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Electrical Transport Properties of $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho) Compounds

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Electrical transport properties of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho) intermetallics crystallized in the orthorhombic $Gd_3Cu_4Ge_4$ structure type (space group *Immm*) were studied in the temperature interval 11 – 300 K. Measurements of the temperature dependencies of electrical resistivity ($\rho(T)$) showed that all the studied compounds are characterized by metallic type of conductivity. The slope change of the resistivity at low temperature part of $\rho(T)$ dependencies for $Gd_3Ag_4Sn_4$, $Tb_3Ag_4Sn_4$ and $Dy_3Ag_4Sn_4$ compounds is connected with their magnetic ordering. Change of the resistivity caused by magnetic ordering was not observed for the $Ho_3Ag_4Sn_4$ compound in the studied temperature interval. Relation between magnetic and electric properties of the investigated $R_3Ag_4Sn_4$ compounds was analyzed.

Keywords: intermetallics, electrical resistivity, X-ray phase analysis.

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Introduction

One of the several well-known series of the compounds formed in the R–M–Sn (R – rare earth metal, M – *d*-element) ternary systems, is the series of the $R_3M_4Sn_4$ intermetallics which crystallize in the $Gd_3Cu_4Ge_4$ structure type (space group *Immm*) [1-5]. The crystal structure of the $Gd_3Cu_4Ge_4$-type stannides is a combination of the structural fragments of the AlB_2-type and $MgCuAl_2$-type. Magnetic property studies of the $R_3M_4Sn_4$ compounds showed that intermetallics with magnetic rare earth elements are characterized by antiferromagnetic ordering at low temperatures, and the magnetic moments are localized on the atoms of the rare earths [5-10]. Magnetic structure investigation for some $R_3M_4Sn_4$ compounds using the method of neutron diffraction showed the different magnetic ordering of the two sublattice of the rare earth atoms which occupy in the structure two symmetry-inequivalent sites 4*e* and 2*d* [9, 10]. In the $R_3Mn_4Sn_4$ (R = Ce, Pr, Nd, Sm) stannides the magnetic moments of the rare earth elements and manganese atoms order at different temperatures [5]. Magnetic properties of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy) compounds were studied by neutron diffraction and Mossbauer spectroscopy [11-13]. Performed investigations showed that these compounds are characterized by antiferromagnetic ordering at low temperatures.

It is well known that the peculiarity of the magnetic state of the intermetallic compounds with rare earths of Yttrium group is displayed on their electrical property behavior. It is caused by the change of the scattering mechanisms depending on the magnetic component in the compound, nature and orientation of the magnetic moments, etc. Thus, the study of electrical properties of the intermetallics gives an additional confirmation of the magnetic ordering or its absence in intermetallic compounds.

In this work we present the results of the electrical property investigation of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho) stannides in the temperature interval 11 – 300 K and an analysis of the relation between their magnetic and electric properties.

