# Evidence of the Non-Magnetic Ordering in $TmRu_2Si_2$ at Low Temperatures

A. Szytuła[a], A. Hoser[b], S. Baran[a] and B. Penc[a,*]

[a]M. Smoluchowski Institute of Physics, Jagiellonian University, S. Łojasiewicza 11, PL–30348 Kraków, Poland

[b]Helmholtz Centrum Berlin für Materialen and Energie GmbH, Hahn-Meitner Pl. 1, D–14109 Berlin, Germany

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The neutron powder diffraction measurements of the $TmRu_2Si_2$ compound in the temperature range 0.47–2.5 K have been performed. The obtained results confirm that this compound in low temperature has a tetragonal $ThCr_2Si_2$-type crystal structure (space group $I4/mmm$). The long range magnetic ordering was not detected up to 0.47 K.

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## 1. Introduction

$TmRu_2Si_2$ compound crystallizes in a tetragonal $ThCr_2Si_2$-type crystal structure [1]. In contrast to other $RRu_2Si_2$ (R = rare earth element) compounds which are antiferromagnets [2, 3], the magnetic properties of $TmRu_2Si_2$ are contradictory. The first magnetic data indicate the ferromagnetic order near 1 K [3] while the new complex measurements (magnetic, specific heat and electrical resistivity) indicate the paramagnetic behaviour down to 0.3 K [4]. The specific heat data (see inset in Fig. 3 in Ref. [4]) give the small intensity maximum at 0.6 K.

For the explanation of these differences the neutron diffraction experiment in mK temperature was performed.

## 2. Experimental

The experiment was performed on the sample whose preparation is described in Ref. [4]. The X-ray analysis at room temperature indicates that the sample has a tetragonal crystal structure with the lattice parameters $a = 4.138(4)$ Å and $c = 9.469(10)$ Å which are in good agreement with those reported in Ref. [4].

The neutron diffraction patterns are determined at temperatures 0.47, 0.6, and 2.5 K with the use of E6 diffractometer at BER II reactor (Helmholtz-Zentrum, Berlin). Low temperatures are obtained using a ILL-type cryostat with a $^3$He inset. The powdered samples were enclosed in a cylindrical copper container with a diameter of 8 mm. For better thermal contact a small amount of deuterated methanol was dropped into the container. The incident neutron wavelength was 2.443 Å. The data were analyzed using the Rietveld-type program Full-Prof [5].

*corresponding author; e-mail: boguslaw.penc@uj.edu.pl

## 3. Results and discussion

Typical diffraction pattern measured at 2.5 K and the differential patterns: 0.47–2.5 K and 0.6–2.5 K are shown in Fig. 1. The strong intensity peaks correspond to Cu container while the small intensity ones correspond to

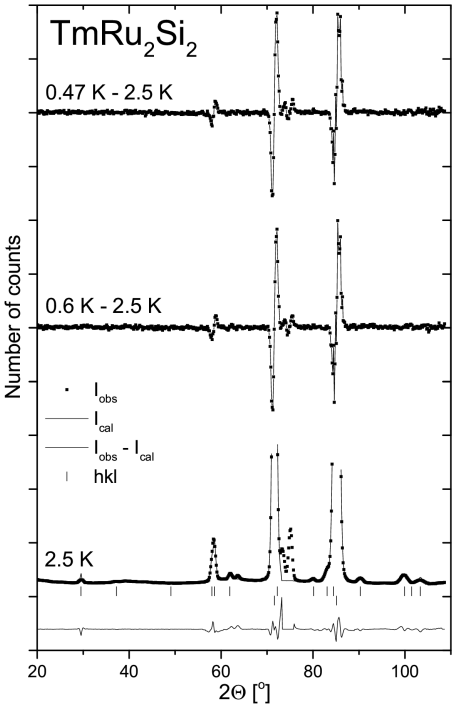


Fig. 1. Neutron diffraction pattern of $TmRu_2Si_2$ collected at 2.5 K. The squares represent the experimental points and the solid line through the data is the calculated profile for the crystal structure model as described in the text. The vertical bars indicate the Bragg peaks of nuclear origin (first row) and Cu (second row). Some region is not taken into account (cryostat). The difference between the observed and calculated intensities is shown by the solid line at the bottom of the panel. The difference patterns 0.47–2.5 K and 0.6–2.5 K are presented on the top.

