Magnetic and Electrical Behavior in CeMgGa

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The intermetallic cerium compound CeMgGa has been synthesized in quantitative yield by reacting the elements in a sealed niobium tube in a high-frequency furnace. The structure was determined from single-crystal X-ray data: ZrNiAl type, $P\bar{6}2m$, $a = 752.7(2)$ pm, $c = 454.8(1)$ pm, $wR2 = 0.0497$, 275 F^2 values, and 14 variables. The structure contains two crystallographically different gallium positions which both have tricapped trigonal prismatic coordinations: [$Ga1Mg_6Ce_3$] and [$Ga2Mg_3Ce_6$]. The cerium atoms have coordination number 15 in the form of a distorted pentacapped pentagonal prism. Together, the magnesium and gallium atoms build a three-dimensional [MgGa] network in which the cerium atoms fill distorted pentagonal channels. CeMgGa exhibits localized magnetism due to the presence of stable Ce^{3+} ions and orders antiferromagnetically at $T_N = 3.1(1)$ K. The compound studied shows metallic conductivity.

Introduction

Equiatomic cerium intermetallics CeTX (T = transition metal, X = element of the 3rd, 4th, or 5th main-group) have attracted considerable interest in recent years with respect to their largely varying magnetic and electrical properties.[1−55] Prominent examples are the Kondo systems CeAuAl,[42] CeNiSn and CeRhSb,[47] the metamagnet CePtAs,[41] mixed-valent CeIrAl[44] and CeNiAl[39], or the ferromagnets CeAgGa[28] and CeAuGe.[55]

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Most structures of these intermetallics derive from the AlB_2 type.[56] The transition metal and *p* element atoms build two- or three-dimensional [TX] polyanionic networks where the cerium atoms separate these networks or fill cages or channels within them.

Very recently, some of the CeTX intermetallics have been investigated with respect to their hydrogen absorption behavior. This leads to very interesting changes in the magnetic properties. To give some examples, the hydrogen insertion in CeNiIn induces a valence transition from intermediate valence to the trivalent state in $CeNiInH_{1.8}$;[57] CeAuAl shows an increase of the Néel temperature upon hydrogenation to $CeAuAlH_{1.4}$;[58] and the Curie temperature of $CePtAlH_{1.1}$ is twice as large as that for CePtAl.[59]

Another recent research strategy concerns the substitution of the X component by a typical divalent metal, i.e., magnesium or cadmium.[60,61] This way it is possible to change the electron count and thus the magnetic properties. An important parameter for all the transition-metal-containing CeTX compounds is the *d*−*f* hybridization between the cerium and transition metal atoms. In going from CeNiIn to CePdIn and CePtIn, the magnetic behavior changes from a valence fluctuating to a paramagnetic and antiferromagnetic heavy fermion state.[34] In this context it seems interesting to investigate similar compounds without a transition metal component. Magnetic ordering has been observed in CeGaGe,[22,62] CeAlSi and CeAlGe,[63] and CeGaSi.[64,65] Our recent investigation in the cerium−magnesium−gallium system led to the new 3.1(1) K antiferromagnet CeMgGa. The synthesis, structure determination, and physical properties are reported herein.

Experimental Procedures

Synthesis. Starting materials for the syntheses of CeMgGa were ingots of cerium (Johnson-Matthey, >99.9%), a magnesium rod (Johnson Matthey, ∅ 16 mm, >99.95%), and gallium rods (VAW, >99.9%). The gallium rods were crushed to small pieces at liquid nitrogen temperature. The surface of the magnesium rod was first cut on a turning lathe to remove surface impurities. Small platelets cut from the remaining rod were then kept under argon. In a next step the cerium ingots were cut into small pieces under dried paraffin oil. The latter was washed off with dried (over sodium wire) *n*-hexane and the pieces were melted to buttons in an arc-melting furnace[66] under an argon atmosphere.

The cerium button, pieces of magnesium, and gallium were mixed in the ideal 1:1:1 atomic ratio and sealed in a small niobium ampule[66] under an argon pressure of 800 mbar. The argon was purified over silica gel, molecular sieves, and titanium sponge (900 K). The tube was placed in a quartz glass sample chamber of a high-frequency furnace (Hüttinger Elektronik, Freiburg, Typ TIG 1.5/300) under flowing argon.[67] The tube was first heated at about 1300 K until the start of the reaction which was indirectly visible by a weak heat flash. Then the sample was held at about 870 K for another 2 h followed by quenching. The sample could be separated easily from the niobium tubes by mechanical fragmentation. No reactions with the crucible material could be detected. Powders and polycrystalline samples of CeMgGa are not stable in moist air over long periods of time. Complete deterioration is observed in water. Single crystals exhibit metallic luster and powders are dark gray.

