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Magnetic Order in TbPdIn

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The magnetic structure in TbPdIn was investigated by neutron diffraction experiments. The results reveal a long-range magnetic structure with propagation vector $(0,0,0)$ and the Tb moments forming a non-collinear structure within the basal plane that is unchanged over the whole temperature region below the ordering temperature of 66 K.

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1. Introduction

TbPdIn belongs to a large family of ternary RTX (R = rare earth, T = transition metal, X = *p*-metal) compounds crystallizing in the hexagonal ZrNiAl-type structure. Several contradictory results have been reported concerning the magnetic properties of TbPdIn. All the previous studies showed clearly that TbPdIn orders magnetically below $T_{\text{ord}} = 66$ K, but the nature of the ordered state was unclear. A possible spin-glass state, which could arise from the spin frustration, was first speculated from the temperature dependence of zero-field-cooled and field-cooled magnetization [1]. On the other hand, powder neutron diffraction measurements revealed a definite 3D magnetic structure described by the $(0,0,0)$ propagation vector [2]. The details of the magnetic structure however were not determined from the powder pattern. Further magnetization and susceptibility studies corroborated the long-range magnetic order in TbPdIn [3, 4], and the magnetization data reported in Ref. [4] indicated furthermore additional magnetic phase transitions at 27 and 6 K, previously not reported in other studies. These transitions were tentatively related to the weak magnetic reflections observed in the neutron powder pattern beside the main magnetic reflections [2]. The unresolved details of the magnetic structure and disagreement between different bulk measurements led us to perform a detailed investigation by new powder neutron diffraction and single-crystal neutron Laue diffraction.

2. Experimental

TbPdIn single crystal used for the Laue diffraction was grown by a Czochralski method in a tri-arc furnace, the polycrystalline sample used for the powder diffraction was prepared in a mono-arc furnace.

The powder diffraction experiment was performed at the Institut Laue–Langevin on the D1B instrument using a wavelength of 2.52 Å. We collected diffraction patterns with a very good counting statistics at 80 and 1.6 K. Additionally, diffraction patterns with lower statistics were measured during continuous heating from 2 to 80 K to map the temperature development. The measured data were analyzed by the Rietveld refinement method, using the FullProf program [5].

A neutron Laue diffraction experiment on a TbPdIn crystal was performed on the VIVALDI instrument at ILL, Grenoble [6]. The crystal dimensions were approximately $1\times1\times1$ mm^3. The Laue patterns were recorded at 2, 10, 40 and 100 K to cover all magnetic phases predicted in Ref. [4]. In order to observe all types of reflections and discover any possible purely magnetic intensities, the crystal was mounted with obvious symmetry axes well away from the vertical axis and six patterns at 15 degree intervals of rotation about the vertical axis were taken at each temperature.

3. Results and discussion

The powder diffraction pattern at 1.6 K (see Fig. 1) confirmed the magnetic structure described by the $(0,0,0)$ propagation vector. The best agreement with the observed data is obtained considering the magnetic structure shown in Fig. 2 (agreement factor $R_{\text{mag}} = 5.6$, compared with $R_{\text{B}} = 4.0$ at 80 K). The Tb moments reach the value of $\mu_{\text{Tb}} = 7.8(3)$ μ_{B} at 1.6 K. The small reduction compared to the free-ion value of 9 μ_{B} can be ascribed to the crystal-field effect. The same magnetic structure was proposed by Ehlers for TbNiIn [7].

Several additional peaks, indicated by arrows in Fig. 1, appear in the 1.6 K pattern, similar to previous study [2]. These reflections cannot be indexed by a single propagation vector. To clarify their nature, the neutron Laue diffraction experiment was performed. As expected, the intensities increase below $T_{\text{ord}} = 66$ K, indicating mag-

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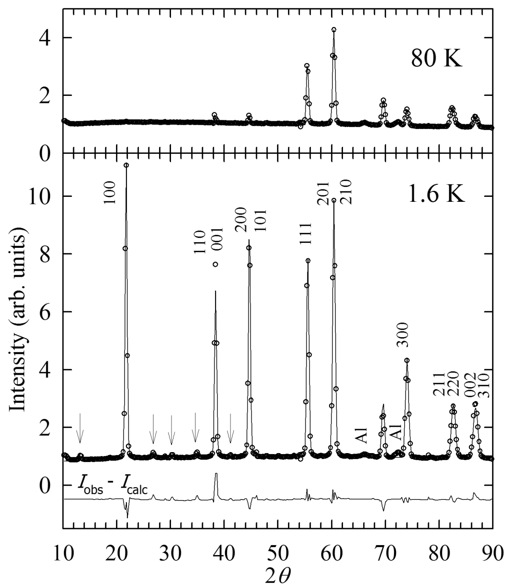

