Magnetic behavior in TmCu$_2$Ge$_2$

A. P. Pikul$^1$, L. Gondek$^2$, D. Kaczorowski$^1$, A. Szytula$^3$

$^1$ Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P Nr 1410, 50–950 Wroclaw 2, Poland
$^2$ Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, Mickiewicza 30, 30-059 Krakow, Poland
$^3$ Institute of Physics, Jagiellonian University, Reymonta 4, 30-059 Krakow, Poland

E-mail: lgondek@agh.edu.pl

Abstract. Basic physical properties of the compound TmCu$_2$Ge$_2$, crystallizing with the tetragonal ThCr$_2$Si$_2$-type crystal structure, were studied down to 350 mK. The temperature dependencies of the magnetic susceptibility, the specific heat and the electrical resistivity exhibit distinct anomalies at $T_N = 3.8$ K that can be associated with the onset of an antiferromagnetic ordered state. Above this temperature the reciprocal susceptibility obeys the Curie–Weiss law with the paramagnetic Curie temperature $\theta_p$ equal to $1.2$ K and the effective magnetic moment $\mu_{eff}$ equal to $7.12 \mu_B$. In the same region, the electrical resistivity shows a metallic character. In the ordered state, another clear peak in the specific heat is observed at $T_t = 1.7$ K that likely manifests a change in the magnetic structure.

1. Introduction

The ternary rare earth RT$_2$X$_2$ intermetallic compounds (R = rare earth, T = 3d, 4d or 5d transition metal and X = Si, Ge), crystallizing mostly in body-centered tetragonal structure of the ThCr$_2$Si$_2$-type (space group $I4/mmm$), exhibit a wide range of electronic and magnetic properties [1, 2]. Among them the phases RCu$_2$Ge$_2$ with R = Ce, Pr, Nd, Eu, Gd, Tb, Dy, Ho and Er are antiferromagnets at low temperatures, and the Tm-based compound was reported to be paramagnetic down to 4.2 K [3]. In this work we report the results of magnetic, electrical resistivity and specific heat measurements of TmCu$_2$Ge$_2$, which demonstrate that actually the ground state in this compound is also antiferromagnetic.

2. Experimental details

Polycrystalline sample of TmCu$_2$Ge$_2$ was prepared by arc melting stoichiometric amounts of the constituent elements (Tm of 99.9 % purity, Cu and Sn of 99.99 % purity) under high-purity argon atmosphere and subsequent annealing in an evacuated quartz tube at 800°C for one week. Quality of the product was checked by X-ray powder diffraction at room temperature using a Philips PW-3710 XPERT diffractometer with CuK$\alpha$ radiation. Evaluation of the diffraction pattern revealed, that the sample exhibits the expected tetragonal crystal structure of the ThCr$_2$Si$_2$-type, with the lattice parameters close to those given in the literature [1]. No impurities were detected in the sample examined.

The magnetic properties were studied in the temperature range 1.7 – 400 K and in external magnetic fields up to 5 T employing a Quantum Design MPMS-5 SQUID magnetometer.
heat capacity and the electrical resistivity were measured by relaxation method and ac four-point technique, respectively. The latter experiments were carried out from room temperature down to 350 mK utilizing a Quantum Design PPMS platform.

3. Results

The magnetic data of TmCu$_2$Ge$_2$ are presented in Fig. 1. Above about 10 K, the magnetic susceptibility obeys the Curie–Weiss law with the paramagnetic Curie temperature $\theta_p = 1.2$ K and the effective magnetic moment $\mu_{\text{eff}} = 7.12$ $\mu_B$. The experimental value of $\mu_{\text{eff}}$ is somewhat smaller than the free ion value (7.56 $\mu_B$). The positive sign of the $\theta_p$ might suggest ferromagnetic interactions between Tm magnetic moments to be dominant. However, a distinct maximum in $\chi(T)$ at $T_N = 3.8$ K clearly manifests the onset of antiferromagnetic ordering at lower temperatures (see the upper inset to Fig. 1). The magnetization measurements taken at 1.72 K as a function of the magnetic field strength corroborated the antiferromagnetic character of the ground state in TmCu$_2$Ge$_2$, revealing a metamagnetic-like anomaly in $\sigma(B)$ near 0.2 T and saturation of the $\sigma(B)$ curve in strong fields (see the lower inset to Fig. 1). The magnetic moment reached in the field-induced ferromagnetic state is 3.2 $\mu_B$ per Tm ion, which is only half the free Tm$^{3+}$ ion value (7 $\mu_B$).

Figure 1. Temperature dependence of inverse magnetic susceptibility $\chi^{-1}$ of TmCu$_2$Ge$_2$. The solid line represents the Curie–Weiss fit. The upper inset shows a low temperature part of $\chi(T)$; the arrow marks the Néel temperature $T_N$. The lower inset: isothermal magnetisation $\sigma$ measured as a function of increasing and decreasing magnetic field $B$ (closed and open symbols, respectively).

Figure 2. Electrical resistivity $\rho$ of TmCu$_2$Ge$_2$ as a function of temperature. The upper inset displays low-temperature part of $\rho(T)$; the arrows indicate the antiferromagnetic ordering temperature $T_N$ and spin-rearrangement temperature $T_s$. 
As shown in Fig. 2, TmCu$_2$Ge$_2$ exhibits metallic electrical conductivity with rather large value of the room temperature resistivity but also quite large residual resistivity ratio, as for a polycrystalline sample. In the temperature range 100 – 300 K, the electrical resistivity increases quasi-linearly with rising the temperature. At lower temperatures the $\rho(T)$ curve slightly rolls over, likely because of crystal field effect. A significant drop of the resistivity at $T_N$ 3.8 K manifests an onset of the magnetically ordered state. As apparent from the inset to Fig. 2, another anomaly at $T_t$ = 1.7 K is visible. The latter feature probably indicates another phase transition of magnetic nature, e.g. a kind of rearrangement of the antiferromagnetic structure.

Fig. 3 displays the specific heat of TmCu$_2$Ge$_2$ measured at low temperatures. Two distinct anomalies are observed at $T_N$ = 3.8 and $T_t$ = 1.7 K, in perfect agreement with the electrical resistivity data. The former lambda-like peak can be associated with the magnetic ordering of the Tm magnetic moments, while the second one may be attributed to the supposed spin reorientation. Unusually high ratio $C/T$ may hint at presence of another anomaly below 350 mK.

4. Summary
The results presented in this work show that the tetragonal TmCu$_2$Ge$_2$ compound exhibits antiferromagnetic ordering at about 3.8 K, followed by another distinct phase transition at 1.7 K, manifesting presumably some reorientation of the Tm magnetic moments.

Acknowledgments
This work was done within the frame of the National Scientific Network "Strongly correlated materials: preparation, fundamental research and applications".

References
[1] Szytula A and Leciejewicz J 1989 Handbook on the Physics and Chemistry of Rare Earths vol 12, ed Gschneider K A Jr and Erwin L (Amsterdam: Elsevier) p 133
[2] Gignoux D and Schmitt D 1997 Handbook of Magnetic Materials vol 10, ed Buschow K H J (Amsterdam: Elsevier) p 239
[3] Kotsanidis P A and Yakinthos J K 1981 Solid State Commun. 40 1041