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Magnetic and electrical properties of R_2CuGe_6 compounds (R=Y, Ce, Nd, Gd, Tb, Dy, Ho, Er, Yb)

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Abstract

The temperature dependence of the magnetic susceptibility of the R_2CuGe_6 (R=Y, Gd, Tb, Dy, Ho, Er, Yb) compounds was measured in the range 84–300 K. It is shown that these rare earth compounds are Langevin type paramagnets except the Y_2CuGe_6 compound which is a Pauli paramagnet with low and temperature independent magnetic susceptibility. The resistivity of the compounds with R=Y, Ce, Nd, Gd, Tb, Ho, Yb was measured in the range 78–380 K. The differential thermopower of the Ce_2CuGe_6 and Tb_2CuGe_6 compounds was investigated in the range 78–380 K. Specific resistivity of the above-mentioned germanides increases linearly with increasing temperature and adopts values normally found in intermetallics. © 2000 Published by Elsevier Science S.A.

Keywords: Intermetallics; X-ray diffraction; Magnetic measurements; Electrical resistivity

1. Introduction

The crystal structure investigation of R_2CuGe_6 (R=Y, Ce, Nd, Gd, Tb, Dy, Ho, Er, Yb) compounds was reported before [1]. The structure of these compounds belongs to the Ce_2CuGe_6 type (*Amm2* space group). In accordance with Ref. [2], the investigation of the temperature dependence of the magnetic susceptibility for the Ce_2CuGe_6, Pr_2CuGe_6 or Nd_2CuGe_6 compounds has shown that Curie–Weiss behaviour is followed whereas Sm_2CuGe_6 behaves as a Van Vleck paramagnet in the 20–100 K temperature region. The aim of this work is to investigate the static magnetic and transport properties of R_2CuGe_6 compounds where R=Y, Ce, Nd, Gd, Tb, Dy, Ho, Er and Yb.

2. Experimental details

Samples were prepared by direct arc melting the constituent elements (rare earths, purity 99.9 wt.%; copper electrolytic, purity 99.99 wt.%; germanium, purity 99.99 wt.%) under high-purity argon atmosphere on a water-cooled copper hearth. The weight losses of as-cast ingots were less than 0.5%. The alloys were then annealed at 870 K in evacuated quartz tubes for 1 month and quenched in cold water. Phase analyses were carried out using powder patterns obtained on a DRON-3.0 powder diffractometer (CuKα radiation).

The magnetic susceptibility was determined using a Faraday balance in the range 84–300 K in magnetic fields up to 0.8 MA m^{-1}. The electrical resistivity and differential thermopower were measured as described previously [3].

3. Results

Magnetic susceptibility measurements on R_2CuGe_6 (R=Y, Gd, Tb, Dy, Ho, Er, Yb) compounds in the range 84–300 K reveal a paramagnetic state in these compounds. The susceptibility of the Y_2CuGe_6 compound is nearly independent of temperature and changes from 0.31×10^{-6} emu g^{-1} at 84 K to 0.39×10^{-6} emu g^{-1} at 300 K when measured in various magnetic fields up to 0.8 MA m^{-1} (Fig. 1). The $\chi(T)$ curve for the other R_2CuGe_6 germanides where R=Gd, Tb, Dy, Ho, Er, Yb, can be described by the Curie–Weiss law. From $\chi^{-1}(T)$ plots (Figs. 1 and 2) the paramagnetic Curie temperature θ_p and the effective magnetic moment (μ_{eff}) per R atom were calculated (Table 1). The μ_{eff}/R values are close to the respective $g_J(J(J+1))^{1/2}$ values for R^{3+} cations. The θ_p

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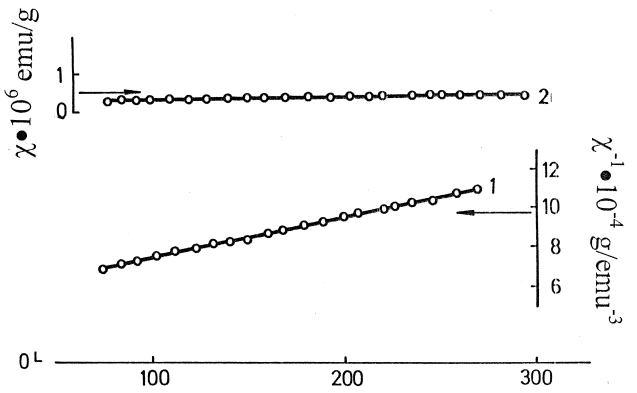

Fig. 1. Inverse susceptibility vs. temperature: (1) Yb_2CuGe_6; (2) Y_2CuGe_6.

