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Formation, crystal chemistry and magnetism of compounds RE_2TGe_6, RE ≡ rare earth, T ≡ Pd, Pt, Cu, Ag and Au

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Abstract

Novel ternary compounds RE_2TGe_6 (RE ≡ Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb; T ≡ Pd, Pt, Ag, Au) were synthesized by argon arc melting followed by a heat treatment at 600 or 800 °C for 150 h. From X-ray powder analysis all compounds were found to be isotypic with structure type Ce_2CuGe_6. Magnetic susceptibilities were measured for RE_2CuGe_6 (RE ≡ Ce, Pr, Nd, Sm) and Ce_2PdGe_6. All compounds exhibit antiferromagnetic ordering below 20 K. The paramagnetic effective moments agree with the values for tripositive RE ion moments. The Sm containing sample reveals typical Van Vleck type paramagnetism above the Néel point.

Keywords: Ternary compounds; X-ray powder diffraction; Antiferromagnetic compounds

1. Introduction

Crystallographic characterization of Ce_2CuGe_6 has been provided by Konyk et al. [1]. Isotypic compounds have been observed in the holmium–noble metal–germanium ternary systems and precise atom parameters have been derived recently for Ho_2PdGe_6 [2]. The continued scan for strongly correlated electron systems among ternary cerium containing alloys has prompted us towards investigation of the compounds RE_2TGe_6, where RE is one of the light rare earth metals.

In this paper we report on the crystal structure of the RE_2TGe_6 series of compounds and magnetic studies of Ce_2TGe_6 (T ≡ Pd, Cu).

2. Experimental details

The alloys, each with a total weight of ca. 1 g, were synthesized by argon arc melting from ingots of high purity elements obtained from Johnson & Matthey and Co, UK (99.9 mass%). Weight losses were less than 1 mass%. The arc melted buttons were sealed in evacuated quartz tubes and annealed for 150 h. Technical details of for example duration and temperature of the homogenizing anneal of the alloys prior to quenching of the silica capsules in water are given in Table 1.

Lattice parameters and standard deviations (see Table 1) were obtained by least squares refinement of room temperature Guinier-Huber X-ray powder data using monochromatic Cu $K\alpha_1$ radiation. The magnetic measurements were performed in the temperature range below 100 K down to 4.5 K using a Lake Shore a.c. susceptometer (a.c. field 1 mT, 133.3 Hz) and a SHE SQUID magnetometer.

3. Results and discussion

3.1. Formation of compounds

Room temperature X-ray patterns of samples of RE_2TGe_6, T ≡ Pd, Pt, Cu, Ag, Au revealed close resemblance to the X-ray intensity pattern of Ce_2CuGe_6 and were indexed completely on the basis of a base-centered orthorhombic unit cell (Table 1). Using the atom parameters derived for Ho_2PdGe_6 [2], excellent agreement is obtained between the experimentally observed and calculated X-ray powder intensities, confirming isotypism with the crystal structure of Ce_2CuGe_6.

The formation of a rather complete RE_2TGe_6 series of compounds is found in the combinations with the nickel group (palladium, platinum) and copper, whereas the stabil-

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Table 1
Crystallographic data of ternary compounds RE_2TGe_6, $T \equiv$ Pd, Pt, Cu, Ag, Au; structure type Ce_2CuGe_6, space group *Amm*2, No. 38, origin on *mm*2 $Z=2$

