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Electrical resistivity and magnetism in some ternary intermetallics

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Abstract

We report on structure and transport properties of novel materials $Er_3Co_6Sn_5$, $Y_3Co_6Sn_5$, $HfCu_6Sb_3$, $Ti_5Cu_{0.45}Sb_{2.55}$, $Tm_2Ni_3Sn_2$ and Er_6Ni_2Sn studied in polycrystalline form in connection to their magnetic behavior. For the latter compound also the specific-heat data are presented. The first two compounds crystallize in the $Dy_3Co_6Sn_5$-type structure. $HfCu_6Sb_3$ forms in a new structure type characterized by space group $P6_3mc$ and lattice parameters $a=4.259$ Å and $c=11.5112$ Å. The W_5Si_3 structure type has been determined for $Ti_5Cu_{0.45}Sb_{2.55}$. Er_6Ni_2Sn possesses the orthorhombic Ho_6Ni_2Ga-type structure with lattice constants $a=9.245$ Å, $b=9.417$ Å and $c=9.819$ Å. Specific features of the temperature dependence of resistivity will be discussed in the context of results of magnetic measurements, e.g. the ρ vs. T curve for Er_6Ni_2Sn exhibits two pronounced cusp-like anomalies at $T_1\approx18$ K and $T_N\approx35$ K. These anomalies are connected with magnetic phase transitions confirmed by magnetization data.

Keywords: Rare-earth intermetallics; Electrical resistivity; Magnetic properties

1. Introduction

Magnetic properties of rare-earth intermetallics are mainly governed by magnetic moments located on 4f sites and their indirect exchange interaction by means of conduction electrons. Consequently, it is not surprising that sharp deviations from ordinary transport behavior, particularly near magnetic transition temperatures, are observed in the rare earth intermetallic compounds. The situation becomes more complex with transition metals involved in the material because of the appearance of the 3d-electron states at the vicinity of E_F [1,2].

It is also worth mentioning that the shape of the Fermi surface of the rare earth intermetallics remarkably deviates from the spherical free electron gas shape and can become more complex at the phase transition temperature [3].

In this paper, we report on the first results of our study of crystal structure and electrical resistivity, specific heat and magnetization measurements of several ternary rare-earth and transition metal intermetallic compounds, namely $Y_3Co_6Sn_5$, $Er_3Co_6Sn_5$, $Ti_5Cu_{0.45}Sb_{2.55}$, $Tm_2Ni_3Sn_2$, Er_6Ni_2Sn and $HfCu_6Sb_3$. Crystal-structure characteristics of some of these materials have been reported by Pöttgen [4].

2. Experimental

Polycrystalline samples were prepared by melting the corresponding amounts of elemental metals in an arc furnace on a water-cooled copper bottom under a protective argon atmosphere. The purity of Er, Y, Tm was 99.8%; Co was 99.9%; Cu (electrolytic), Sn and Sb was 99.99%; Ti was 99.97%; Hf was 99.93%. The alloys with Ti and Hf were annealed at 770 K, Er_6Ni_2Sn at 870 K and $(Er,Y)_3Co_6Sn_5$ at 670 K for 1 month and then quenched in cold water. The phase composition was checked by analyzing the X-ray powder patterns obtained on the DRON-2.0 diffractometer (FeK_α radiation) and HZG-4a (MoK_α radiation).

Electrical resistivity was measured by four-probe ac-method using standard lock-in technique in the temperature range from 4 K up to room temperature. Bars with typical dimensions $1.0\times1.5\times4$ mm were cut from polycrystalline ingots by spark erosion in the oil bath to prevent oxidation and internal stresses. The electrical contacts were made by

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colloidal silver paste, and then annealed at approximately 100°C for several hours. Depending on the sample resistance, which was several milliohms for all samples at room temperature, current through the sample was chosen in the range of 10–50 mA to increase the signal-to-noise ratio.

Measurements of specific heat and AC-susceptibility in the temperature range of 2–00 K were performed by means of a PPMS-14 measurement system (Quantum Design).

In addition, DC susceptibility data vs. temperature and magnetic fields have been collected at the Quantum Design SQUID magnetometer.