The sample was investigated by EDX analyses using a Leica 420 I scanning electron microscope with CeO_2, gallium phosphide, and MgO as standards. The composition was close to the ideal value. No impurity elements could be detected.

Structural Characterization. The sample was characterized through a Guinier powder pattern using Cu $K\alpha_1$ radiation and α-quartz (a = 491.30 pm, c = 540.46 pm) as an internal standard. The pattern could be indexed readily on the basis of a hexagonal unit cell with the lattice parameters listed in Table 1. To ensure correct indexing, the observed pattern was compared with a calculated one[68] taking the positions of the refined structure.

Several small irregularly shaped single crystals of CeMgGa could be isolated from the annealed sample by mechanical fragmentation. They were first investigated on a Buerger precession camera equipped with an image plate system (Fujifilm BAS-1800) in order to establish suitability for intensity data collection. Intensity data were recorded at room temperature by use of a four-circle diffractometer (CAD4) with graphite monochromatized MoKα radiation (λ = 71.073 pm) and a scintillation counter with pulse-height discrimination.

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Table 1. Crystal Data and Structure Refinement for CeMgGa

empirical formula	CeMgGa
molar mass	234.15 g/mol
lattice parameters	$a = 752.7(2)$ pm
(Guinier powder data)	$c = 454.8(1)$ pm
	$V = 0.2232$ nm^3
formula units per cell	$Z = 3$
space group	$P\bar{6}2m$
calculated density	5.23 g/cm^3
crystal size	$10 \times 30 \times 50$ μm^3
transmission (max:min)	1.23
absorption coefficient	24.0 mm^{-1}
F(000)	303
θ range	3 to 30°
range in hkl	$\pm 10, \pm 10, -6 \le l \le +3$
total no. reflections	2121
independent reflections	275 ($R_{int} = 0.0989$)
reflections with $I > 2\sigma(I)$	265 ($R_{sigma} = 0.0423$)
data/restraints/parameters	275/0/14
goodness-of-fit on F^2	1.073
final R indices [$I > 2\sigma(I)$]	$R1 = 0.0246$; $wR2 = 0.0495$
indices (all data)	$R1 = 0.0266$; $wR2 = 0.0500$
extinction coefficient	0.006(1)
absolute structure parameter	0.05(9)
largest diff. peak and hole	1.15 and −1.02 e/Å^3

The scans were taken in the $\omega/2\theta$ mode and an empirical absorption correction was applied on the basis of psi-scan data.

The isotypy of CeMgGa with the hexagonal ZrNiAl type[69] was already evident from the Guinier powder data. The atomic parameters of CeAgMg[70] were taken as starting values, and the structure was refined with anisotropic displacement parameters using SHELXL-97[71] (full-matrix least-squares on F^2). Refinement of the Flack[72,73] parameter indicated the wrong absolute structure. The atomic parameters were inverted for the subsequent least squares cycles.

As a check for the correct compositions, the occupancy parameters were varied in a separate series of least-squares cycles along with the displacement parameters. All sites were fully occupied within two standard deviations, and in the final cycles the ideal composition was assumed. The X-ray data gave no hint for gallium/magnesium mixing. A final difference Fourier synthesis revealed no significant residual peaks. All relevant crystallographic details are listed in Table 1. Atomic coordinates and interatomic distances are given in Tables 2 and 3. Listings of the anisotropic displacement parameters and the structure factors are available.[74]

Physical Property Investigations. Magnetic measurements were carried out in the temperature range 1.7−400 K in applied fields up to 5 T using a Quantum Design MPMS-5 SQUID magnetometer. The electrical resistivity was measured over the temperature range 1.2−270 K employing a conventional dc four-point technique. The isothermal transverse magnetoresistivity was taken at $T = 4.2$ K up to $B = 8$ T.

Results and Discussion

Crystal Chemistry. CeMgGa is a new compound in the large family of intermetallics with ZrNiAl type structure.[69,75] As already mentioned in the Introduction, a large series of CeTX compounds with ZrNiAl structure have a three-dimensional [TX] polyanionic network. In CeMgGa this network is formed only by main group elements. A projection of the CeMgGa structure onto the *xy* plane is presented in Figure 1. From a geometrical point of view, this structure is built up from two types of gallium centered trigonal prisms, [Ga1Mg_6-Ce_3] and [Ga2Ce_6Mg_3]. The square faces of these trigonal prisms are capped by cerium atoms for Ga1 and by magnesium atoms for Ga2 resulting in coordination number nine for both gallium sites. The trigonal prisms are condensed via common faces and edges.

