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Magnetic and galvanomagnetic properties of CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions in magnetic fields up to 13 T

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Abstract

The magnetoresistance and magnetization in magnetic fields up to 13 T have been investigated for the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions depending on composition. The intermediate valence state, the Kondo lattice state or the stable state of the Ce ions co-exist with the anti-ferromagnetic or ferromagnetic state of the Mn sublattice. For some of samples the Hall effect was measured. The substitution of Si → Ge leads to the increase of the magnetoresistance [R(B) − R(0)]/R(0). At 4.2 K and in a magnetic field of 13 T it changes its value from + 70% for $x = 0$ to $\leq |\pm 5|\%$ for $x \geq 0.3$. The behaviour of the galvanomagnetic effects is discussed. © 1997 Elsevier Science S.A.

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1. Introduction

The CeMn$_2$Si$_2$ ternary compound is a very interesting material, because in this compound the intermediate valence (IV) state of the Ce ions, with an effective valence of 3.13 [1,2], co-exists with an antiferromagnetic ordering of the Mn sublattice [2,3]. Due to the presence of different types of ions with localized magnetic moment and strong electron–electron interactions, it is interesting to investigate the behaviour of the electrical resistivity in high magnetic fields for various compositions based on the CeMn$_2$Si$_2$ compound.

It was found that for CeMn$_2$Si$_2$ [4] the magnetoresistance [R(B) − R(0)]/R(0) has an unusually large positive value (+70% at 4.2 K and in a magnetic field of 13 T). It is known that the substitution of Si → Ge in the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions leads to a change in the type of the Mn sublattice magnetic ordering from the collinear antiferromagnetic to the conical magnetic structure and of the valence state type of the Ce ions from the IV state to the Ce^{3+} stable state [5–7]. It was also shown that the magnetic phase diagram for the CeMn$_2$(Ge$_2$Si$_{1-x}$)$_2$ solid solutions in the region of compositions $0.5 < x < 0.7$ has a complex character [5,6], while in the region of compositions $0.1 \leq x \leq 0.5$ the Kondo lattice state is achieved for the Ce ions [6,8].

Here we present the results of the electrical resistivity and magnetization measurements for the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions in magnetic fields up to 13 T. For some of these samples the Hall effect was also measured.

2. Experimental details

Samples of the CeMn$_2$(Si$_x$Ge$_{1-x}$)$_2$ system were prepared by arc-melting in an argon atmosphere under a pressure of 10^{-5} Pa. The purity of the raw components was as follows: Ce — 99.00%; Mn — 99.80%; Si — 99.99%; Ge — 99.99%. The phase composition of the alloys was checked by the X-ray structural analysis. It was stated that all alloys crystallise in the structure of the CeGa$_2$Al$_2$ (ThCr$_2$Si$_2$) type [8]. Samples for measurements of galvanomagnetic properties were produced by grinding into a parallelepiped form with typical dimensions of 5 × 2 × 2 mm^3. The electrical contacts were made by fall out a copper film with further indium soldering. Mea-

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surements were carried out on the four-probe scheme with the dc method and applying an electric current *j*-value of 100 mA in the temperature range 4.2–120 K. Magnetic fields up to 13 T for the galvanomagnetic investigation were created by a superconducting solenoid of 'Intermagnetic' type with its orientation perpendicular to *j*. The measurements of a magnetization were made by the ballistic method using the Bitter magnet. The measurements were conducted at the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland).

3. Experimental results and discussion

The results of the measurements of the electrical resistivity vs. applied magnetic fields for the $CeMn_2(Ge_xSi_{1-x})_2$ samples with $x = 0$, 0.2, 0.5 and 0.9 are presented in Fig. 1. The electrical resistivity for these compositions, as well as for other samples of these solid solutions [6,8], changes over a wide range of values depending on composition. For this reason, the R(B)/R(0) and [R(B,T) − R(0,T)]/R(0,T) ratios were used in order to unify the magnetic field influence on the electrical resistivity. At 4.2 K the ratio R(B)/R(0) for the samples with $x = 0$, 0.2 and 0.5 has a positive sign and increases linearly with the increase of the intensity of the magnetic field. For $CeMn_2Si_2$ ($x = 0$) this dependence was presented for the first time by Levin [4]. The ratio R(B)/R(0) for the $x = 0.9$ sample has a small value and negative sign (−1.03).

The temperature dependence of the electrical resistivity for various values of the magnetic field for the $CeMn_2(Ge_xSi_{1-x})_2$ samples with $x = 0$, 0.2 and 1 are

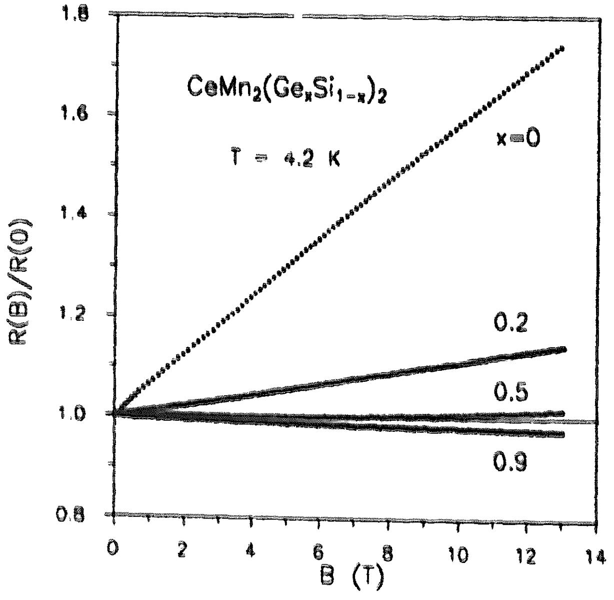

Fig. 1. The dependence of the electrical resistivity R(B) vs. a magnetic field, normalized to its zero-field value R(0), for samples of the $CeMn_2(Ge_xSi_{1-x})_2$ solid solutions.

