used as the pressure transmitting medium and a manganin manometer was used for determination of pressure inside the cell.

3. Results and discussion

Measurements at the ambient pressure confirmed magnetic ordering occurring at T_C, observable by all methods involved (Fig. 1). At low temperatures, specific heat and AC susceptibility show an anomaly at the temperature T_R, caused by the reorientation of the easy direction of magnetization [7]. A tiny anomaly at $T_f \sim 125$ K for $HoCo_2$ and $T_f \sim 117$ K for $x = 0.025$ can be found on the AC susceptibility data. These anomalies are rapidly decaying in the applied magnetic field and vanish already in the field of 10 mT. T_f has been ascribed to the flip of the magnetization of the Co clusters in $HoCo_2$ [3]. We assume analogous origin of the anomaly at T_f for the substituted sample.

All characteristic temperatures shift with Si substitution in a non-monotonous way (T_C increases, T_f and T_R decrease). Substitution of Si for Co leads to dilution of Co-magnetic sublattice. According to [8] the reason for increasing of T_C with substitution may be due to experimentally estimated increased Co moment. Dilution thus can result in higher net magnetic moment of the system although the magnetic interaction will be weaker. The shift of T_f is probably due to strengthening spin fluctuations of Co moments or disturbance of Co magnetic clusters by Si. No T_f related anomaly can be recognized for $x = 0.075$ above T_C. When considering the approaching tendency of T_C and T_f with Si doping up $x = 0.025$,

*corresponding author; e-mail: valeja@centrum.cz

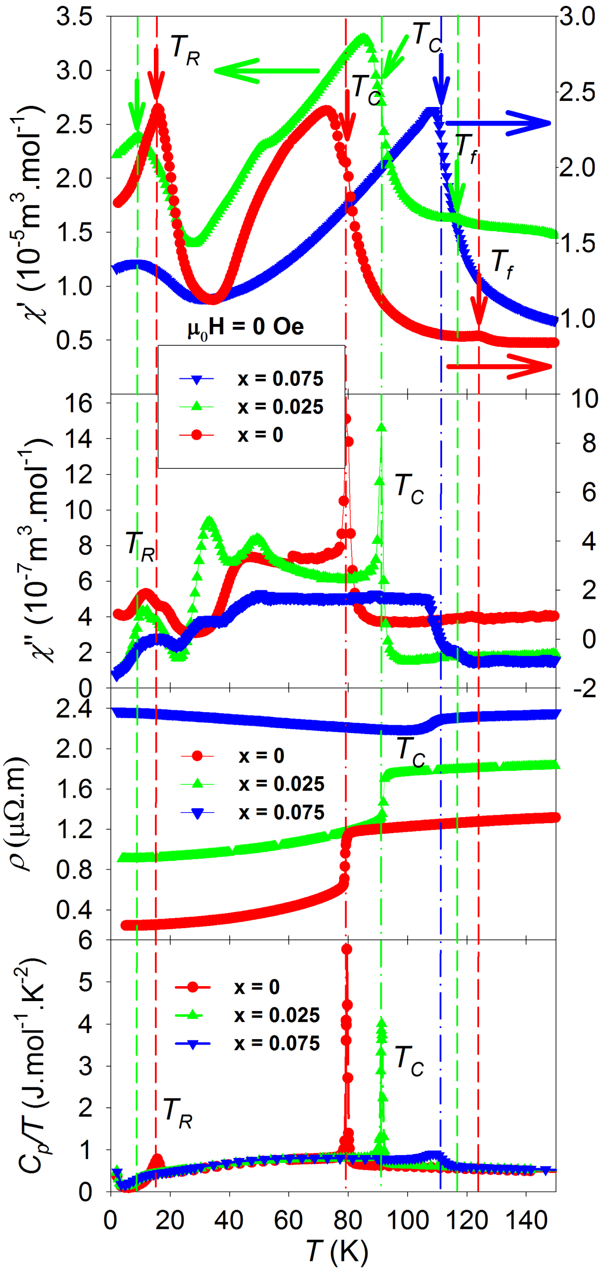

Fig. 1. Temperature dependences of AC magnetic susceptibility, electric resistivity and specific heat for $Ho(Co_{1-x}Si_x)_2$ with $x = 0.00$, 0.025 and 0.075.