Fig. 1. Neutron powder diffraction patterns of TbPdIn at 80 K and 1.6 K. The full line at 1.6 K pattern corresponds to the magnetic structure shown in Fig. 2, the bottom line is the difference between the calculated and observed pattern. The arrows indicate magnetic peaks of the impurity phase (see text).

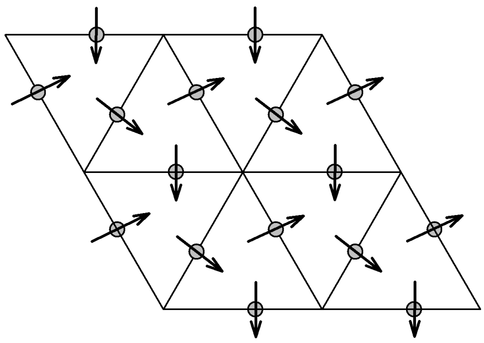

Fig. 2. Proposed magnetic structure in TbPdIn.

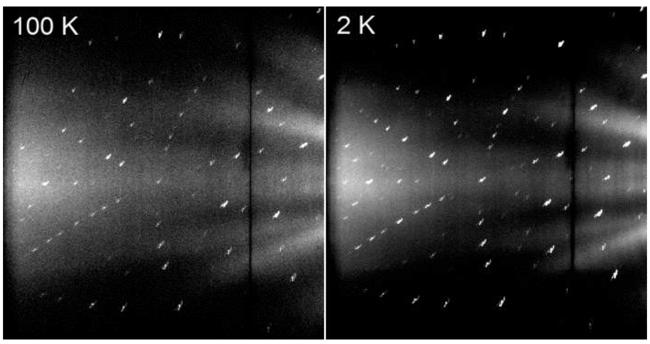

Fig. 3. Part of the Laue diffraction patterns taken at 100 and 2 K. All the observed spots at 2 K can be indexed with integer (hkl) in the hexagonal symmetry.

netic ordering characterized by the $(0, 0, 0)$ propagation vector (see Fig. 3). Detailed comparison of the Laue patterns taken above and below T_{ord} did not reveal any additional diffraction spots at any measurement temperature. We made a considerable effort to detect any possible weak diffraction spots of purely magnetic origin (with non-integer hkl) taking highly overexposed patterns at 2 K, but no such spots were found. It means that the additional magnetic intensities observed in the powder experiment are most probably due to a magnetic impurity phase.

The temperature dependence of the intensities, as observed in both powder and Laue diffraction, did not reveal any indications of further magnetic phase transitions below T_{ord}. All the intensities increase monotonously with decreasing temperature in a similar way. We believe that the phase transitions reported in Ref. [4] at 27 and 6 K are also due to impurity phases. Such a conclusion is also corroborated by the fact that other bulk measurements [1, 3] did not show these transitions.

4. Conclusion

In conclusion, our results confirm the long-range magnetic structure with the $(0, 0, 0)$ propagation vector and reveal the non-collinear magnetic structure with Tb moments within the basal plane which remains unchanged in the whole temperature region below the ordering temperature.

Acknowledgments

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References

[1] S. Nishigori, Y. Hirooka, T. Ito, *J. Magn. Magn. Matter.* **177-181**, 137 (1998).

[2] P. Javorský, P. Svoboda, S. Nishigori, M. Hofmann, *Mater. Sci. Forum* **321-324**, 705 (2000).

[3] D.X. Li, Y. Shiokawa, T. Nozue, T. Kamimura, K. Sumiyama, *J. Magn. Magn. Mater.* **241**, 17 (2002).

[4] M. Bałanda, A. Szytuła, M. Guillot, *J. Magn. Magn. Mater.* **247**, 345 (2002).

[5] J. Rodriguez-Carvajal, *Physica B* **192**, 55 (1993).

[6] G.J. McIntyre, M.-H. Lemée-Cailleau, C. Wilkinson, *Physica B* **385-386**, 1055 (2006).

[7] G. Ehlers, Ph.D. Thesis, Technische Universität Berlin, 1996.