Proceedings of the European Conference "Physics of Magnetism" (PM'08), Poznań 2008

Magnetic Properties of $TbNi_{1-x}Au_xIn$ Compounds

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Polycrystalline samples of $TbNi_{1-x}Au_xIn$ for $x = 0.1$, 0.2, 0.4, 0.6 and 0.8 were prepared and studied by powder X-ray and neutron diffraction and ac magnetic susceptibility measurements. These compounds crystallize in the hexagonal ZrNiAl-type structure. With increasing Au content a change in the magnetic structure is observed. For $x = 0.1$, 0.2 and 0.4 the magnetic order is described by the propagation vector $\boldsymbol{k} = (1/2, 0, 1/2)$ while for $x = 0.8$ by $\boldsymbol{k} = (0, 0, 1/2)$. Between 1.5 K and the Néel temperature the magnetic order is stable.

PACS numbers: 71.20.Lp, 75.25.+z, 75.50.Ee

1. Introduction

TbNiIn and TbAuIn are isostructural compounds which crystallize in the hexagonal crystal structure of the ZrNiAl-type (space group $P\bar{6}2m$) [1, 2]. The distribution of the rare-earth atoms in the basal plane is similar to the *kagomé* lattice. In the case of antiferromagnetic coupling between nearest neighbor rare-earth magnetic moments frustration of the magnetic interactions is induced by this topology.

TbNiIn and TbAuIn have different magnetic properties. For TbNiIn magnetic and neutron diffraction data indicate complex magnetic properties. In the temperature range 1.5–32 K the magnetic order is described by two phases: a noncollinear antiferromagnetic phase of the 120°-type and a modulated one with the propagation vector $\boldsymbol{k} = (1/2, 0, 1/2)$. The modulated phase disappears at 32 K. At low temperatures the Tb magnetic moments of the noncollinear structure form the angle of 30(5)° with the $a-b$ plane while for the modulated one they lie in the $a-b$ plane [3, 4]. In TbAuIn a noncollinear antiferromagnetic ordering of the 120°-type was found below $T_N = 35$ K. The Tb-moments lie in the $a-b$ plane. Magnetic ordering is described by the propagation vector $\boldsymbol{k} = (0, 0, 1/2)$ [5].

This work reports the results of X-ray and neutron powder diffraction and magnetic measurements of the $TbNi_{1-x}Au_xIn$ solid solutions. The work has in view to determine the parameters of the crystal and magnetic structure as a function of Au content x.

2. Experimental

Polycrystalline samples of $TbNi_{1-x}Au_xIn$ for x equal to 0.1, 0.2, 0.4, 0.6 and 0.8 were prepared by arc melting of high-purity respective metals (Tb: 3N, Ni and Au: 4N and In: 5N) in a titanium gettered argon atmosphere. The samples were then annealed in evacuated vycor capsules at 600°C for 1 week.

X-ray powder diffraction patterns were recorded at room temperature using Cu K_α line with a Philips PW-3710 X'PERT type diffractometer.

The ac magnetic susceptibility was measured using a 7225 Lake Shore instrument. Both in-phase, χ', and out-of-phase, χ'', components of the $\chi_{ac} = \chi' - i\chi''$ are measured at the frequency $f = 125$ Hz. Neutron powder diffraction studies of the $TbNi_{1-x}Au_xIn$ compounds with x equal to 0.1, 0.2, 0.4 and 0.8 were performed on the E6 diffractometer installed at the Berlin Neutron Scattering Center, Hahn–Meitner Institute (Berlin). The measurements were carried out in the temperature range 1.5–60 K with the incident neutron wavelength 2.444 Å. The neutron diffraction data were analyzed using the Rietveld-type program FullProf [6].

3. Results

As mentioned in Introduction, the crystal structure of TbNiIn and TbAuIn compounds is described as the ZrNiAl-type (space group $P\bar{6}2m$) with the atoms occupying the following positions Tb in 3(g): x_{Tb}, 0, 1/2, In in 3(f): x_{In}, 0, 0 and Ni and Au in 1(b): 0, 0, 1/2 and 2(c): 1/3, 2/3, 0. The X-ray powder diffraction at room temperature and neutron diffraction patterns collected at paramagnetic state could be easily indexed assuming this type structure for all the investigated compounds. The determined a and c lattice constants and unit cell volume V increase with increasing Au content x. The a/c ratio shows an anomalous dependence with the maximum for

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$x = 0.4$ (Fig. 1). The value of the x_{Tb} and x_{In} parameters do not change with increasing Au content x. The Au atoms preferentially occupy the 2(c) positions for which larger atomic volume is characteristic.