3. Results and discussion

The results of X-ray powder analysis performed on $Y_3Co_6Sn_5$, $Er_3Co_6Sn_5$, $Ti_5Cu_{0.45}Sb_{2.55}$, Er_6Ni_2Sn and $HfCu_6Sb_3$ are summarized in Table 1. It contains information about crystal structure type, space group, lattice constants and corresponding references if any were found. The $HfCu_6Sb_3$ compound crystallizes in a new structure type. $Tm_2Ni_3Sn_2$ forms a new modulated crystal structure, which has not yet been fully determined.

Fig. 1 presents the temperature dependence of electrical resistivity for all studied compounds measured in the temperature range 4.2–300 K. Absolute values of resistivity are rather high (hundreds of μΩ cm), which can be mainly caused by internal cracks and imperfect sample geometry.

In all the studied compounds the electrical resistivity decreases with temperature. The temperature dependence of resistivity of $Ti_5Cu_{0.45}Sb_{2.55}$ and $HfCu_6Sb_3$ exhibits behavior typical for transition metals without long-range magnetic ordering. Paramagnetic behavior of the former compound down to 4.5 K has been confirmed by magnetization measurements. The electrical resistivity in both compounds decreases linearly with temperature and then saturates obeying the power law at lower temperatures and exhibiting residual resistivity values of 168 and 78 μΩ cm, respectively. The ρ vs. T dependencies observed for the two isostructural compounds $Y_3Co_6Sn_5$ and $Er_3Co_6Sn_5$ significantly deviate from linearity, which is usually characteristic of materials with a strong scattering of conduction electrons in the Co 3d-band states.

The most interesting electrical resistivity behavior was found in Er_6Ni_2Sn. The temperature dependence of electrical resistivity exhibits two pronounced cusp-like anomalies at ≈35 and≈18 K. The specific heat as a function of temperature shown in Fig. 2 reveals three anomalies, namely at ≈35, 17 and 7.5 K, which indicate phase transitions in the system. These transitions are magnetic phase transitions as follows from magnetization and susceptibility data. All three transitions are revealed by the dc susceptibility measured in 0.1 T, whereas the ac susceptibility data displays two clear relatively sharp maxima pointing to the two lower-temperature transitions (Fig. 3). The anomaly around 7.5 K disappears already in a field of 0.5 T. The magnetization curves measured at various temperatures between 5 and 50 K, confirm that all magnetic phases below 35 K are of antiferromagnetic character (see Fig. 4), i.e. T_N≈35 K. The character of the specific-heat anomaly at T_N and lack of maximum on the temperature dependence of susceptibility (only strong field dependence of susceptibility is observed to set below T_N) is rather strange. The lower two transition temperatures are

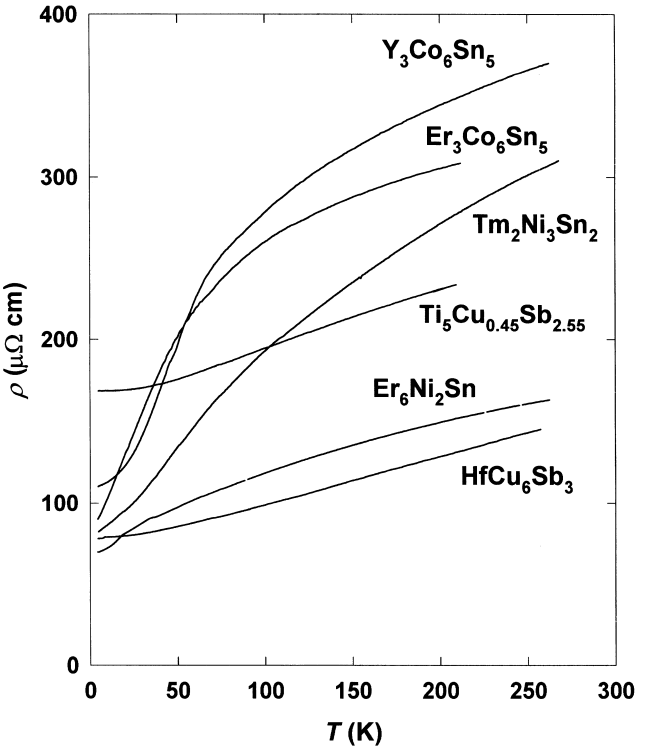

Fig. 1. Temperature dependence of electrical resistivity of $Y_3Co_6Sn_5$, $Er_3Co_6Sn_5$, $Tm_2Ni_3Sn_2$, $Ti_5Cu_{0.45}Sb_{2.55}$, Er_6Ni_2Sn and $HfCu_6Sb_3$.

