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Magnetic Properties of $Tb_{1-x}Y_x$ NiIn System

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Polycrystalline samples of $Tb_{1-x}Y_x$NiIn ($x = 0.2$ and 0.4) were prepared and studied by powder X-ray diffraction and AC and DC magnetic measurements. The compounds crystallize in the hexagonal ZrNiAl-type structure. At low temperatures the ferromagnetic properties are detected. With increasing Y content a change in the magnetic properties, decrease of the Curie temperatures and magnetic moment in the ordered state are observed.

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1. Introduction

TbNiIn and YNiIn are isostructural compounds which crystallize in the hexagonal crystal structure of the ZrNiAl-type (space group $P\bar{6}2m$) [1, 2]. In this structure R (Tb or Y) atoms are stacked in the R–In layers separated by the nonmagnetic Ni–In layers. The R atoms occupy the 3(g) positions: x, 0, 1/2; 0, x, 1/2; $\bar{x}, \bar{x}$, 1/2 and form a triangular structure which is a deformed kagomé lattice: equilateral triangles of three R atoms are joined on each apex to build up deformed hexagons. 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. The noncollinear structure is stable up to the Curie temperature equal to 70 K [3, 4]. Neutron diffraction data have not detected magnetic moment on the Ni atom.

This work reports the results of X-ray powder diffraction and magnetic measurements of the $Tb_{1-x}Y_x$NiIn solid solutions for $x = 0.2$ and 0.4. The work has in view to determine the parameters of the crystal structure and magnetic properties as a function of the Y content x.

2. Experimental

The polycrystalline samples of $Tb_{1-x}Y_x$NiIn for x equal to 0.2 and 0.4 were prepared by arc melting of high-purity elements (Tb and Y: 3N, Ni: 4N and In: 5N) in a titanium gettered argon atmosphere. Afterwards the samples were annealed in evacuated vycor capsules at 600 °C for 1 week.

X-ray powder diffraction patterns were recorded at room temperature (Cu K_α; Philips X'Pert PRO diffractometer). The data were analyzed with the Rietveld structure refinement program Fullprof [5].

Dc and ac magnetic measurements were carried out using a commercial MPMS SQUID magnetometer in the temperature range 2–300 K in the magnetic field up to 50 kOe. Then, a zero-field-cooling (ZFC) and field-cooling (FC) measurement at low field (≈ 10 Oe) was made to get precise information about temperature of possible magnetic transitions. Both ZFC and FC curves were measured during heating.

3. Results

The X-ray diffraction analysis confirms that the obtained samples with $x = 0.2$ and 0.4 are single phase, with the ZrNiAl-type structure (space group $P\bar{6}2m$). The values of a and c lattice parameters do not change with increasing Y content x which is in good relation with the atomic radius of Tb (1.78 Å) and Y (1.80 Å).

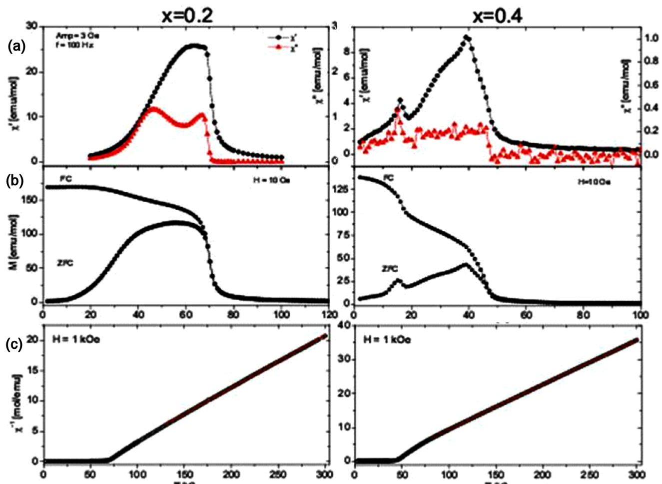

Fig. 1. Temperature dependence of the magnetic susceptibility of $Tb_{1-x}Y_x$NiIn: (a) ac χ' and χ'', (b) ZFC and FC dc magnetization, (c) reciprocal magnetic susceptibility.

Figure 1 presents the results of the magnetic measurements for $x = 0.2$ and 0.4. For $Tb_{0.8}Y_{0.2}$NiIn temperature dependence of the χ' component of ac magnetic susceptibility shows a broad maximum near 65 K and then decreases rapidly at 71 K. The χ'' component has

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two maxima, at 46.5 K and 66.5 K, and decreases to zero at 71 K. Dc magnetization reveals the large difference between ZFC and FC curves. ZFC dependence is similar to that observed for $\chi'(T)$ whereas the FC curve indicates the additional phase transition near 40 K.