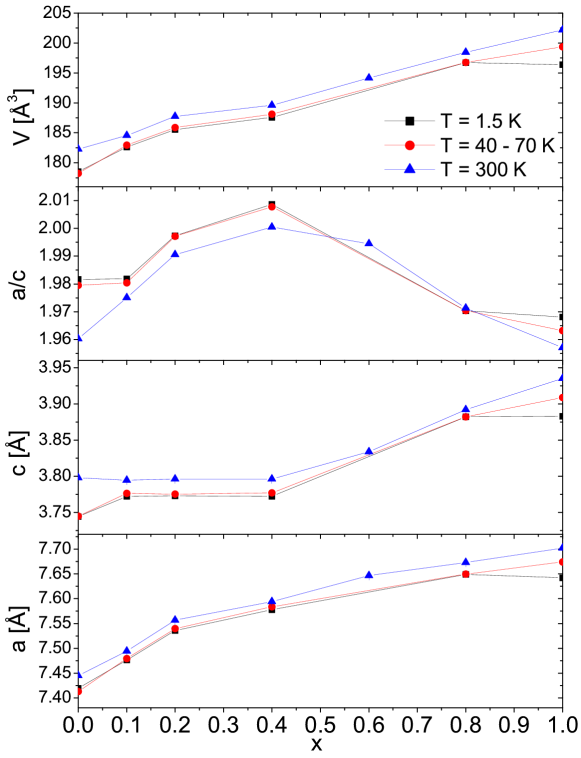

Fig. 1. Lattice parameters a and c, a/c ratio and unit cell volume V for $TbNi_{1-x}Au_xIn$ versus x for $0 \le x \le 1$ at $T = 1.5$ K (squares), 40–70 K (circles) and 300 K (triangles). The data for $x = 0$ and 1.0 are from Ref. [4].

The temperature dependence of the magnetic susceptibility, presented in Fig. 2, shows the change in the magnetic properties with changing Au content. The magnetic data give the following results:

— $x = 0.1$, temperature dependence of the real and imaginary part of the magnetic susceptibility χ' and χ'' indicates some transitions between 20 and 55 K,

— $x = 0.2$, temperature dependence of $\chi''(T)$ shows the maximum at T_N equal to 52 K and a small increase in the susceptibility at low temperatures. The out-of-phase component of the magnetic susceptibility indicates also the glass properties,

— $x = 0.4$, magnetic data show an anomaly corresponding to the Néel temperature at 41 K. The χ', χ'' and $d\chi'/dT$ indicate an additional phase transition at 20 K,

— $x = 0.6$ in $\chi'(T)$ and $\chi''(T)$ an anomaly at 37.4 is observed. These data are not shown because they are similar to those for $x = 0.8$,

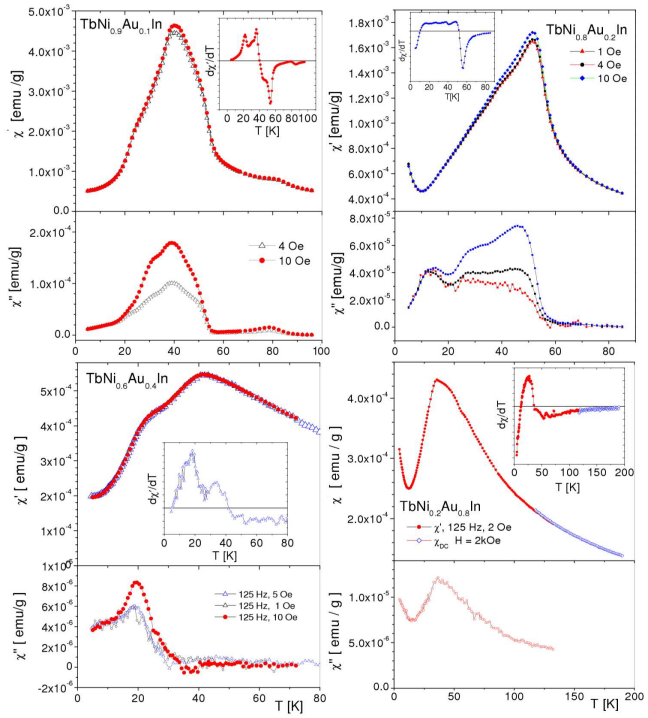

Fig. 2. Temperature dependence of χ' and χ'' for $TbNi_{1-x}Au_xIn$ for $x = 0.1$, 0.2, 0.4 and 0.8. The insets show the thermal dependence of the $d\chi'/dT$. The horizontal lines in the insets indicate the zero level on the $d\chi'/dT$ scale.