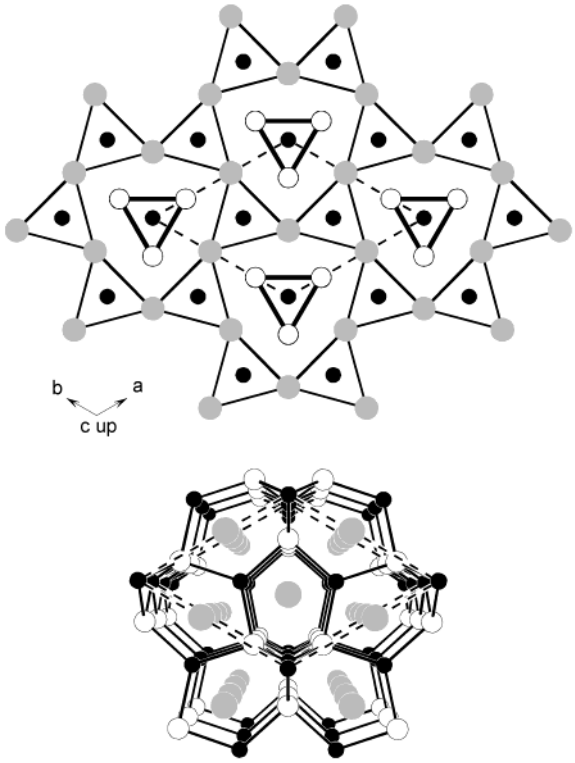

Figure 1. Crystal structure of CeMgGa. The upper drawing shows a projection onto the *xy* plane. All atoms lie on mirror planes at $z = 0$ (thin lines) and $z =$ (thick lines), respectively. The cerium, magnesium, and gallium atoms are drawn as gray, open, and filled circles, respectively. A perspective view along the *c* axis is shown in the lower drawing. The three-dimensional [MgGa] network is emphasized.

Together, the magnesium and gallium atoms form a three-dimensional [MgGa] network. Each magnesium atom has four gallium neighbors at Mg−Ga distances of 291 and 292 pm, significantly larger than the sum of the covalent radii of 261 pm.[76] However, a similar range of Mg−Ga distances has been observed in the binary compounds $MgGa_2$ (280−331 pm)[77] and Mg_2Ga_5 (287−297 pm).[78] The cerium atoms have coordination number 15 (5 Ga + 6 Mg + 4 Ce). The Ce−Ga distances (318 and 320 pm) are in the same range as in $Ce_{4.8}Ga_{3.2}$ (318−342 pm),[79] and also the Ce−Mg distances (340−358 pm) compare well with the shortest Ce−Mg dis-

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Table 2. Atomic Coordinates and Anisotropic Displacement Parameters for CeMgGa

atom	Wyckoff position	*x*	*y*	*z*	U_{11}	U_{22}	U_{33}	U_{12}	U_{eq}[a]
Ce	3*a*	0.4224(1)	0	0	108(3)	91(3)	67(3)	46(2)	91(2)
Mg	3*g*	0.7572(6)	0	1/2	97(17)	110(20)	99(20)	55(10)	100(9)
Ga1	1*a*	0	0	0	119(7)	U_{11}	60(12)	60(4)	99(5)
Ga2	2*d*	1/3	2/3	1/2	101(5)	U_{11}	82(9)	51(3)	95(4)

[a] U_{eq} (pm^2) is defined as one-third of the trace of the orthogonalized U_{ij} tensor. $U_{13} = U_{23} = 0$.

