

Magnetic Phase Transitions in TbNi(Al,In) Compounds

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The magnetic phase transitions in TbNiAl_{1-x}In_x compounds were investigated by ac-susceptibility measurements. Our data reveal magnetic ordering with transition temperatures between 40 and 70 K, depending on the In concentration. All the studied compounds exhibit two or more phase transitions accompanied by distinct anomalies in the real and imaginary part of the ac-susceptibility. The paramagnetic Curie temperatures remain positive in the whole series.

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1. Introduction

The ternary RTX compounds (R = rare earth, T = transition *d*-metal, X = *p*-metal) crystallizing in the hexagonal ZrNiAl-type structure (space group $P\bar{6}2m$) form a large group of intermetallics. Most of these compounds order magnetically at low temperatures, often with complex magnetic structures (see e.g. [1–3]). Generally, the type of magnetic order is very sensitive to the number of conduction electrons and the development of magnetic properties in pseudo ternary series like R(Ni,Cu)Al is very complex [4]. On the other hand, magnetocrystalline anisotropy seems to be strongly influenced by the ratio of lattice parameters which is most noticeable in the Tb-based compounds showing the strongest anisotropy. To bring deeper information about the relation between magnetic anisotropy and crystal structure, the TbNi(Al,In) series seems to be ideal. The Al–In substitution is isoelectronic, so the changes of magnetic structure are primarily driven by the large change of the lattice parameters that occurs in this series [5, 6]. Both parent compounds have been previously studied. TbNiAl orders antiferromagnetically below $T_N = 47$ K and undergoes further magnetic phase transition at $T_1 = 23$ K [5]. One third of Tb moments is reduced to almost zero between T_N and T_1 , change of the propagation of the frustrated moments occurs below T_1 . The Tb magnetic moments are oriented parallel to the *c*-axis in both magnetic phases [5]. TbNiIn orders magnetically below $T_{\text{ord}} \approx 70$ K with additional magnetic phase transitions indicated by ac-susceptibility at $T_1 = 59$ K and $T_2 = 29$ K [6]. As inferred from neutron diffraction, the Tb moments lie within the basal plane

and form a non-collinear structure below the transition temperature T_{ord} , while an additional weaker component develops below T_2 [7]. We give a basic characterization of the magnetic properties of TbNi(Al,In) compounds in this paper.

2. Experimental

Polycrystalline TbNiAl_{1-x}In_x samples were prepared by arc-melting of pure elements in a mono-arc furnace under the protection of an argon atmosphere. Small excess of In was used ($\approx 5\%$) to compensate higher evaporation of In during the melting. The X-ray diffraction patterns obtained using Cu K_α radiation confirmed single-phase samples within ZrNiAl-type of structure and the energy dispersive X-ray (EDX) analysis confirmed homogeneous In–Al distribution. To determine the paramagnetic characteristics, magnetization was measured between 50 and 300 K in the field of 2 T. The temperature variation of the ac-susceptibility was measured during cooling between 80 and 3 K using the PPMS (Quantum Design) instrument. The data in 0 T were obtained for frequencies 13, 113, 1113 and 9993 Hz, the frequency of 9993 Hz was additionally used for measurements in magnetic fields of 0.05, 0.10, 0.20 and 0.50 T. Both magnetization and ac-susceptibility were measured on powder samples randomly fixed by a glue.

3. Results and discussion

In the paramagnetic region, the $M/H(T)$ dependences follow the Curie–Weiss behavior. The determined values of the effective moment, μ_{eff} , are close or slightly higher than the Tb³⁺ free ion value of $9.72 \mu_B$ (see Table). The small enhancement can be attributed to polarization of conduction electrons, similar to pure TbNiAl [5]. The

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paramagnetic Curie temperature, θ_p , is positive for all In concentrations and weakly increases for the In rich compounds. The positive θ_p in TbNiAl despite its antiferromagnetic structure points to prevailed ferromagnetic interactions in the paramagnetic region [5].

TABLE

Magnetic phase transition temperatures as determined from ac-susceptibility and the paramagnetic characteristics (not measured for $x = 0.0, 0.3, 0.7$ and 1.0) in TbNiAl $_{1-x}$ In $_x$ compounds.

x	T_{ord} [K]	T_1 [K]	T_2 [K]	μ_{eff} [μ_B]	θ_P [K]
0.0 [5]	47	23	–	10.4	30
0.0	46	24	18		
0.1	48	25	18	9.9	36
0.2	46	22	–	10.0	32
0.3	45	15	–		
0.4	42	12	–	10.1	34
0.5	42	12	–	9.6	34
0.6	45	14	–	9.9	36
0.7	47	20	–		
0.8	51	20	13	9.7	40
0.9	60	49	21	9.6	44
1.0	72	54	20		
1.0 [6]	70	59	29	9.5	55

The temperature dependence of ac-susceptibility reveals two or more phase transitions for all studied compounds. The anomalies related to the temperature of magnetic ordering are frequency independent which indicates the long-range magnetic order in the whole series, contrary to some R(Ni,Cu)Al compounds where the spin-glass state is manifested by frequency dependent signal [8]. The ac-susceptibility of Al rich compounds show qualitatively similar behaviour to the parent TbNiAl with a relatively sharp anomaly at T_N and less pronounced maximum around T_1 (see Fig. 1). With increasing the In content, the anomaly around the ordering temperature becomes broader and the low-temperature anomaly becomes more pronounced, especially in χ'' . Two maxima are observed at the low-temperature region at 20 and 13 K for $x = 0.8$, indicating more complex behaviour. The main maximum in the In rich compounds TbNiAl $_{0.1}$ In $_{0.9}$ and TbNiIn is preceded by certain shoulder at higher temperatures (see Fig. 2), which was observed also in the previous study on TbNiIn [6]. All the observed anomalies are summarized in Table. The magnetic field generally suppresses all the anomalies but causes also a shift of some of them to lower temperatures which could indicate the antiferromagnetic character (see Figs. 1 and 2).