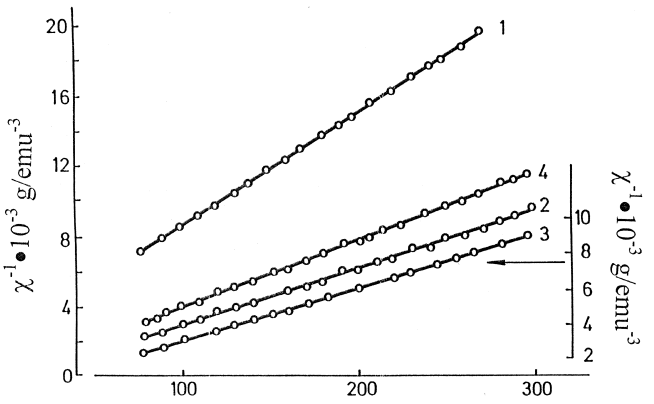

Fig. 2. Inverse susceptibility vs. temperature: (1) Gd_2CuGe_6; (2) Dy_2CuGe_6; (3) Ho_2CuGe_6; (4) Er_2CuGe_6.

values are negative for all compounds except for Er_2CuGe_6.

The temperature dependencies of the resistivity in the range 78–380 K are linear for these ternary germanides where R=Ce, Nd, Gd, Tb, Ho (Figs. 3 and 4). The $\rho(T)$ plots are characterized by a negative curvature at low temperature only for the Yb_2CuGe_6 and Y_2CuGe_6 compounds (Fig. 4). The values of the resistivity at 80 K and 380 K for the various compounds are listed in the table.

The differential thermopower (α) of the Ce_2CuGe_6 and Tb_2CuGe_6 compounds was measured in the range 78–400 K. The thermopower is positive and gradually increases with increasing temperature for the Tb_2CuGe_6 compound (0.1–3.1 $\mu V\,K^{-1}$). The differential thermopower with respect to copper in the Ce_2CuGe_6 compound has a maximum. There are signs of reversal of α at both low and high temperatures. The absolute values of the differential thermopower for the Ce_2CuGe_6 compound are very low.

Table 1
Magnetic and electrical data of the ternary compounds R_2CuGe_6 (R=Gd, Tb, Du, Ho, Er, Yb)

Compound	μ_{eff}^{meas} (μ_B)	Θ_p (K)	$\mu_{eff/R}^{theor}$ (μ_B)	ρ ($\mu\Omega$ m)	
				80 K	380 K
Ce_2CuGe_6	–	–	–	1.9	2.7
Nd_2CuGe_6	–	–	–	0.8	2.5
Y_2CuGe_6	–	–	–	0.3	1.3
Gd_2CuGe_6	7.3	−43	7.94	0.8	2.3
Tb_2CuGe_6	10.2	−20	9.72	0.8	2.0
Du_2CuGe_6	10.5	−12	10.65	–	–
Ho_2CuGe_6	10.6	−2	10.61	0.4	1.1
Er_2CuGe_6	9.3	1	9.58	–	–
Yb_2CuGe_6	4.1	−202	4.54	0.8	2.8

4. Discussion

The investigation of the magnetic properties of the R_2CuGe_6 compounds (where R=Y, Gd, Tb, Dy, Ho, Er, Yb) offers the possibility to establish the R atoms' valence state. We may conclude that there is no localized magnetic moments on the Cu atoms ($3d^{10}$ configuration, $^1S_0$ ground state). Also the Ge atoms have no magnetic moments. The effective magnetic moments μ_{eff} of the R_2CuGe_6 compounds calculated per R atom are close to the respective free ion values of the R^{3+} ions and this is evidence of the trivalent rare earth atomic state in the investigated compounds.

The negative values of the paramagnetic Curie points for Gd_2CuGe_6, Dy_2CuGe_6 and Ho_2CuGe_6 open the possibility of antiferromagnetic ordering in these compounds at low temperature.

The θ_p value for Yb_2CuGe_6 essentially differs from those of the other compounds (Table 1). The moment μ_{eff}/Yb is less than the respective $g_J(J(J+1))^{1/2}$ value for the Yb^{3+} ion. This difference in magnetic characteristics may be explained by the possibility of a mixed valence state of the Yb atoms in this compound. The small energy difference between the Yb^{2+} and Yb^{3+} states allows the ytterbium atoms to be in a mixed valence state (inter-configurational fluctuations). In any case, the μ_{eff}/Yb values show that the valence of Yb is not equal to +3. There is a small maximum for the Yb_2CuGe_6 compound in the dependence of the cell volume V on the R atomic number (Fig. 5). This also points to an intermediate valence state of Yb in this ternary germanide.

The Pauli paramagnetism in the Y_2CuGe_6 compound is obvious because all ingredients have no localized magnetic moment. The insignificant increase in the magnetic susceptibility with increasing temperature indicates that the Fermi level resides in a density-of-state minimum in this compound.