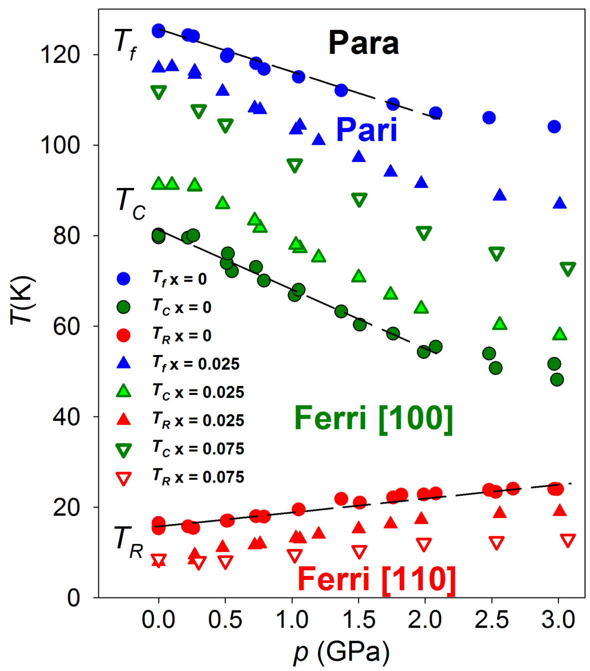

Fig. 2. Phase diagram of $Ho(Co_{1-x}Si_x)_2$ compounds with pressure development of T_R, T_C and T_f.

one may expect that parimagnetism does not occur for $x = 0.075$. The reorientation temperature T_R decreases as well, due to increased structural disorder.

Upon application of hydrostatic pressure, all the characteristic temperatures shift in a bit different way. As it is shown in the Fig. 2 the characteristic temperatures shift in the same sense for all concentration of Si.

T_C and T_f shift to lower temperatures with increasing pressure in a similar manner, suggesting that the hierarchy of exchange interactions responsible for controlling inset of parimagnetism and ferrimagnetism is the same for fixed $Ho(Co_{1-x}Si_x)_2$ composition. The pressure coefficient $\partial T_C/\partial p$ slightly increases with increasing Si concentration.

Decreasing T_C with increasing pressure is usually explained as an influence of the pressure on the destabilization of Co magnetism and consequent weakening the RE-Co-RE exchange interaction. Analogous scenario can explain the shift of T_f. This temperature is also controlled by the RE-Co-RE exchange interaction [9]. The observed variations of T_R with Si doping and external pressure can be explained by the varying crystal field interaction. This, however, requires further efforts mainly by theoreticians.

4. Conclusions

We have studied the AC susceptibility, specific heat and electrical resistivity of $Ho(Co_{1-x}Si_x)_2$ compounds and determined variations of the characteristic temperatures for magnetism with alloying (Si replacing Co) and applying hydrostatic pressure. Si doping leads to increase (decrease) of T_C (T_f) and consequently to possible ceasing parimagnetism for $x = 0.075$. Pressure suppresses T_C and T_f simultaneously, reflecting that both the characteristic temperatures are controlled by the RE-Co-RE exchange interaction.

Acknowledgments

The experiments were performed in MLTL (http://mltl.eu/), which is supported within the program of Czech Research Infrastructures (project no.

LM2011025). The work was also supported by the Czech Science Foundation (project No. P204/12/0692).

References

[1] D. Bloch, R. Lemaire, *Phys. Rev. B* **2**, 2648 (1970).

[2] J. Herrero-Albillos, F. Bartolomé, L.M. García, A.T. Young, T. Funk, J. Campo, G. J. Cuello, *Phys. Rev. B* **76**, 094409 (2007).

[3] C.M. Bonilla, I. Calvo, J. Herrero-Albillos, A.I. Figueroa, C. Castam-Guerrero, J. Bartolomé, J.A. Rodriguez-Velamazan, D. Schmitz, E. Weschke, D. Paudyal, V.K. Pecharsky, K.A. Gschneidner Jr., F. Bartolomé, L.M. García, *J. Appl. Phys.* **111**, 07E315 (2012).

[4] C.M. Bonilla, J. Herrero-Albillos, A.I. Figueroa, C. Castam-Guerrero, J. Bartolomé et al., arXiv/1302.0775 (2013).

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

[6] K. Murata, H. Yoshino, H. Om Yadav, Y. Honda, N. Shirakawa, *Rev. Sci. Instrum.* **68**, 2490 (1997).

[7] E. Gratz, *Solid State Commun.* **48**, 825 (1983).

[8] T.D. Cuong, *Magnetism and related properties of* $RECo_2$ *compounds: Co-dilution*, Ph.D. thesis, Charles University in Prague (1998).

[9] O. Syshchenko, T. Fujita, V. Sechovský, M. Diviš, H. Fujii, *J. Alloys Comp.* **317-318**, 438 (2001).