$TmRu_2Si_2$ sample and the cryostat (Al). Analysis of the peaks connected with the investigated sample confirm the tetragonal structure (space group $I4/mmm$) in which the atoms occupy the following positions: Tm atoms at 2a site (0,0,0), Ru atoms in 4d site (0,1/2,1/4) and Si atoms in 4c site (0,0,$z$). Determined values of lattice parameters $a$ and $c$, $a/c$ ratio, unit cell volume $V$ and positional parameter $z$ are listed in Table I.

TABLE I

Crystal structure parameters of $TmRu_2Si_2$.

| $T$ [K] | $a$ [Å] | $c$ [Å] | $c/a$ | $V$ [Å$^3$] | $z$ | $R_{\mathrm{Bragg}}$ | $R_{\mathrm{RF}}$ |
|---|---|---|---|---|---|---|---|
| 0.47 | 4.108(1) | 9.384(4) | 2.285 | 158.33(10) | 0.373(2) | 8.8 | 8.3 |
| 0.6 | 4.107(1) | 9.382(4) | 2.284 | 158.29(9) | 0.373(2) | 8.6 | 8.7 |
| 2.5 | 4.107(1) | 9.393(6) | 2.287 | 158.41(13) | 0.374(3) | 8.5 | 8.9 |
| 293 | 4.138(4) | 9.469(10) | 2.288 | 162.14(47) | 0.377(3) | 4.9 | 5.1 |

Presented in the work data indicate that the change of the crystal structure and existence of the magnetic order at low temperatures are not observed. In majority of $TmT_2Si_2$ compounds the Tm moments order at low temperatures [6]. Absence in Tm sublattice of magnetic order is detected in isostructural $TmCr_2Si_2$ [7] in which the electrical crystal field (CF) indicates a non-magnetic singlet as a ground state.

The obtained data indicate the large decrease of the unit cell volume between 0.47 K and 300 K equal to 3.81 Å$^3$ which corresponds to the ratio $\Delta V/V = 2.35\%$. This decomposition is larger than that in isostructural $TmCu_2Si_2$ (0.7%) [8] and $TmNi_2Ge_2$ (1.75%) [9].

The anomaly is hard to understand. Some explanation can give the comparison of the physical parameters for ordered and non-ordered compounds:

- quadrupole interaction ($\Delta E_Q$) at low temperature is bigger in compounds with magnetic ordering compound, for example 140 mm/s in $TmCu_2Si_2$ [10] than in non-magnetic compounds $TmCr_2Si_2$ (70 mm/s) [7];
- energy gap between the ground state and first excited CF level is equal to 6.09 K in $TmCr_2Si_2$ [11] and nearly 20 K in $TmRu_2Si_2$ [4] and 13 K in $TmCr_2Si_2$ [7].

## 4. Conclusion

Above data suggest that anomalous properties of $TmRu_2Si_2$ are connected with the electronic properties of those compounds.

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## References

[1] Z. Ban, M. Sikirica, *Acta Crystallogr.* **18**, 594 (1965).
[2] I. Felner, I. Nowik, *J. Phys. Chem. Solids* **45**, 419 (1984).
[3] K. Hiebl, C. Horvath, P. Rogl, *J. Magn. Magn. Mater.* **73**, 287 (1983).
[4] L. Gondek, D. Kaczorowski, A. Szytula, *Solid State Commun.* **149**, 2181 (2009).
[5] J. Rodriguez-Carvajal, *Physica B* **192**, 55 (1993).
[6] A. Szytula, in: *Handbook of Magnetic Materials*, Ed. K.H.J. Buschow, Vol. 6, North Holland Publ. Co., Amsterdam 1991, Ch. 2, p. 85.
[7] B. Saensunon, G.A. Stewart, K. Nishimura, *J. Alloys Comp.* **476**, 49 (2009).
[8] W. Schlabitz, J. Baumann, G. Neumann, D. Plumacher, K. Reggentin, *Crystalline Electric Field Effects in F-Electron Magnetism*, Eds. P. Guertin, W. Suski, Z. Żołnierek, Plenum Press, New York 1982, p. 289.
[9] J.K. Yakinthos, *J. Magn. Magn. Mater.* **99**, 123 (1991).
[10] G.A. Stewart, J. Żukrowski, in Ref. [8], p. 319.
[11] Y. Takeda, N.D. Dung, Y. Nakano, T. Ishikura, S. Ikeda, T.D. Matsuda, E. Yamamoto, Y. Haga, T. Takeuchi, R. Settai, Y. Ōnuki, *J. Phys. Soc. Japan* **77**, 104710 (2008).