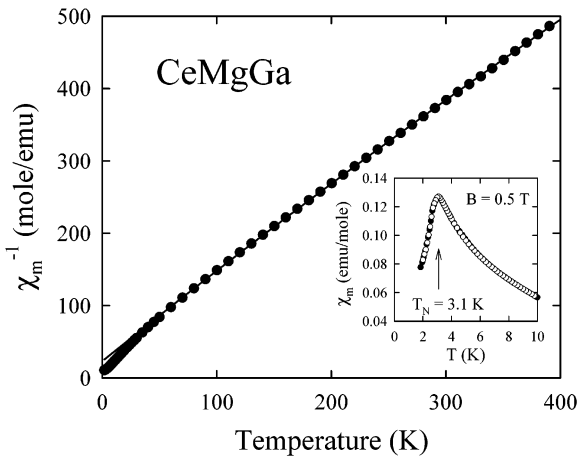

Figure 2. Temperature dependence of the reciprocal molar magnetic susceptibility of CeMgGa, measured in a magnetic field of 5 T. The solid line represents a modified Curie−Weiss fit with the parameters $\mu_{eff} = 2.49\ \mu_B$, $\theta = -18$ K, and $\chi_0 = 1.57 \times 10^{-4}$ emu/mol. Inset: the low−temperature susceptibility taken in a field of 0.5 T with increasing (open circles) and decreasing (full circles) temperature.

Table 3. Interatomic Distances (pm) Calculated with the Powder Lattice Parameters in the Structure of CeMgGa

Ce:	1	Ga1	317.9(1)	Ga2:	3	Mg	291.0(3)
	4	Ga2	319.9(1)		6	Ce	319.9(1)
	2	Mg	339.5(3)	Mg:	2	Ga2	291.0(3)
	4	Mg	357.9(1)		2	Ga1	291.7(3)
	4	Ce	389.7(1)		2	Mg	316.5(8)
Ga1:	6	Mg	291.7(3)		2	Ce	339.5(3)
	3	Ce	317.9(1)		4	Ce	357.9(1)

tances (342−353 pm) in Ce_5Mg_{42}.[80] Each cerium atom has four nearest cerium neighbors at a Ce−Ce distance of 390 pm, well above the Hill limit[81] of about 340 pm for *f* electron localization, in agreement with magnetic ordering discussed below.

Finally, we draw back to the Mg−Mg interactions. The magnesium atoms build the trigonal prisms around the origin of the unit cell. Within the triangular faces we observe Mg−Mg distances of 317 pm, slightly smaller than the average Mg−Mg distance of 320 pm in *hcp* magnesium.[82] We can thus assume a significant degree of Mg−Mg bonding in the CeMgGa structure.

Magnetic Properties. The temperature dependence of the inverse magnetic susceptibility of CeMgGa is displayed in Figure 2. Above 50 K, $\chi^{-1}(T)$ can be well described by the modified Curie−Weiss law (MCW)

$$\chi = \chi_0 + \frac{C}{T - \theta} \quad (C = (N_A \mu_{eff}^2 / 3k_B))$$

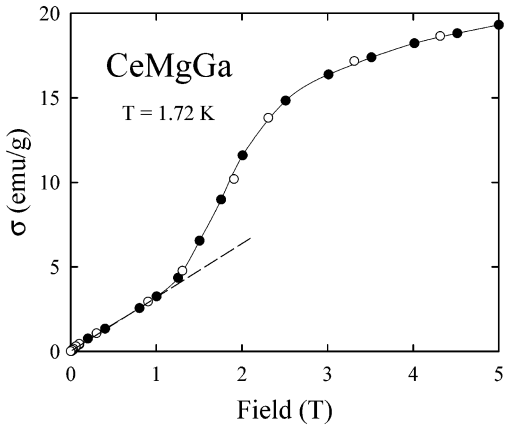

Figure 3. Field dependence of the magnetization of CeMgGa measured at $T = 1.72$ K with increasing (full circles) and decreasing (open circles) field. The dashed straight line emphasizes a metamagnetic behavior above 1 T.

with the effective magnetic moment $\mu_{eff} = 2.49(1)\ \mu_B$ (i.e., close to that expected for a free Ce^{3+} ion of $2.54\ \mu_B$), the paramagnetic Curie temperature $\theta = -18(2)$ K, and the temperature-independent term $\chi_0 = 1.57(2) \times 10^{-4}$ emu/mol. Apparent deviation of $\chi^{-1}(T)$ from the MCW behavior, seen at lower temperatures, presumably results from crystal field interactions.