Compound	Heat treatment (°C)	Lattice parameters (nm) a	b	c	V (nm³)
Y_2PdGe_6	600	0.40790(4)	0.40168(5)	2.1525(2)	0.3526(1)
La_2PdGe_6	600	0.42117(3)	0.41100(3)	2.2265(5)	0.3854(13)
Ce_2PdGe_6	800	0.41784(3)	0.40900(2)	2.2043(2)	0.3767(1)
Pr_2PdGe_6	600	0.4152(2)	0.4073(2)	2.197(1)	0.3716(2)
Nd_2PdGe_6	600	0.4139(2)	0.4060(2)	2.187(1)	0.3674(3)
Sm_2PdGe_6	600	0.4116(3)	0.4044(3)	2.170(2)	0.3612(3)
Gd_2PdGe_6	600	0.40953(5)	0.40320(6)	2.1624(5)	0.3570(1)
Tb_2PdGe_6	600	0.40830(6)	0.40227(6)	2.1530(5)	0.3536(1)
Dy_2PdGe_6	600	0.40738(5)	0.40163(6)	2.1485(7)	0.3515(1)
Ho_2PdGe_6	600	0.40441(8)	0.40026(9)	2.1571(5)	0.3491(1)
Er_2PdGe_6	600	0.40574(4)	0.40051(5)	2.1402(4)	0.3477(1)
Tm_2PdGe_6	600	0.40479(6)	0.39954(7)	2.1346(5)	0.3452(5)
Yb_2PdGe_6	600	0.40755(3)	0.39934(3)	2.1851(3)	0.3556(1)
Y_2PtGe_6	800	0.40574(4)	0.40125(3)	2.1677(4)	0.3529(1)
Ce_2PtGe_6	600	0.41503(9)	0.40858(9)	2.2183(7)	0.3761(1)
Pr_2PtGe_6	600	0.41348(7)	0.40705(6)	2.2084(5)	0.3716(1)
Nd_2PtGe_6	600	0.41201(12)	0.40630(11)	2.2007(7)	0.3684(2)
Sm_2PtGe_6	600	0.40955(6)	0.40412(6)	2.1871(2)	0.3619(1)
Gd_2PtGe_6	600	0.40693(5)	0.40239(4)	2.1791(5)	0.3568(1)
Tb_2PtGe_6	600	0.40634(6)	0.40155(7)	2.1648(5)	0.3532(1)
Dy_2PtGe_6	800	0.40430(3)	0.40050(4)	2.1672(3)	0.3509(1)
Ho_2PtGe_6	600	0.40441(8)	0.40026(9)	2.1571(5)	0.3491(1)
Er_2PtGe_6	800	0.40315(5)	0.39977(5)	2.1573(5)	0.3477(1)
Tm_2PtGe_6	800	0.40247(6)	0.39936(6)	2.1539(6)	0.3461(1)
Yb_2PtGe_6	600	0.40482(4)	0.39771(4)	2.1940(5)	0.3532(1)
Ce_2CuGe_6 [a]	600	0.40756(2)	0.42152(7)	2.15408(7)	0.3621(1)
	600	0.42116(5)	0.40725(4)	2.1584(4)	0.3702(1)
Pr_2CuGe_6	800	0.41950(3)	0.40585(2)	2.1485(2)	0.3657(1)
Sm_2CuGe_6	600	0.41496(5)	0.40275(5)	2.1221(3)	0.3546(1)
Tm_2CuGe_6	800	0.40657(4)	0.3963(4)	2.0767(5)	0.3346(1)
La_2AgGe_6	600	0.43409(6)	0.41598(5)	2.1901(4)	0.3954(1)
Ce_2AgGe_6	600	0.43089(3)	0.4144(5)	2.1674(5)	0.3870(1)
Pr_2AgGe_6	600	0.42887(6)	0.41295(6)	2.1610(5)	0.3827(1)
Nd_2AgGe_6	600	0.42709(5)	0.41169(6)	2.1517(5)	0.3783(1)
Sm_2AgGe_6	600	0.42394(5)	0.40956(5)	2.1346(8)	0.3706(1)
Gd_2AgGe_6	600	0.41946(7)	0.40583(8)	2.1166(7)	0.3603(1)
Ce_2AuGe_6	600	0.42587(5)	0.41263(5)	2.1852(4)	0.3840(1)
Pr_2AuGe_6	600	0.42412(5)	0.41135(4)	2.1772(4)	0.3798(1)
Nd_2AuGe_6	600	0.42265(5)	0.41272(6)	2.1689(4)	0.3764(1)
Sm_2AuGe_6	600	0.41945(5)	0.40792(4)	2.1507(5)	0.3679(1)
Gd_2AuGe_6	600	0.41325(4)	0.40234(4)	2.1187(3)	0.3522(1)
Tb_2AuGe_6	600	0.41325(4)	0.40234(4)	2.1187(3)	0.3522(1)
Dy_2AuGe_6	600	0.41065(6)	0.39918(6)	2.1006(3)	0.3443(1)

