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Neutron diffraction studies of pseudoternary $TbRu_{2-x}Pd_xSi_2$

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Abstract

The $TbRu_{2-x}Pd_xSi_2$ series of solid solutions ($x = 0.25$, 0.5, 0.75, 1.0, 1.5 and 1.75) was studied by neutron powder diffraction. They crystallize in the body-centered tetragonal $ThCr_2Si_2$-type structure. Analysis of the magnetic component of the neutron diffraction patterns shows the change of the magnetic structure from a sine-modulated LSW II-type for $x \leq 1.0$ to a short-range magnetic order for $x = 1.5$ and a sine-modulated LSW IV-type for $x = 1.75$. In the LSW II-type structure the Tb-moments are parallel to the *c*-axis while for the LSW IV-type they are parallel to the *a*-axis. With increasing *x* a sharp decrease in the Néel temperature is observed. For $0.75 \leq x < 1.0$ two magnetic phases coexist.

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1. Introduction

The compounds with the general formula RT_2X_2 (R is rare earth, T is transition d-electron element and X is Si or Ge) which crystallize in the body-centered tetragonal $ThCr_2Si_2$-type structure (space group *I*4/*mmm*) still attract special interest because of the wide variety of physical properties going along with the simplicity of the crystal structure. The neutron diffraction data indicate that at low temperatures the rare earth magnetic moments form a wide variety of magnetic structures [1]. The type of the magnetic structure is correlated with the T-element. To determine the influence of the T-element on the type of magnetic ordering, systematic investigations of the magnetic properties of the pseudoternary $RT_{2-x}T_x'Si_2$ systems were carried out. This work reports the results for $TbRu_{2-x}Pd_xSi_2$ series. Complementary investigations are reported in Refs. [2–4]. The parent compounds have modulated magnetic structures described by different propagation vectors: $\mathbf{k} = (k_x, 0, 0)$, where $k_x = 0.233$ for $TbRu_2Si_2$ [5,6] and $\mathbf{k} = (0, k_y, k_z)$ where $k_y = 0.4057$ and $k_z = 0.1671$ for $TbPd_2Si_2$ [7].

2. Experiment details and results

All specimens were obtained by means of induction melting followed by a solid state diffusion process. Detailed description of sample preparation is given in Ref. [4]. X-ray diffraction was used to check the purity of the samples. Neutron diffraction patterns at several selected temperatures were collected using the E6 diffractometer ($\lambda \approx 2.4$ Å) at the BERII reactor of the BENSC, Hahn-Meitner Institute, Berlin, Germany. The Rietveld-type profile-refinement program FULLPROF [8] was used to refine the structural and magnetic parameters simultaneously.

Powder neutron-diffraction pattern collected in the paramagnetic region confirms the body-centered tetragonal $ThCr_2Si_2$-type crystal structure of the investigated samples. The R atoms occupy the 2a site: 0, 0, 0, Ru and Pd atoms occupy the 4d site: 0, 1/2, 1/4 and Si atoms occupy the 4e site: 0, 0, *z*. The analysis of the nuclear peaks intensities suggests that the Pd atoms are statistically distributed at the 4d site. The determined *a* and *c* lattice parameters, the unit cell volume *V* and the positional parameter of Si atoms z_{Si} in the paramagnetic and magnetically ordered (1.5 K) region are plotted in Fig. 1. Both *c* and z_{Si} parameters increase with increasing Pd content *x* while the *a*-parameter decreases.

The changes in the magnetic structure are studied by powder neutron diffraction. The low angle part of the neutron diffraction patterns collected for the $TbRu_{2-x}Pd_xSi_2$ series of solid solutions ($x = 0.25$, 0.75, 1.0, 1.5 and 1.75) at low temperatures is shown in Fig. 2. For $x = 0.25$ the distribution of the magnetic peaks is similar to those observed for $TbRu_2Si_2$ [5,6]. Magnetic order is a sine-modulated one described by the propagation vector $\mathbf{k} = (k_x, 0, 0)$ with $k_x = 0.266(1)$. The magnetic moment equal 8.12(11) μ_B is parallel to the *c*-axis. Temperature dependence of the $000^{\pm}$ peak intensity (at $2\theta = 9.2^{\circ}$) gives the Néel temperature equal 45 K. In the neutron pattern of $TbRu_{1.25}Pd_{0.75}Si_2$ the peak $000^{\pm}$ of the magnetic origin (about $2\theta = 10^{\circ}$) has a complex structure and con-

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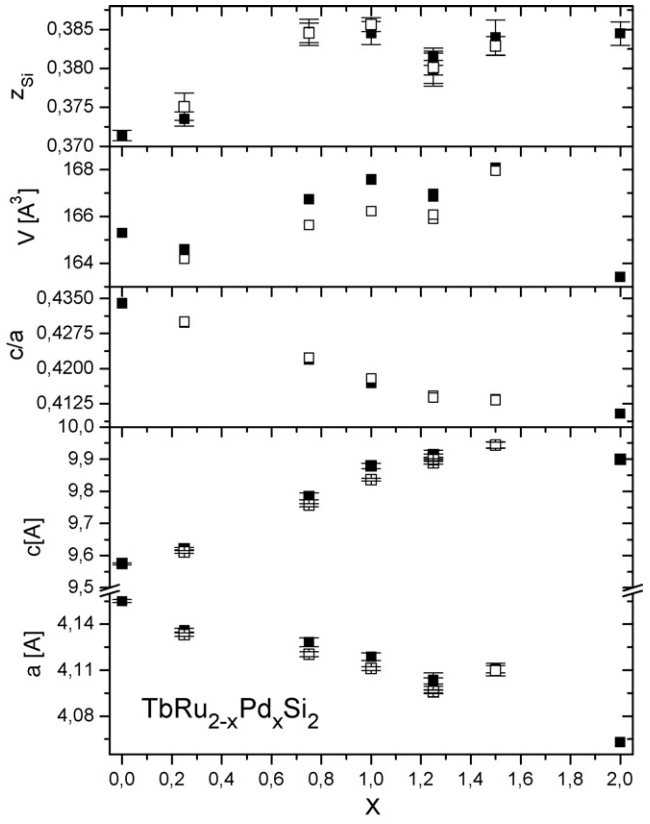

