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MAGNETIC PHASE TRANSITIONS IN INTERMETALLIC $GdNi_{5-x}Sn_{1+x}$ AND $TbNi_{5-x}Sn_{1+x}$ COMPOUNDS

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Magnetization measurements were performed for intermetallic compounds $GdNi_{5-x}Sn_{1+x}$ and $TbNi_{5-x}Sn_{1+x}$ in the temperature range from 4.5 K to 280 K and in magnetic field up to 50 kOe. Complex magnetic phase diagram was revealed for both compounds. The results are discussed assuming three-sublattice magnetic structure and partial disordering of the structure.

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1. Introduction

Recently a series of new compounds $RNi_{5-x}Sn_{1+x}$, where R = Y, Sm, Gd to Yb, has been synthesized [1]. It was found that the crystal structure of these compounds is described by the space group *Pnnm*. From the detailed X-ray studies of $HoNi_5Sn$ it was stated that a part of Ni atoms in sites 4(g) and 4(g') are substituted by extra Sn atoms distributed statistically in these positions. Therefore the formula of this compound should be given in the form $HoNi_{4.9}Sn_{1.1}$. It is assumed that the similar stoichiometry is common for other members of presented series, therefore the formula $RNi_{5-x}Sn_{1+x}$ is taken for them (with x about 0.1). A majority of mentioned compounds (beside those with R = Y, Er, Tm) undergo a magnetic ordering at temperatures below 40 K [1]. In the present paper the results of more detailed studies of magnetic properties of $GdNi_{5-x}Sn_{1+x}$ and $TbNi_{5-x}Sn_{1+x}$ are given. These results are also completed with transport measurements of $GdNi_{5-x}Sn_{1+x}$.

2. Experiment and results

The method of samples preparation was described in [1]. The magnetization measurements were carried out, using the SQUID magnetometer, in the temperature range from 4.5 K to 280 K in magnetic field up to 50 kOe. The electrical resistivity as a function of temperature was measured by the four probe method using the a.c. bridge.

2.1. $GdNi_{5-x}Sn_{1+x}$

In higher temperature range the magnetic susceptibility, χ, measured both in magnetic field 50 Oe and 3000 Oe follows the modified Curie–Weiss law, $\chi(T) = \chi_0 + c_g/(T - \Theta_p)$, where $\chi_0 = 7 \times 10^{-6}$ emu/(g · Oe), $c_g = 0.0155$ (emu · K)/(g · Oe) and $\Theta_p = 7.5$ K. The effective moment for a formula unit $\mu_{eff} = 8.4\mu_B$ (calculated from c_g) is close to the theoretical value of magnetic moment on Gd^{3+} ion. It agrees well with the fact that Ni atoms in such compounds practically do not contribute to the magnetic moment due to hybridization of the $3d$ band of Ni and $6s$–$5d$ band of Gd. The temperature dependence of magnetization below 40 K, when measured in low fields, suggests the occurrence of several magnetic phase transitions (see Fig. 1). Distinct kinks between 10 and 35 K on the $M(T)$ curve, when measured in 5 Oe, become practically invisible in 3000 Oe. In the same temperature range a distinct difference occurs between low field magnetization of zero field cooled (ZFC) sample and of field cooled (FC) sample (Fig. 1). This behavior suggests an existence of a spin-glass state. The magnetization curves as a function

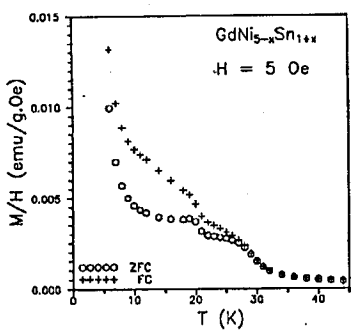

Fig. 1. Temperature dependence of dc magnetic susceptibility of ZFC and FC sample of $GdNi_{5-x}Sn_{1+x}$ measured in magnetic field of 5 Oe.

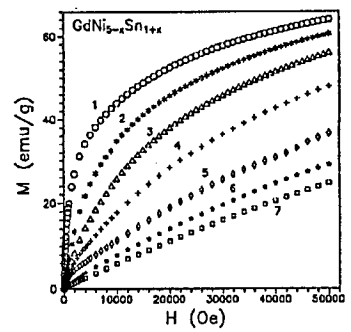

Fig. 2. Magnetization vs. magnetic field for $GdNi_{5-x}Sn_{1+x}$ at various temperatures (1 — 4.5 K; 2 — 7 K; 3 — 10 K; 4 — 15 K; 5 — 23 K; 6 — 30 K; 7 — 36 K).

