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Electrical Resistivity and Electronic Structure of the $Tb_xGd_{1-x}Ni_3$ System

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In the paper the electric properties and electronic structure of the intermetallic $Tb_xGd_{1-x}Ni_3$ compounds are presented. The partial replacement of Gd by Tb atoms causes the decrease of the Curie temperature (T_C) and the increase of the residual resistivity. According to the Matthiessen rule the scattering mechanisms in $\rho(T)$ have been analyzed. Moreover, the reduced form of the electrical resistivity $\rho_Z(T-T_0)$ indicates a deviation from the linearity for $x > 0.2$. This kind of behaviour can be attributed to density of d states near by the Fermi level (E_F) which are dominated by Ni $3d$ states. The valence band spectra as well as the core level lines have been analyzed as the influence of Tb/Gd substitution on the electronic structure.

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1. Introduction

The RT_3 compounds where R is rare earth and T is a transition metal show interesting properties due to the combination of $3d$ itinerant and $4f$ localized magnetism [1–17]. One of the first studied group is RNi_3. Obviously, its properties depends on the type of R atoms. In the $GdNi_3$ compound the Curie temperature $T_C = 115$ K and the saturation magnetic moment equals 6.55 μ_B/f.u. [1, 4, 12, 13]. The $TbNi_3$ compound exhibits non-collinear magnetic structure where the terbium atoms in the same crystallographic position are arranged ferromagnetically but they are turned between two different positions [16]. Thus, the magnetic susceptibility exhibits a large magnetic anisotropy with two different paramagnetic Curie temperatures $\theta_{\parallel} = 9$ K and $\theta_{\perp} = 63$ K. The saturation magnetic moment equals 7.52 μ_B/f.u. The electrical resistivity exhibits two magnetic transitions, one at about 55 K (helimagnetic structure of terbium moments along c axis) and the second at about 80 K (transition from ferromagnetic to paramagnetic state) [16].

In this paper we are focused on the influence of terbium substitution on the electric properties and the electronic structure in the $Tb_xGd_{1-x}Ni_3$ series.

2. Experimental details

The polycrystalline samples $Tb_xGd_{1-x}Ni_3$ ($x = 0.0$, 0.2, 0.5, 0.8, 1.0) were prepared by arc-melting from high purity elements under argon atmosphere. All samples were wrapped in tantalum foil, placed in quartz tubes and annealed at 900°C for one week. The crystal structure was checked by means of X-ray diffraction (XRD) using Siemens D5000 diffractometer. The electrical resistivity $\rho(T)$ was measured quasi-continuously at a slowly changing temperature (4.2–300 K) by a standard four-probe technique. The X-ray photoelectron spectroscopy (XPS) measurements were performed with the use of PHI 5700/660 Physical Electronics spectrometer. The spectra were analyzed at room temperature using monochromatized Al K_α radiation (1486.6 eV). The samples were fractured and measured in vacuum of 10^{-10} Torr.

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3. Results and discussion

3.1. Electrical resistivity

The $\rho(T)$ dependence for the $Gd_{1-x}Tb_xNi_3$ systems is presented in Fig. 1a. The Tb/Gd substitution causes the decrease of the temperature of the magnetic phase transition (T_C). Similar behaviour has been observed from magnetic measurements [18]. According to the Matthiessen rule the electrical resistivity can be described as

$$\rho(T) = \rho_0 + \rho_{ph}(T) + \rho_{mag}(T), \quad (1)$$

where ρ_0 — residual resistivity, ρ_{ph} — phonon contribution, ρ_{mag} — magnetic part. The ρ_{ph} for pure metals in the high temperature range follows the Bloch–Grüneisen dependence and changes linearly with T. The ρ_{mag} represents a spin-disorder contribution which is caused by the scattering of the conduction electrons on $4f$ moments and contribution correlated with spin fluctuations. In our case the value of ρ_0 increases with the terbium substitution (see Table). It may be a result of crystal disorder which appears in the crystal lattice during substitution.

Analyzing the $\rho(T)$ curve below T_C we have to take into account the phonon contribution ($\rho_{ph} \sim T^5$) and the scattering of s electrons to free d states ($\rho_{sd} \sim T^3$). Therefore, the following expression has been used:

$$y = ax^5 - bx^3 + cx^n. \quad (2)$$

For the compounds with $x \leq 0.2$ the n is close to 2 which indicates the scattering on the spin waves (magnons). In the case of compounds with $x > 0.2$ the resistivity below

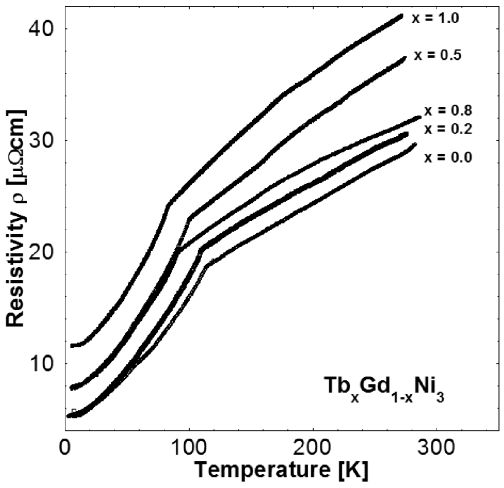

Fig. 1. Temperature dependence of the electrical resistivity for the $Tb_xGd_{1-x}Ni_3$ system.

T_C can be rather fitted by using the equation

$$y = ax^5 - bx^3 + cx^2 \exp(-d/x), \quad (3)$$

where a, b, c are constants and parameter d is proportional to the energy gap ($d = \Delta/2k_B$). The exponential contribution may be the result of a magnetic anisotropy which was already observed by Hashimoto *et al.* in the $TbNi_3$ compound [16].