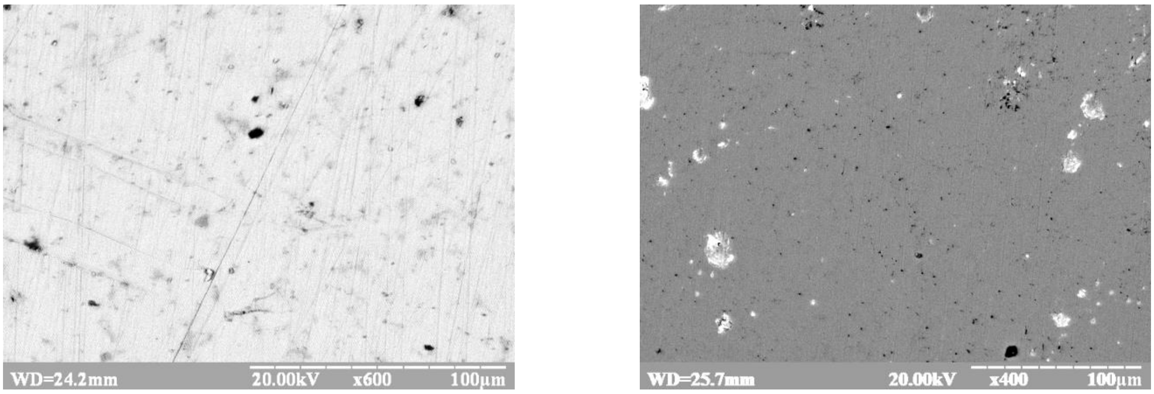
I. Experimental

The samples of the nominal composition $R_{28}Ag_{36}Sn_{36}$, where R = Gd, Tb, Dy, Ho, were synthesized by direct arc-melting of the constituent elements (purity of rare earths 99.9 wt. %, Ag purity 99.99 wt. %, and Sn purity 99.999 wt. %) under Ti-gettered, argon atmosphere on a water-cooled copper hearth. The pieces of the alloys were sealed in evacuated silica tubes, homogenized at 770 K for 720 h and subsequently quenched in ice water. Phase analysis was performed using X-ray powder diffraction of the synthesized samples (diffractometer DRON-2.0M with

Table 1

Lattice parameters and EPMA data of the $R_3Ag_4Sn_4$ compounds

Compound	EPMA data, at. %	Lattice parameters, nm		
		a	*b*	*c*
$Gd_3Ag_4Sn_4$	$Gd_{28.93}Ag_{35.46}Sn_{36.61}$	0.4566(3)	0.7306(3)	1.5222(4)
$Tb_3Ag_4Sn_4$	$Tb_{28.09}Ag_{35.56}Sn_{36.35}$	0.4558(2)	0.7282(4)	1.5182(7)
$Dy_3Ag_4Sn_4$	$Dy_{27.99}Ag_{36.30}Sn_{35.71}$	0.4532(1)	0.7288(2)	1.5123(4)
$Ho_3Ag_4Sn_4$	$Ho_{27.98}Ag_{36.28}Sn_{35.74}$	0.4520(2)	0.7281(3)	1.5092(3)



a *b*

Fig. 1. Electron microphotographs of the $Gd_3Ag_4Sn_4$ (*a*) and $Tb_3Ag_4Sn_4$ (*b*) compounds.

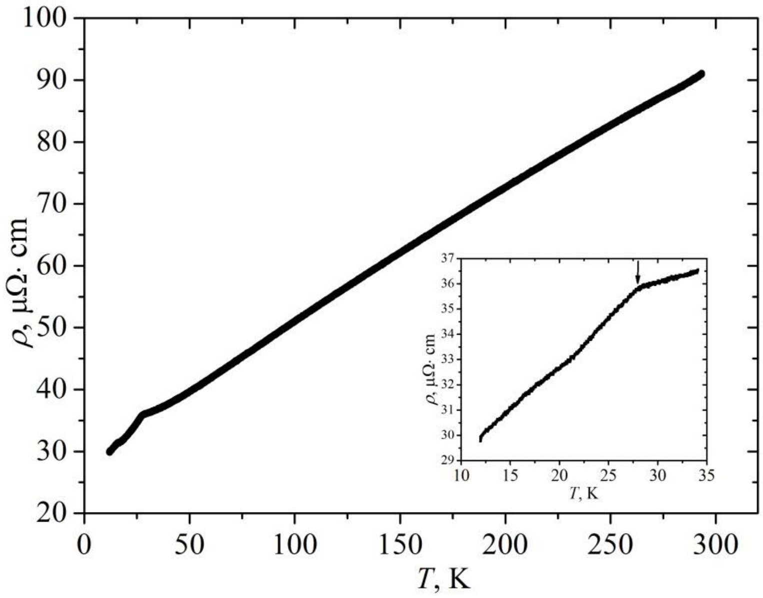


Fig. 2. Temperature dependence of the electrical resistivity *ρ(T)* of the $Gd_3Ag_4Sn_4$ compound. The insert presents the low-temperature dependence of the electrical resistivity.

FeK_α radiation). Chemical and phase compositions of the samples were tested by electron microprobe analysis (EPMA) (scanning electron microscope REMMA 102-02). The calculation of the lattice parameters was performed using the WinCSD program package [14].