Table 1
Crystal structures and lattice parameters

Compound	Structure type	Space group	a (Å)	b (Å)	c (Å)	Ref.
$Er_3Co_6Sn_5$	$Dy_3Co_6Sn_5$	*Immm*	4.295(1)	12.292(1)	9.626(1)	[4]
$Y_3Co_6Sn_5$	$Dy_3Co_6Sn_5$	*Immm*	4.303(1)	12.369(2)	9.701(1)	[4]
Er_6Ni_2Sn	Ho_6Co_2Ga	*Immm*	9.245(5)	9.417(9)	9.819(6)	[6]
$Ti_5Cu_{0.45}Sb_{2.55}$	W_5Si_3	*I4/mcm*	10.4741(4)	–	5.2374(2)	–
$HfCu_6Sb_3$	$HfCu_6Sb_3$	$P6_3mc$	4.259(6)	–	11.5112(5)	–

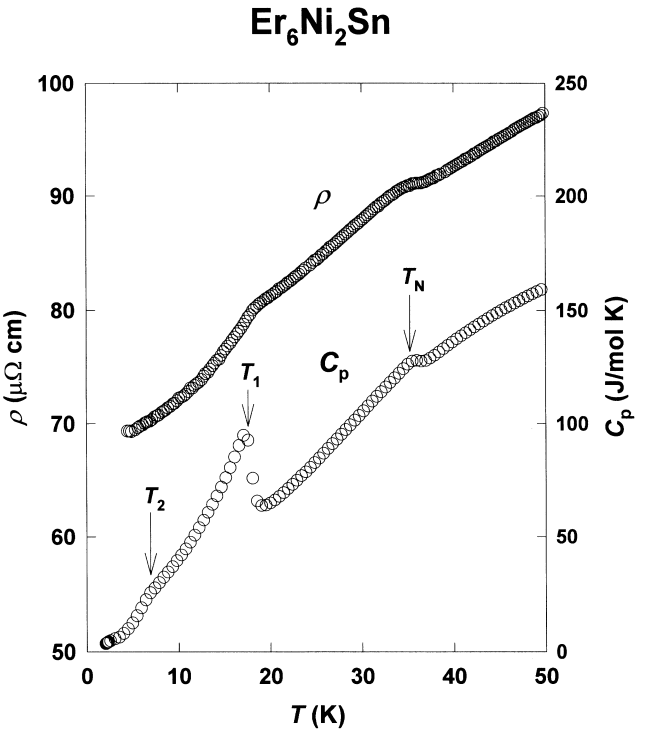

Fig. 2. Low temperature details of temperature dependence of electrical resistivity and specific heat measured on Er_6Ni_2Sn.