Fig. 1. Concentration dependence of the a and c lattice parameters, a/c ratio, unit cell volume V and z_{Si} positional parameter of Si atoms for $TbRu_{2-x}Pd_xSi_2$ in the paramagnetic (solid symbols) and magnetically ordered state (open symbols). Error bars are marked if bigger than the appropriate symbol.

sists of two peaks with the maxima at 10° and 12°. This suggests that in this case the magnetic ordering is described by two sine-modulated structures with the propagation vectors $\mathbf{k} = (k_x, 0, 0)$ equal 0.299(1) and 0.333(1) and Tb magnetic moments parallel to the c-axis and equal 4.0(1) and 4.6(1) μ_B, respectively. The temperature dependence of the magnetic peaks intensities gives the Néel temperature 27 K.

Similar sine-modulated magnetic ordering, with $k_x = 0.355(1)$ and the small value of the magnetic moment equal 3.82(8) μ_B, is observed for $x = 1.0$. About $2\theta = 20°$ a broad maximum of small intensity forms below 15 K. Thermal dependence of the $000^{\pm}$ peak intensity gives the Néel temperature equal 22 K.

For $x = 1.5$ a broad peak about 2θ equal 20° is well developed below 15 K. This indicates that only short-range order similar to those observed in the parent compound $TbPd_2Ge_2$ [7] exists in this sample.

The neutron pattern of $TbRu_{0.25}Pd_{1.75}Si_2$ is similar to the one observed for $TbPd_2Si_2$ [7]. Magnetic peaks were indexed by the propagation vector $\mathbf{k} = (0, 0.403(1), 0.149(1))$ which is close to the one reported for $TbPd_2Si_2$. The terbium magnetic moments equal 5.8(1) μ_B at 1.5 K are parallel to the a-axis and form a sine-modulated structure in which the magnetic moment at (1/2, 1/2, 1/2) is antiparallel to that at (0, 0, 0). Temperature dependence of the magnetic peaks intensities give the Néel temperature equal 14 K. A diffuse magnetic peak corresponding to a short-range order is observed above T_N.

3. Discussion and summary

The data presented in this work confirm that the $TbRu_{2-x}Pd_xSi_2$ solid solutions exist in the whole range of Pd content ($0 \leq x \leq 2$) and crystallize in the tetragonal crystal structure of

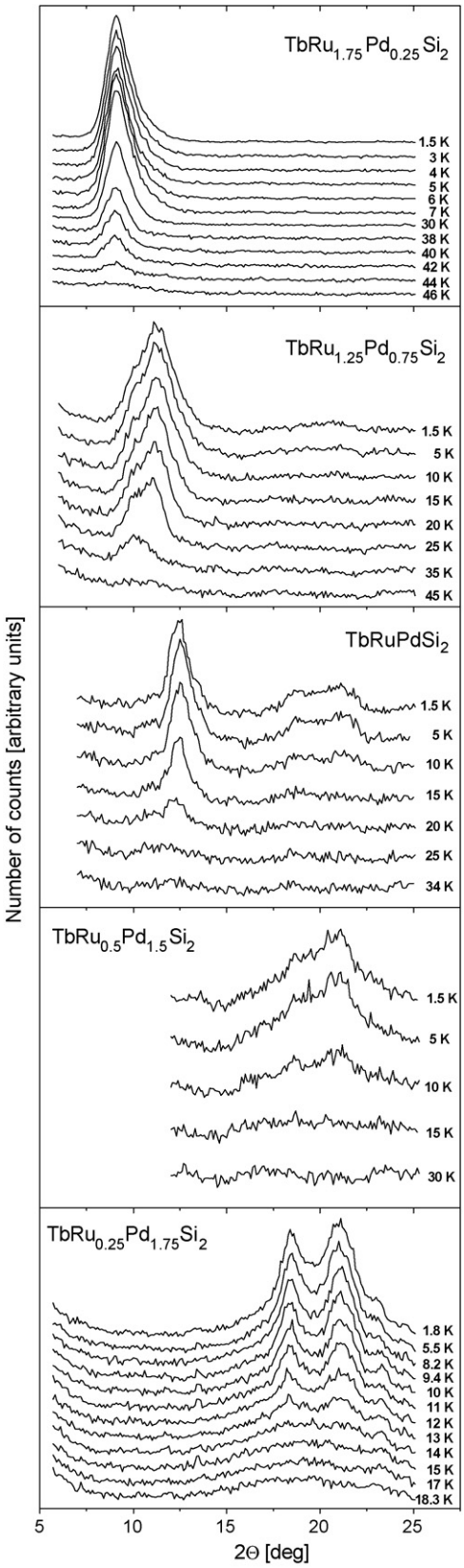

Fig. 2. The low angle part of the neutron diffraction patterns of $TbRu_{2-x}Pd_xSi_2$ ($x = 0.25$, 0.75, 1.0, 1.5 and 1.75) at different temperatures.