The temperature dependencies of electrical resistivity were measured in the temperature range 11 - 300 K employing two-probe method on millimeter-scale, well-shaped pieces cut by spark erosion from the polycrystalline samples using the helium cooler with closed cycle (Advanced Research systems, USA).

II. Results and discussion

According to performed X-ray phase analysis the prepared $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho) compounds crystallize in the $Gd_3Cu_4Ge_4$ structure type (space group *Immm*). Calculated lattice parameters and EPMA data of

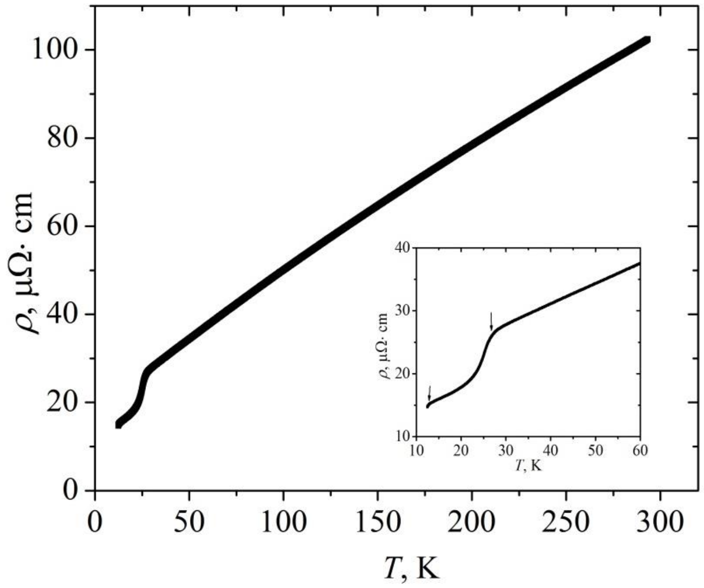


Fig. 3. Temperature dependence of the electrical resistivity *ρ(T)* of the $Tb_3Ag_4Sn_4$ compound. The insert presents the low-temperature dependence of the electrical resistivity.

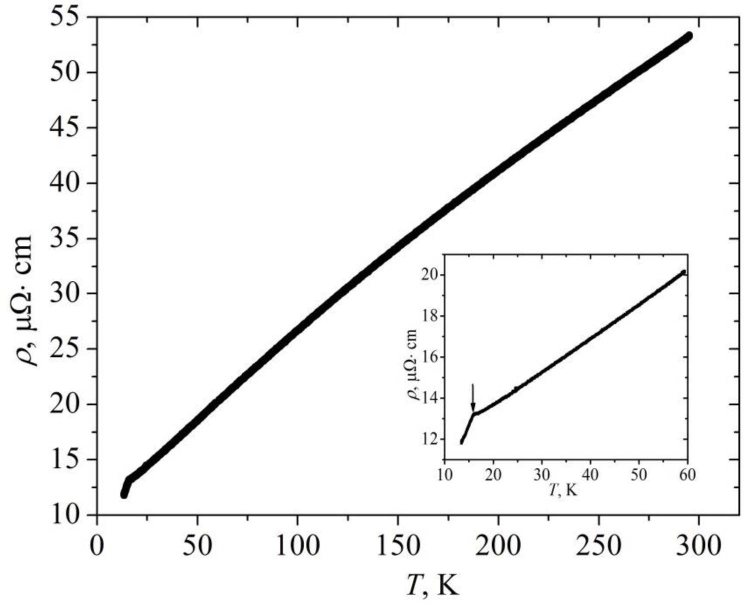


Fig. 4. Temperature dependence of the electrical resistivity *ρ(T)* of the $Dy_3Ag_4Sn_4$ compound. The insert presents the low-temperature dependence of the electrical resistivity.

the $R_3Ag_4Sn_4$ compounds are gathered in Table 1. Electron microphotographs of the $Gd_3Ag_4Sn_4$ and $Tb_3Ag_4Sn_4$ compounds are shown in Fig. 1.