For $Tb_{0.6}Y_{0.4}NiIn$ the temperature dependence of χ' indicates two maxima at 16 and 39 K. The $\chi''(T)$ has the maximum at 16 K and decreases to zero at 48 K. The temperature dependence of the ZFC magnetization at $H = 10$ Oe is similar as that of χ' whereas FC indicates the change at 16 K.

Magnetization curves for both samples measured at 2 K in the field up to $H = 50$ kOe (see Fig. 2) indicate the hysteresis loops with the coercive fields equal to 1 kOe for $x = 0.2$ and 3.3 kOe for $x = 0.4$. Magnetization is not saturated and in the magnetic field $H = 50$ kOe magnetic moment equals 8.0 μ_B for $x = 0.2$ and 4.55 μ_B for $x = 0.4$.

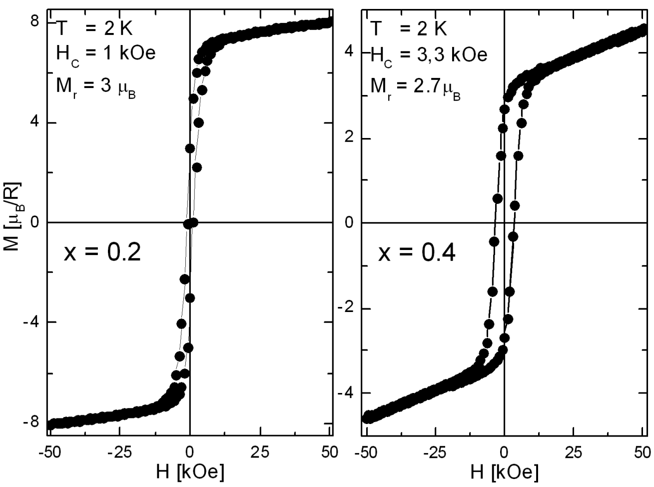

Fig. 2. A hysteresis loop for $Tb_{1-x}Y_xNiIn$ for $x = 0.2$ and 0.4 measurement at $T = 2$ K and magnetic field up to 50 kOe.

TABLE

Magnetic data of $Tb_{1-x}Y_xNiIn$ for $x = 0$, 0.2, and 0.4.

x	T_C [K]	T_t [K]	θ_p [K]	$\mu_{eff}(\mu_B)$ /Tb atom	T_{max} [K]		Ref.
0	70	32	55	9.5	50	28	[3]
0.2	71	40	58	10.3	67	46.5	this work
0.4	48	16	30	10.0	39	16	this work

Above 100 K the reciprocal magnetic susceptibility obeys the Curie–Weiss law with the positive values of the paramagnetic Curie temperature θ_p (see Table) and the effective magnetic moment close to the free Tb^{3+} ion value (9.72 μ_B).

4. Discussion and summary

The X-ray diffraction data indicate that in the $Tb_{1-x}Y_xNiIn$ system the compounds with the hexagonal ZrNiAl-type structure exist for $x = 0.2$ and 0.4.

With increasing x the number of nonmagnetic Y atoms decreases the number of paramagnetic Tb atoms with localized magnetic moments, therefore the doped diamagnetic Y atoms, which probably are statically distributed in the Tb sublattices, should influence magnetic interactions.

Magnetic data indicate that at low temperatures the magnetic properties are similar to those observed in TbNiIn. The two magnetic phase transitions at T_C and T_t are observed. The values of T_C and T_t decrease with increasing x (Table). The magnetization curves at 2 K have ferromagnetic-like character with the magnetic moment smaller than free Tb^{3+} ion value (9 μ_B).

The interatomic Tb–Tb distances in plane and between the planes in these compounds are large and because of that magnetic properties of these compounds can be described in the Ruderman–Kittel–Kasuya–Yosida (RKKY) [6] or Campbell [7] model. The first model suggests the long range magnetic interaction via conduction electrons in which the exchange integral has the oscillatory character, whereas the second one suggests the indirect interaction of $4f$ levels across the $5d$ bands of rare earth atoms and $3d$ bands of Ni atoms.

Observed dependence of the T_C and T_t, first constant (or larger) for $x = 0.2$ and next smaller ($x = 0.4$) suggests that the interaction is most probably of the RKKY type.

The large difference between the ZFC and FC curves are related to domain effects in powder samples with strong magnetocrystalline anisotropy. The similar behavior was observed in RNiIn [3] and RPdIn (R = Tb, Dy) compounds with complex magnetic structures and with domain-wall pinning effects [8].

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