We can conclude that TbNi(Al,In) compounds order magnetically with transition temperatures between 40 and 70 K, and show existence of at least two different magnetic phases. To investigate the magnetic structures

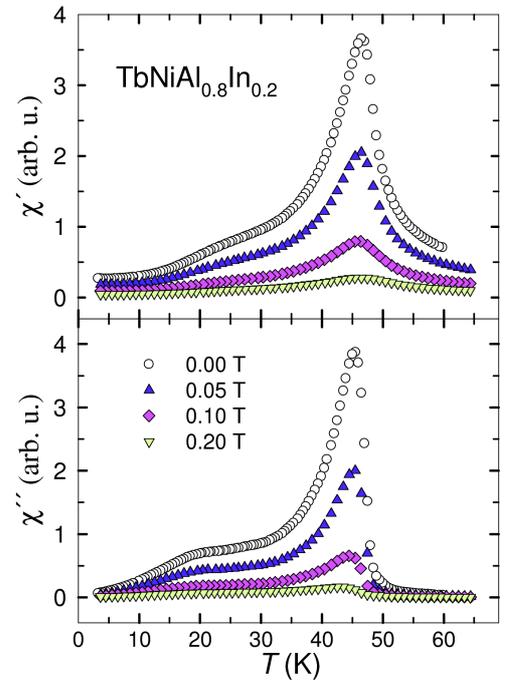


Fig. 1. Ac-susceptibility (real (a) and imaginary (b)) of TbNiAl $_{0.8}$ In $_{0.2}$ measured in different magnetic fields. The data obtained for 0 T were divided by a factor of 5×10^4 for better lucidity.

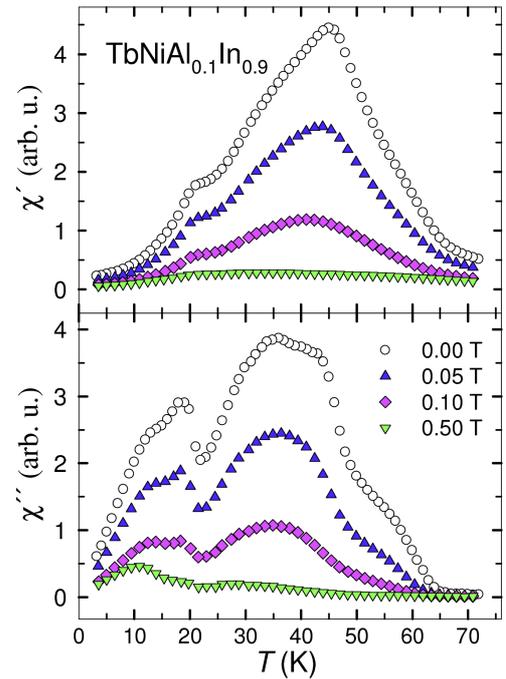


Fig. 2. Ac-susceptibility (real (a) and imaginary (b)) of TbNiAl $_{0.1}$ In $_{0.9}$ measured in different magnetic fields. The data obtained for 0 T were divided by a factor of 3×10^4 and the 0.5 T data were multiplied by a factor of 4 for better lucidity.

of individual phases, neutron diffraction study will be performed.

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References

- [1] G. Ehlers, H. Maletta, *Z. Phys. B* **101**, 317 (1996).
- [2] O. Garlea, E. Morosan, S.L. Bud'ko, J.L. Zarestky, P.C. Canfield, C. Stassis, *J. Appl. Phys.* **95**, 6921 (2004).
- [3] S. Baran, A. Arulraj, D. Kaczorowski, B. Penc, A. Szytuła, *Intermetallics* **18**, 42 (2010).
- [4] J. Prchal, T. Naka, M. Míšek, O. Isnard, P. Javorský, *J. Magn. Magn. Mater.* **316**, e499 (2007).
- [5] G. Ehlers, H. Maletta, *Z. Phys. B* **99**, 145 (1996).
- [6] Yu.B. Tyvanchuk, Ya.M. Kalyczak, Ł. Gondek, M. Rams, A. Szytuła, Z. Tomkiewicz, *J. Magn. Magn. Mater.* **277**, 368 (2004).
- [7] L. Gondek, A. Szytuła, S. Baran, J. Hernandez-Velasco, *J. Magn. Magn. Mater.* **272-276**, e443 (2004).
- [8] J. Prchal, E. Šantavá, D. Schmoranzer, *Physica B* **404**, 3056 (2009).