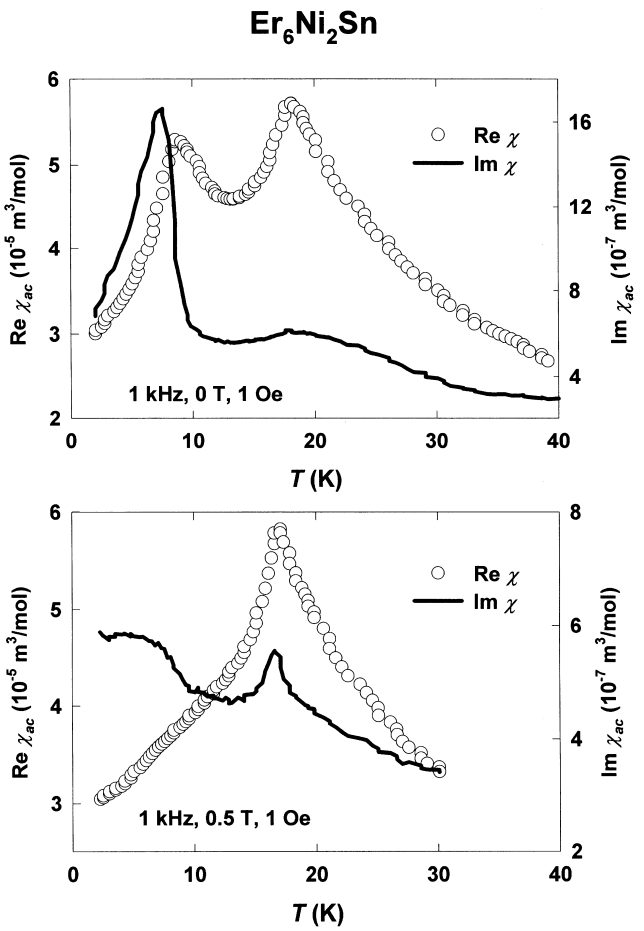

Fig. 3. Temperature dependence of ac-susceptibility of Er_6Ni_2Sn measured in fields of 0 and 0.5 T.

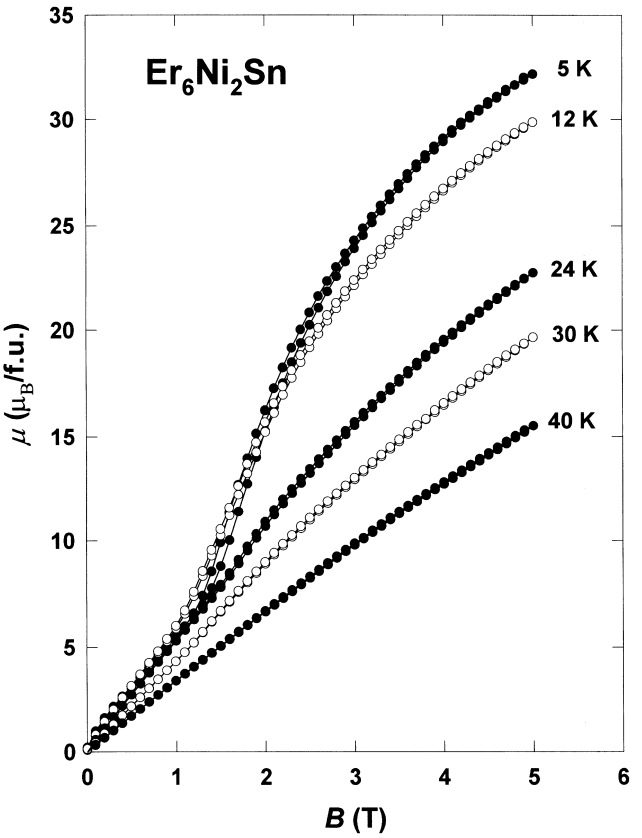

Fig. 4. Magnetization curves measured on Er_6Ni_2Sn at different temperatures.

denoted as T_1 and T_2 in Fig. 2. At 5 K, a metamagnetic transition is observed on a magnetization curve with the onset around 1 T. This transition is lifted up to ≈1.3 T when the temperature increases above 18 K; then it persists at temperatures up to ≈30 K, although it gradually becomes broader and less pronounced with increasing temperature. The metamagnetic transition above 1 T (1.3 T at temperatures above 18 K) observed on magnetization curves is apparently connected with a relatively easy collapse of antiferromagnetism under applied magnetic fields. Detailed results of magnetic study of this compound [5] showed direct connection of these anomalies with magnetic phase transitions. Note that the C_p vs. T data reveal an additional peculiarity at 7.5 K, which makes accurate determination of the electronic specific heat coefficient difficult from the presented data. Measurement of heat capacity down to 0.3 K is planned.

To determine the true nature of magnetic structures in Er_6Ni_2Sn, neutron diffraction experiments are in progress.

Acknowledgements

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