The inset to Figure 2 shows the low-temperature susceptibility of CeMgGa. The pronounced maximum occurring at $T_N = 3.1(1)$ K unambiguously manifests a transition to a long-range antiferromagnetic state. The antiferromagnetic character of the spin arrangement below T_N is further corroborated by the field-dependent behavior of the magnetization taken at 1.72 K (Figure 3) with a distinct metamagnetic-like anomaly above 1 T. In high magnetic fields $\sigma(B)$ shows a tendency to saturation reaching at 5 T a value of 19.3(1) emu/g that corresponds to a magnetic moment of $0.81(1)\ \mu_B$. The latter is reduced when compared with the theoretical saturation magnetization for Ce^{3+} of $2.14\ \mu_B$, similar to CeAgMg[83] or $Ce_3Pd_4Sn_6$.[84]

Electrical Properties. The temperature dependence of the resistivity of CeMgGa is presented in Figure 4. The overall shape of $\rho(T)$ has a metallic character but the absolute magnitude of the resistivity is quite enhanced. Above ca. 7 K, $\rho(T)$ can be well described by the formula[85]

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

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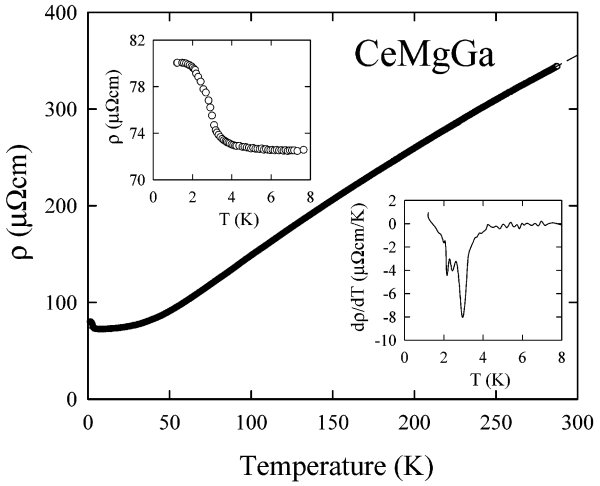

Figure 4. Temperature dependence of the electrical resistivity of CeMgGa. The dashed line represents the resistivity curve calculated with the Bloch−Grüneisen−Mott formula (see text). Upper inset: the low-temperature resistivity. Lower inset: temperature derivative of the resistivity in the vicinity of the antiferromagnetic phase transition.

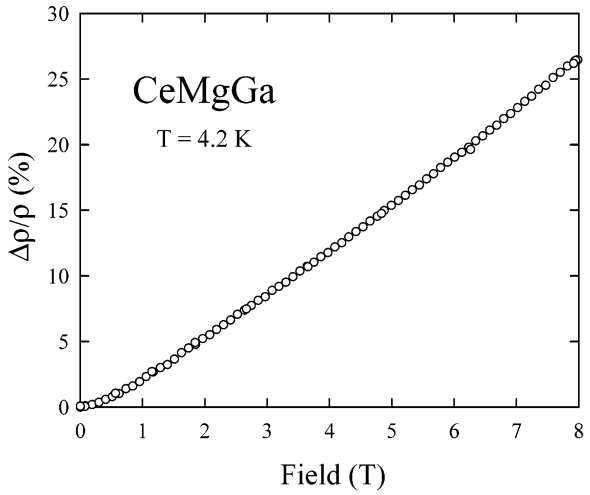

Figure 5. Field dependence of the transverse magnetoresistivity of CeMgGa taken at $T = 4.2$ K.

where the first term stands for the sum of temperature-independent residual (ρ_0) and spin disorder (ρ_0^{∞}) resistivities, the second term is a Bloch−Grüneisen-type resistivity due to scattering of conduction electrons on phonons, and the third term accounts for interband scattering processes. The least-squares fitting of this expression to the experimental data for CeMgGa yields the following parameters: $\rho_0 + \rho_0^{\infty} = 73.7$ $\mu\Omega$cm, $R = 1.04$ $\mu\Omega$cm/K, $\Theta_D = 246$ K, and $K = 6.76 \times 10^{-7}$ $\mu\Omega$cm/K^3.

Below the antiferromagnetic phase transition the resistivity slightly increases (see the upper inset to Figure 4). This behavior is reminiscent of systems exhibiting the opening of a gap on portions of the Fermi surface due to the onset of antiferromagnetism. The Neél temperature, defined as a deep minimum in the derivative dρ/dT vs T (see the lower inset to Figure 4), amounts to $T_N = 3.0(2)$ K, thus being in good agreement with the magnetic data.

Upon applying magnetic field, the resistivity of CeMgGa measured in the paramagnetic region shows a substantial increase. Figure 5 shows the field dependence of the transverse magnetoresistivity

$$\Delta\rho/\rho = \frac{\rho(B) - \rho(B = 0)}{\rho(B = 0)} \quad (B \perp i)$$

taken at $T = 4.2$ K. As seen, the magnetoresistivity is positive and large, arriving at 26% in a field of 8 T.

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