— $x = 0.8$, a maximum at 36 K in $\chi'(T)$ and $\chi''(T)$ is observed and a slight increase in the value at low temperatures.

TABLE

Magnetic data of $TbNi_{1-x}Au_xIn$ compounds. $T_{C,N}$ is the Curie or Néel temperature, T_t — temperature of the magnetic phase transition in the ordered state, μ — Tb magnetic moment determined from the neutron diffraction data.

x	$T_{C,N}$ [K]	T_t [K]	μ [μ_B]	Ref.
0	68	32	7.1(1)	[3, 4]
0.1	55		7.9(1)	this work
0.2	52		8.0(1)	this work
0.4	41	20	6.8(4)	this work
0.6	37.4		–	this work
0.8	36		6.6(3)	this work
1.0	35		7.8(1)	[5]

The magnetic data are summarized in Table.

The neutron diffraction data for $TbNi_{0.9}Au_{0.1}In$ measured at 1.5, 25, 36 K do not show any change in the magnetic structure. Similar neutron diffraction patterns are observed for $x = 0.2$ and 0.4 at 1.5 K. The peaks of magnetic origin are indexed by the propagation vector $\boldsymbol{k}$

= (1/2, 0, 1/2), similarly to those observed in TbNiIn [3, 4] and in some RTAl compounds [7]. Good agreement with the experimental results is observed for the noncollinear structure of the 120°-type with the Tb moments equal to 8.0(1) μ_B, 8.0(1) μ_B and 5.3(2) μ_B at 1.5, 25 and 36 K, respectively.

Neutron diffraction data for $TbNi_{0.8}Au_{0.2}In$ at 1.65 K (see Fig. 3a) show additional peaks of magnetic origin. Numerical analysis of the intensities of the magnetic peaks indicates that the Tb moments, equal to 8.0(1) μ_B, lie in the $a-b$ plane and form a noncollinear structure in which the Tb moments form the angles of 180°, 60° and −60° with the a-axis for Tb_1 (x, 0, 1/2), Tb_2 ($\bar{x}$, $\bar{x}$, 1/2) and Tb_3 (0, x, 1/2), respectively. For this magnetic structure model R_{magn} = 8.6% was obtained. Similar structure is observed at 25 K with the magnetic moments equal to 6.4(2) μ_B (R_{magn} = 9.6%). The type of the magnetic ordering does not change with increasing temperature.

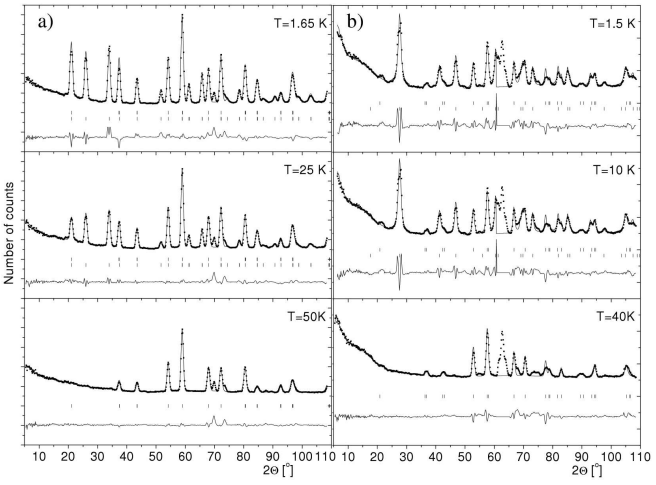

Fig. 3. Neutron diffraction patterns of $TbNi_{1-x}Au_xIn$ for (a) $x = 0.2$ at 1.65, 25 and 50 K and (b) $x = 0.8$ at 1.5, 10 and 40 K. The squares represent experimental points, the solid lines are the calculated profiles for the crystal and magnetic structure models (as described in the text) and the difference between the observed and the calculated intensity (the bottom of each diagram). The vertical bars indicate the positions of the Bragg peaks of nuclear and magnetic origin, respectively. The peak at $2\theta = 63°$ originates from the cryostat.