The temperature dependencies of the electrical resistivity *ρ(T)* of the $R_3Ag_4Sn_4$ compounds measured in the temperature range 11 - 300 K are shown in Figs. 2-5. As evidently from Figs. 2-5, the small resistivity values (6.7 - 101.5 μΩ·cm) of the all studied compounds and increasing of their resistivity with temperature indicated metallic type of conductivity.

The slope change of the resistivity at low temperature part of the *ρ*(*T*) dependencies (Figs. 2-4) observed for the stannides with magnetic rare earths R = Gd, Tb, Dy, is connected with their magnetic ordering, previously reported in Refs. [11-13]. Analysis of the temperature dependence of electrical resistivity of the $Gd_3Ag_4Sn_4$ compound (Fig. 2, insert) indicated the presence of the transition at low temperature which confirmed the magnetic ordering of this compound at 28 K [11]. Unfortunately, the used in our work temperature interval of measurements is not sufficient to confirm the second transition temperature at 8 K connected with spin-reorientation transition.

Two transitions, at 27.2 K and 13.1 K, were observed on the temperature dependence of the electrical resistivity for the $Tb_3Ag_4Sn_4$ compound (Fig. 3, insert). Obtained result is in a good agreement with behavior of the magnetic properties, where the temperature 27.2 K corresponds to the antiferromagnetic ordering (T_N = 28 K), and at 13.1 K spin-reorientation transition

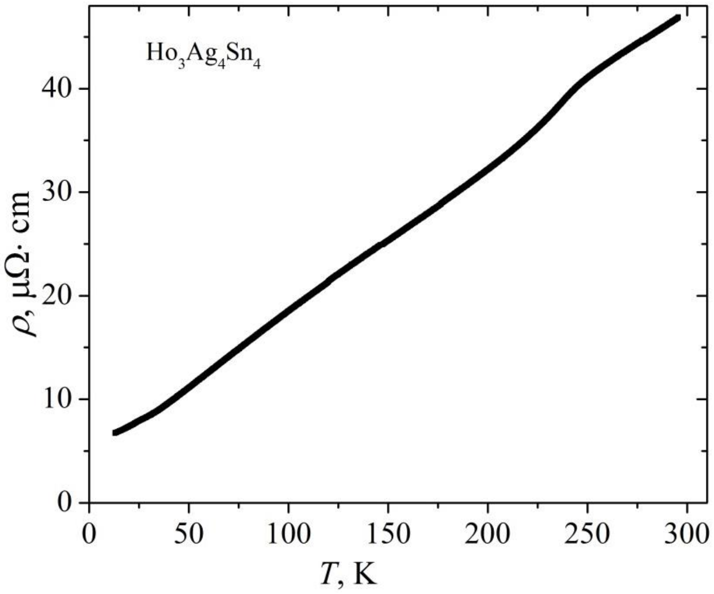


Fig. 5. Temperature dependence of the electrical resistivity *ρ(T)* of the $Ho_3Ag_4Sn_4$ compound.

Table 2

Parameters of the temperature dependencies of electrical resistivity of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho) compounds

Compound	$\rho_{290\ K}$, μΩ·cm	$\rho_{11\ K}$, μΩ·cm	$T_{order.}$, *K*	
			R*	M**
$Gd_3Ag_4Sn_4$	90.3	29.9	27.5	28; 8
$Tb_3Ag_4Sn_4$	101.5	14.7	27.2; 13.1	28; 13
$Dy_3Ag_4Sn_4$	52.7	11.9	15.7;	16
$Ho_3Ag_4Sn_4$	46.2	6.7	-	-

R* Data of the electrical property measurements.
M** Data of the magnetic property measurements [11-13].

Table 3

Approximation parameters of *ρ(T)* dependencies of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy) compounds (Bloch–Gruneissen–Mott formula, K_0 – residual resistivity; K_1 – temperature independent constant; K_2 –Debay temperature; K_3 – Mott coefficient)

Compound	K_0 (ρ_0, μΩ·cm)	K_1 (C)	K_2 (Θ_D, *K*)	K_3 (A, μΩ·cm/K^3)
$Gd_3Ag_4Sn_4$	34.98	160.4	201.1	$3.787\cdot10^{-8}$
$Tb_3Ag_4Sn_4$	20.99	83.2	69.6	$2.335\cdot10^{-7}$
$Dy_3Ag_4Sn_4$	12.36	50.0	83.9	$1.066\cdot10^{-7}$

takes place [12].