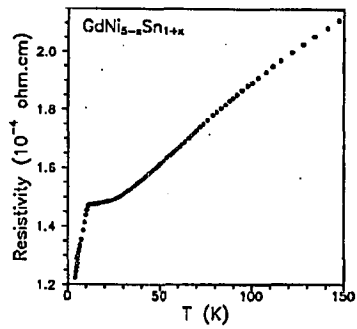

Fig. 3. Resistivity versus temperature for $GdNi_{5-x}Sn_{1+x}$.

of the applied field are presented in Fig. 2. A spontaneous magnetization (M_0), which can be determined from the Arrott plot, takes place only below 10 K with magnetic moments μ_0 equal to $4.01\mu_B$ and $2.30\mu_B$ at 4.5 K and 7 K, respectively. The temperature dependence of resistivity has a shape which is characteristic of rare-earth compounds with a transition to the magnetic order (Fig. 3). Because the electric conductivity of this compound has the metallic character, one can expect that the main contribution to χ_0 has a Pauli paramagnetic origin.

2.2. $TbNi_{5-x}Sn_{1+x}$

The temperature dependence of susceptibility of $TbNi_{5-x}Sn_{1+x}$ is similar to that of $GdNi_{5-x}Sn_{1+x}$, but not so complex because there are only two transitions. The temperature dependence of magnetization, measured in magnetic field of 5 Oe for ZFC and FC sample, is shown in Fig. 4. As one can see a spin-glass-like behavior is also observed in this case. In the low temperature region the susceptibility depends also on a magnetic field intensity. The $M(H)$ dependence is not linear at 6 K, however, a spontaneous magnetization ($2.98\mu_B$) determined from Arrott plot was found only for 4.5 K.

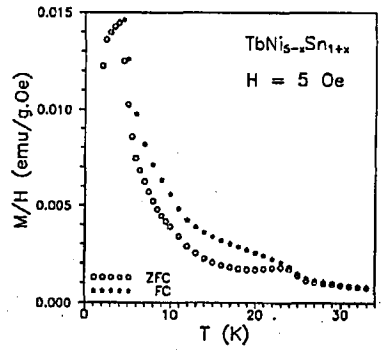

Fig. 4. Temperature dependence of dc magnetic susceptibility of ZFC and FC sample of $TbNi_{5-x}Sn_{1+x}$ measured in magnetic field of 5 Oe.

3. Discussion

The complicated magnetic behavior of $GdNi_{5-x}Sn_{1+x}$ and $TbNi_{5-x}Sn_{1+x}$ can be related to their complex crystal structure, and especially to the presence of three nonequivalent positions of the rare-earth atoms and to the partial disordering of the structure. The kinks on $M(T)$ curves and the ZFC/FC hysteresis of the magnetization suggest an existence of magnetic phase transitions and a spin-glass formation. One can assume that, in the case of $GdNi_{5-x}Sn_{1+x}$, at about 33 K one of three Gd sublattices begins to order ferro- or ferrimagnetically. Simultaneously the rest of Gd ions undergoes to spin-glass-like state (with freezing temperature $\approx$ 27 K). Then, between 22–19 K, the next Gd sublattice orders, whereas a third Gd sublattice is still in the spin-glass state. The magnetization of the ordered Gd sublattices is masked by spin glass, therefore the spontaneous magnetization is not observed. Finally, at about 10 K the last transition begins and an ordering of all Gd atoms occurs. This last magnetic transition is very well seen on the $R(T)$ dependence (Fig. 3).

An intermediate magnetic transition is not observed for $TbNi_{5-x}Sn_{1+x}$. The first transition (of one or two Tb sublattices) takes place between 26–23 K, being associated also with the spin-glass formation. The next (probably ferrimagnetic) transition takes place at about 4.6 K. The magnetization curves of $TbNi_{5-x}Sn_{1+x}$ in weak applied fields are similar to those observed in the TbCoSn compounds [2, 3]. This last compound crystallizes also in orthorhombic structure of the TiNiSi type, but Tb atoms occupy only one position. The magnetic and neutronographic measurements of TbCoSn indicate the formation of a modulated non-collinear magnetic structure at T_n = 20.5 K. Next below T_n two additional phase transitions at 11.6 K and 5.4 K are observed. It is remarkable that in TbCoSn neutronographic measurements indicate that a part of the Tb moments remains in a disorder state. We think that these moments can be in a spin-glass state as well. The complex magnetic behavior of TbCoSn is explained by subtle balance between the long-range RKKY coupling and the crystal field effects [2]. However, this last effect which could be taken into consideration for $TbNi_{5-x}Sn_{1+x}$, should be excluded in the case of Gd compounds. Apparently in $GdNi_{5-x}Sn_{1+x}$ (as well in $TbNi_{5-x}Sn_{1+x}$) the complex magnetic behavior could be due to a competition between ferro- and antiferromagnetic exchange interactions. In the $RNi_{5-x}Sn_{1+x}$ structure the R atoms form planes which are perpendicular to x-axis. Ferromagnetic interactions could be dominating in the planes, whereas antiferromagnetic interactions could be realized between the planes.

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