The neutron diffraction patterns of $TbNi_{0.6}Au_{0.4}In$ at 1.65 and 25 K are similar to the one of $TbNi_{0.8}Au_{0.2}In$. In both temperatures similar magnetic structure with the Tb moments equal to 6.8(4) μ_B and 4.5(2) μ_B, respectively, is present.

For $x = 0.8$ a different distribution of magnetic reflections in the neutron diffraction patterns collected at 1.5 and 10 K is observed (Fig. 3b). These peaks are indexed by the propagation vector $\boldsymbol{k}$ = (0, 0, 1/2), similarly to TbAuIn [5]. Analysis of the intensities of the magnetic peaks confirms that: the Tb magnetic moments equal to 6.5(3) μ_B at both temperatures lie in the $a-b$ plane and form a noncollinear structure of the 120°-type which does not change up to T_N equal to 36 K.

In all the investigated compounds no magnetic moment was detected on the Ni atoms.

4. Discussion and summary

The X-ray and neutron diffraction data presented in the work indicate that the $TbNi_{1-x}Au_xIn$ solid solutions with the hexagonal ZrNiAl-type structure exist in the whole Au content range. With increasing x the values of the a and c lattice parameters and unit cell volume V increase. The dependence of the a/c ratio versus the dilution parameter x is anomalous showing the maximum for $x = 0.4$.

Magnetic data show that with increasing x the value of the critical temperature of the magnetic order decreases. Analysis of the magnetic and crystal structure parameters indicates that for $x > 0.4$ the Néel temperature dependence on the lattice parameter a and unit cell volume V is not as strong as for lower Au content but almost linear in the mentioned regions (Fig. 4).

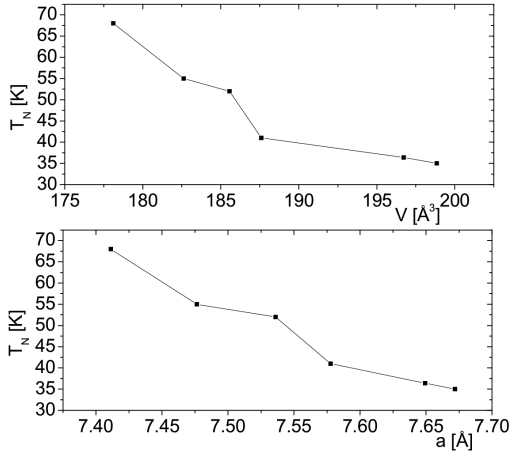

Fig. 4. Dependence of the Néel temperature versus the lattice parameter a and the unit cell volume V.

Neutron diffraction data indicate a change in the magnetic structure. In the compounds with the high Ni concentration magnetic ordering is described by the propagation vector $\boldsymbol{k}$ = (1/2, 0, 1/2) similarly as observed in TbNiIn while in those with the small Ni concentration magnetic order is described by the propagation vector $\boldsymbol{k}$ = (0, 0, 1/2) similarly to the structure of TbAuIn. In all the compounds the Tb magnetic moments form locally noncollinear magnetic structure of the 120°-type. The values of the magnetic moments are smaller than the free Tb^{3+} ion value (9.0 μ_B, see Table). This suggests strong influence of the crystal electric field on the magnetic order.

The difference in magnetic and neutron diffraction data concerning the additional phase transitions for the

compounds with $x \leq 0.4$ results from different sensitivity of the two experimental methods. The results are similar to those reported for TbRhSn [8] and TbPdAl [9]. In these compounds the anomaly at T_t equal to 11 K [8] and 100 K [9] is of the first order and is associated with the small jump in the volume (see Fig. 8 in Ref. [8]).

The observed magnetic order could be described in the $X-Y$ model introducing two exchange integrals J_1 and J_2 between nearest and next nearest spins [10] and an additional exchange integral $J_c < 0$ which describes the antiferromagnetic interactions along the c-axis.

Acknowledgments

This work was partially supported by the Ministry of Science and Higher Education in Poland under the grant No. 1 P03B 111 29. This research project was partially supported by the European Commission under the 6th framework. Programme through the Key Action Strengthening the European Research Area, Research Infrastructures. Contract No. RII3-CT-2003-505925 (NMI3). Kind hospitality and financial support extended for two of us (S.B. and B.P.) by the Berlin Neutron Scattering Centre, Hahn–Meitner Institute is gratefully acknowledged.

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