The resistivity of R_2CuGe_6 where R=Y, Nd, Gd, Tb, Ho, Yb, linearly increases with increasing temperature and is typical for intermetallics. The linearity in the $\rho(T)$ plots indicates that there is mainly phonon scattering above

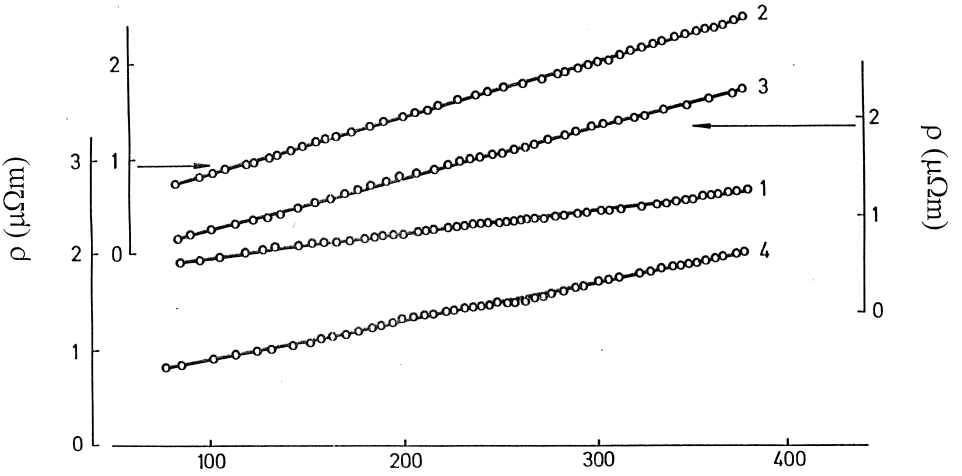

Fig. 3. Temperature dependence of the electrical resistivity: (1) Ce_2CuGe_6; (2) Nd_2CuGe_6; (3) Gd_2CuGe_6; (4) Tb_2CuGe_6.

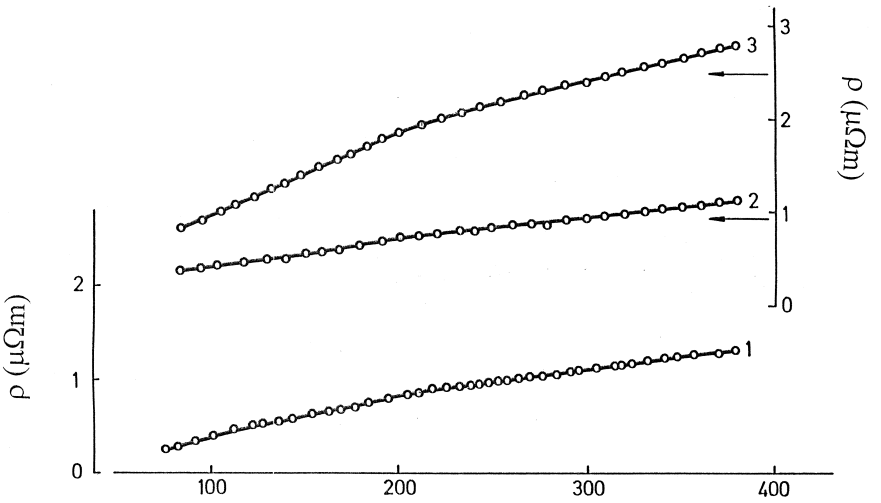

Fig. 4. Temperature dependence of the electrical resistivity: (1) Y_2CuGe_6; (2) Ho_2CuGe_6; (3) Yb_2CuGe_6.

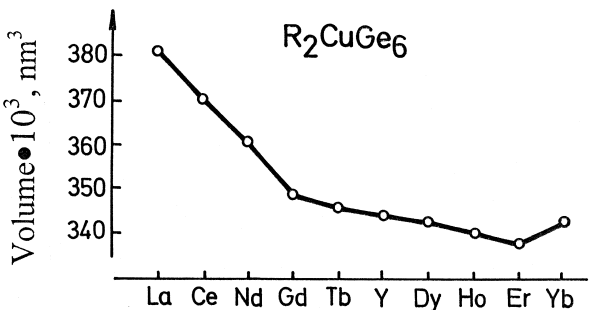

Fig. 5. Unit cell volume of R_2CuGe_6.

$(2/3)\theta_D$ (Debye temperature) in these compounds. The negative curvature of $\rho(T)$ in Y_2CuGe_6 below ~220 K may correspond to the transition from $\rho(T)$ linearity to $\rho(T^n)$ dependence. It is seen that the resistivity of the Y_2CuGe_6 compound has the lowest values compared to the other isostructural compounds. The higher resistivity in the other compounds may be the result of spin disorder scattering. This additional resistance is temperature independent.

Yb_2CuGe_6 is characterized by a considerable negative curvature in its $\rho(T)$ plot. The unusual behaviour of this resistivity compared with the $\rho(T)$ curves of the other

R_2CuGe_6 phases also emphasizes the conclusion about a mixed valence state of ytterbium in its compound. This resistance peculiarity is caused by additional scattering due to charge fluctuations associated with variable occupation of the Yb 4f shell. The increase with temperature of the resistivity in Ce_2CuGe_6 is less than in the other germanides. However, neither $\rho(T)$ nor $\alpha(T)$ are typical for cerium in a mixed valence compound.

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