[a] Data from Ref. [1].

ity of silver- and gold-containing compounds is reduced with decreasing radius of the rare earth element and Gd_2TGe_6 ($T \equiv$ Ag, Au) are the smallest end members observed.

3.2. Crystallochemistry

The plot of the unit-cell dimensions of the isostructural compounds vs. the rare earths is shown in Fig. 1. The cerium

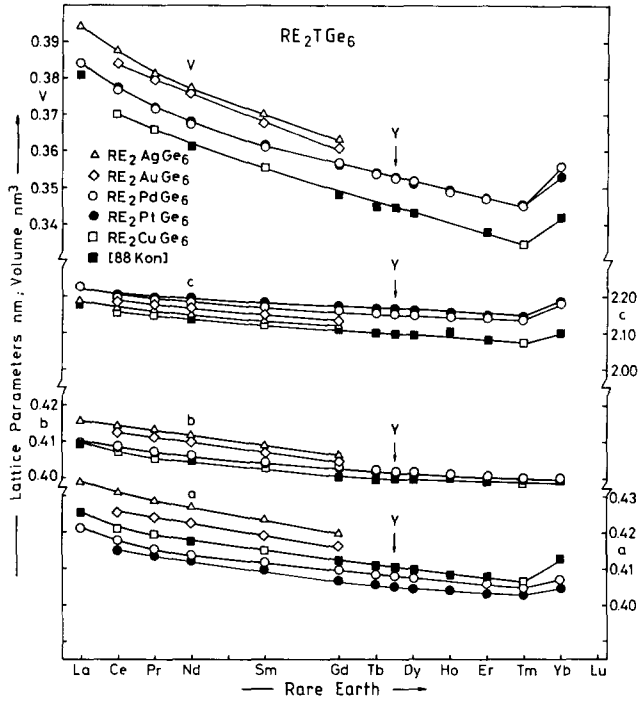

Fig. 1. Lattice parameters for the ternary compounds RE_2TGe_6 vs. rare earths. The positions of the values for yttrium are marked by an arrow.

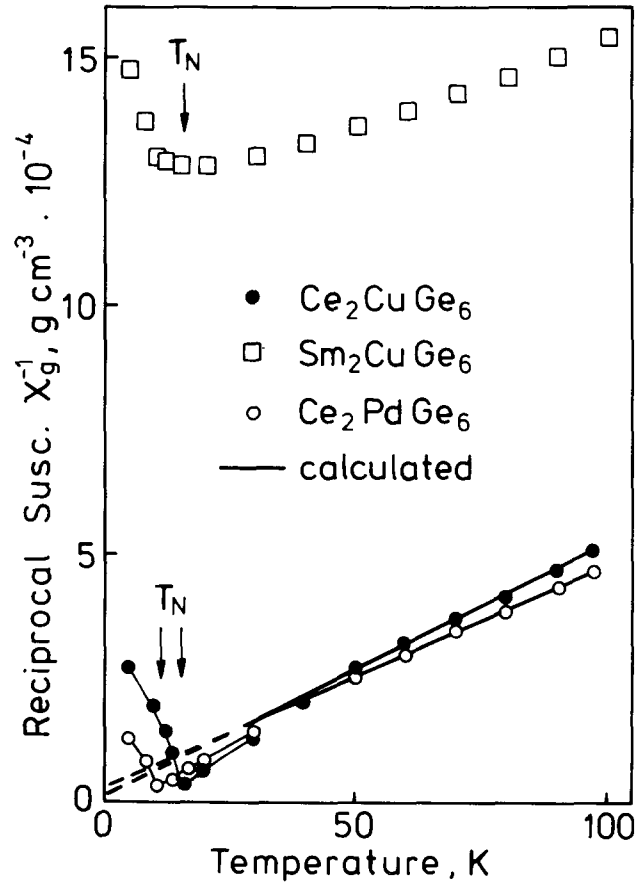

Fig. 2. Inverse susceptibility vs. temperature for $Ce_2(Cu,Pd)Ge_6$ and Sm_2CuGe_6.