The analysis of parameters obtained from the fitting of $\rho(T)$ dependence indicates that all of them increase with Gd/Tb substitution. At the same time the value of energy gap Δ in the spin waves spectra also increases (see Table). This change is caused by using additional energy which is needed to reverse the localized spins opposite to the anisotropy field.

The reduced form of the electrical resistivity can be represented as

$$\rho_Z = \rho(T) - \rho(T_0)/\rho(T_1) - \rho(T_0), \quad (4)$$

in the assumption where $T_0 = 140$ K and $T_1 = 260$ K.

TABLE

The transport properties of the $Tb_xGd_{1-x}Ni_3$ compounds.

x	T_C [K]	ρ_0 [$\mu\Omega$ cm]	$a \times 10^{-10}$	$b \times 10^{-5}$	$c \times 10^{-3}$	n	
			[$\mu\Omega$ cm K^{-5}]				
0.0	115	5.17	55–113 K				
			3.68	1.33	1.45	2.08	
			4.2–50 K				
			4.19	1.97	3	1.90	
			$T < T_C$				
0.2	110	5.29	3.69	1.51	2.73	1.97	
						d [K]	Δ [meV]
0.5	101	7.75	6.02	2.04	3.02	2.89	0.25
0.8	90	7.79	8.50	2.64	3.50	8.22	0.71
1.0	83	11.45	16.2	3.63	4.31	8.44	0.73

The $\rho_Z(T - T_0)$ concerns paramagnetic range and represents the phonon contribution and the s–d scattering on the non-ordered spins. Thus, ρ_Z exhibits linear behaviour above T_C only for the samples with $x \leq 0.2$. In the case of higher Tb content the $\rho_Z(T - T_0)$ resistivity deviates from the linearity and shows a positive curvature. This kind of behaviour can be explained as the strong dependence on density of d states nearby the Fermi level.

3.2. Electronic structure

For all studied samples valence bands (VB) spectra nearby the Fermi level (E_F) are dominated by the Ni $3d$ states (Fig. 2a). The terbium substitution causes the decrease of the intensity of Gd $4f$ line which is located at 8 eV. Simultaneously, the multiplet structure of Tb appears. This structure consists of several lines located at 2 eV, 7.4 eV, 9.1 eV and 10.2 eV. The broadening of states nearby to E_F is particularly visible for $x = 0.8$ and 1.0 terbium concentration. It is the result of Tb substitution and the increase of the intensity of Tb $4f$ states (Fig. 2a and b). For the compounds with $x \geq 0.5$ the slight increase of intensity of states on the E_F is visible. This behaviour could be the result of f–d interaction. Simultaneously in the resistivity measurements it has been shown that for these compounds the resistivity increases with the temperature faster than for other compounds ($x \leq 0.2$). On the one hand, it may be the effect of decrease of $5d$ electrons number (terbium has two $4f$ electron more but one none $5d$ electron than gadolinium). On the other hand, it may be the result of the decrease of relaxation time between collisions as a consequence of higher probability of electrons scattering into free d states.

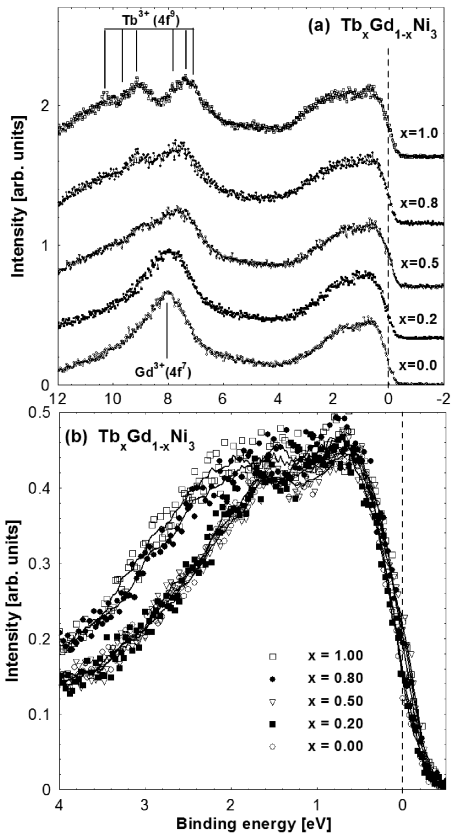

Fig. 2. (a) Valence band spectra in the broad binding energy range and (b) near by the Fermi level for the $Tb_xGd_{1-x}Ni_3$ series where $x = 0.0$, 0.2, 0.5, 0.8, and 1.0.

4. Summary

The aim of presented work was to study the electric properties as well as the electronic structure of the $Tb_xGd_{1-x}Ni_3$ series. All results can be concluded as follows:

- The value of the temperature of phase transition T_C obtained from $\rho(T)$ decreases with Gd/Tb substitution. The fitting of $\rho(T)$ dependence for $x \leq 0.2$ below T_C indicates the scattering on the spin waves whereas for $x > 0.2$ is rather connected with a magnetic anisotropy. Additionally, the reduced resistivity $\rho_Z(T - T_0)$ exhibits linear dependence for $x \leq 0.2$ samples and for higher Tb content deviates from the linearity. This deviation can be connected with the change of the electronic structure.
- The VB spectra nearby E_F are dominated by the Ni $3d$ states. For the compounds with $x \geq 0.5$ the slight increase of intensity of states on the E_F has been observed. Simultaneously, the $\rho_Z(T - T_0)$ indicates the deviation from the linearity. It may be the effect of decrease of $5d$ electron number but also the result of the decrease of relaxation time between collisions as a consequence of higher probability of electrons scattering into free d states.

Acknowledgments

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