The slope change of the resistivity on the low-temperature part of the dependence of the electrical resistivity at 15.7 K for $Dy_3Ag_4Sn_4$ compound (Fig. 4, insert) is in a good agreement with temperature of the antiferromagnetic ordering at T_N = 16 K, determined from the studied behavior of the magnetic properties in Ref. [13].

As evidently from Fig. 5, the transition on the *ρ(T)* dependence for the $Ho_3Ag_4Sn_4$ compound was not observed up to 11 K. The magnetic properties of this compound were not studied, and we can only assume such transition at the temperatures lower than 11 K. The electrical transport parameters of the $R_3Ag_4Sn_4$ compounds are given in Table 2.

Above transition temperatures the temperature dependencies of the electrical resistivity of the $Gd_3Ag_4Sn_4$, $Tb_3Ag_4Sn_4$ and $Dy_3Ag_4Sn_4$ compounds are nearly linears caused by prevailing mechanism of phonon scattering. In this case $\rho(T)$ dependencies in the paramagnetic state can be approximated by Bloch–Grüneisen–Mott (BGM) formula [15]

$$\rho(T) = \rho_0 + 4RT\left(\frac{T}{\Theta_D}\right)^4 \int_0^{\Theta_D/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})} - AT^3$$

where the first term represents the scattering of the conduction electrons on static defects in the crystal structure (residual resistivity ρ_0); the second term describes the phonon contribution in total resistivity (parameter Θ_D is Debay temperature); third term accounts for Mott-type interband scattering processes. The calculated Debye temperature Θ_D of the $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy) compounds and approximation parameters of $\rho(T)$ dependencies are given in Table 3.

Obtained in our work results of the behavior of electrical properties of the $R_3Ag_4Sn_4$ compounds are in a good agreement with performed calculation of the density of electronic states (DOS) for isotypic $Sm_3Ag_4Sn_4$ stannide [16]. The performed calculations of the total DOS showed that the Fermi level is located in the continuous energy band and provided the metallic properties of the $Sm_3Ag_4Sn_4$ compound. Analysis of the electrical transport properties of the $R_3Ag_4Sn_4$ compounds showed their similarity to other series of the ternary compounds of rare earths and tin, e.g. $RAgSn_2$ [17], $R_3Cu_4Sn_4$ [18], RNi_3Sn_2 [19], $RNiSn_4$ [20], characterized by small values of the electrical resistivity and metallic type of conductivity.

Conclusions

Performed measurements of electrical properties of the $R_3Ag_4Sn_4$ compounds showed, that compounds with magnetic rare earth metals are characterized by presence of the transitions on electrical properties which connect to the temperatures of their magnetic orderings. Change of the resistivity caused by magnetic ordering was not observed for the $Ho_3Ag_4Sn_4$ compound in the used temperature range. Thus, electrical property measurements give an additional possibility to characterize the magnetic state of the intermetallics.

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Електротранспортні властивості сполук сполук $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho)

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Електротранспортні властивості інтерметалідів $R_3Ag_4Sn_4$ (R = Gd, Tb, Dy, Ho), які належать до структурного типу $Gd_3Cu_4Ge_4$ (просторова група *Immm*), досліджено в інтервалі температур 11 – 300 K. За результатами вимірювання температурних залежностей питомого електроопору встановлено, що досліджені сполуки характеризуються металічним типом провідності. Для станідів з Gd, Tb та Dy проаналізовано зв'язок магнітних та електричних властивостей. Для сполуки $Ho_3Ag_4Sn_4$ зміни питомого електроопору, пов'язаної з магнітним упорядкуванням, не виявлено в дослідженому температурному інтервалі.

Ключові слова: інтерметаліди, питомий електроопір, рентгенівський фазовий аналіз.