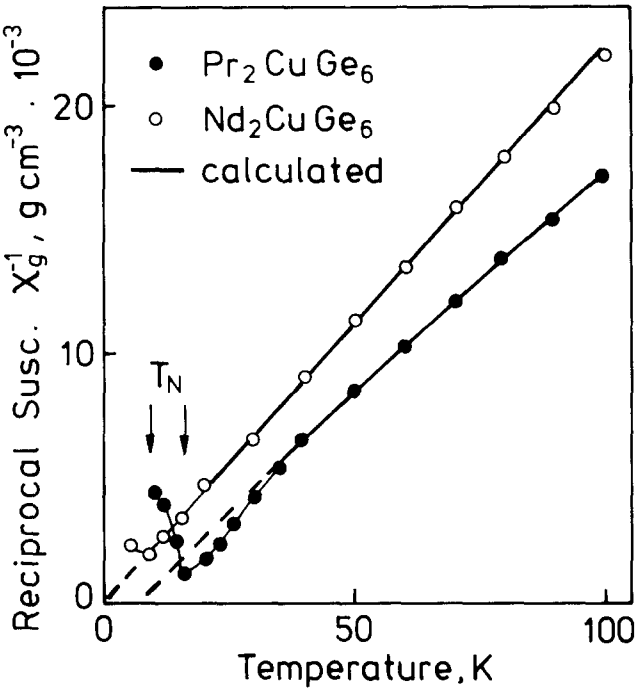

Fig. 3. Inverse susceptibility vs. temperature for $(Pr,Nd)_2CuGe_6$.

compounds do not display any deviation from the decreasing trivalent lanthanide mean atomic volume.

Positive deviations are observed for the ytterbium-containing compounds, suggesting a substantial presence of divalent ytterbium. As usual the unit-cell volume of the yttrium-containing compounds is found between the values for the corresponding terbium and dysprosium phases.

3.3. Magnetic behavior

The magnetic behavior of the RE_2CuGe_6 compounds as well as Ce_6PdGe_6 is summarized in Figs. 2, 3 and Table 2. Above $T = 20$ K the samples reveal the typical temperature dependent paramagnetism, closely obeying the Curie–Weiss law, with the exception of Sm_2CuGe_6. The effective moments were calculated by least squares fit using the relation $\chi = C/(T-\theta)+\chi_0$ and are in good accord with theoretical values for RE^{3+} ions. Sm_2CuGe_6 shows paramagnetic behavior owing to the closely spaced multiplets. On cooling the samples below 20 K the magnetic susceptibilities reveal typical maxima (Néel points) owing to an antiparallel spin alignment with the rare-earth sublattices. The positive paramagnetic Curie temperature in the case of $(Pr,Nd)_2CuGe_6$ indicate the possible formation of ferromagnetic layers which are antiferromagnetically coupled.

Table 2
Magnetic data of ternary compounds RE_2TGe_6, RE ≡ Ce, Pr, Nd, Sm; T ≡ Cu, Pd

Compound	T_N (K)	Θ_P (K)	μ_{eff}^{meas} (μ_B)	$\mu_{eff/RE}^{theor}$ (μ_B)
Ce_2CuGe_6	15.8	−2	2.5	2.54
Pr_2CuGe_6	16.0	7	3.7	3.62
Nd_2CuGe_6	9.0	1	3.6	3.64
Sm_2CuGe_6	16.0	−106	1.2 [a]	–
Ce_2PdGe_6	11.5	−7	2.7	2.54

[a] Van Vleck paramagnetism.

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