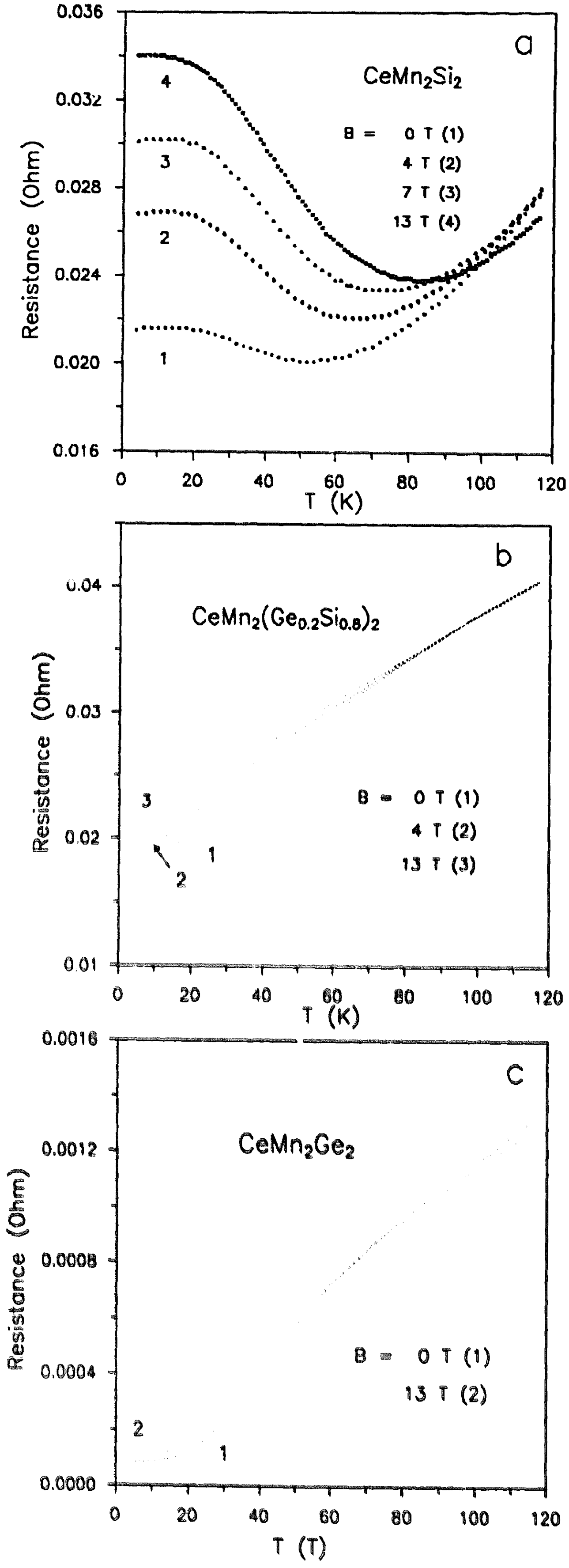

Fig. 2. The temperature dependence of the electrical resistance in various values of the magnetic field for samples: (a) $CeMn_2Si_2$; (b) $CeMn_2(Ge_{0.2}Si_{0.8})_2$; (c) $CeMn_2Ge_2$.

presented in Fig. 2a,b,c. On increasing the temperature, the magnetoresistance of all the samples decreases and reaches approximately zero above 100 K.

Our data for the temperature dependence of the electrical resistivity in zero magnetic field for samples of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions are in agreement with the data from [6]. We found that the character of the temperature dependence of the magnetoresistance for these samples depends on the composition. The substitution of Si → Ge leads to a decrease of the magnetoresistance and can even change its sign. At 4.2 K and in a magnetic field of 13 T the magnetoresistance changes from + 70% for $x = 0$ to $\leq |\pm 5|\%$ for $x \geq 0.3$.

The magnetic-field and temperature dependence of the Hall resistivity have been measured for samples with $x = 1$ and 0.2. The value of the Hall resistivity in these samples has an unusually large value and it is approximately more than two orders higher than that of a simple metallic compound or a heavy fermion compound of the CeRu$_2$Si$_2$ type [9]. With increasing temperatures, the Hall resistivity decreases and reaches the usual value above 100 K.

The magnetic-field dependence of the magnetization for the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ samples at 4.2 K are given in Fig. 3. The value of the magnetization at this temperature and in a magnetic field of 13 T changes from 3 cm^3 g^{-1} (0.03 μ_B F.U.$^{-1}$) for CeMn$_2$Si$_2$ to 30 cm^3 g^{-1} (2.2 μ_B F.U.$^{-1}$) for CeMn$_2$Ge$_2$. No metamagnetic phase transition at 4.2 K in magnetic fields up to 13 T for the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions with an antiferromagnetic ordering have been observed.

The behaviour of the magnetoresistance in a complex metallic system, in which the strong electron-electron interactions and the magnetic ordering coexist, is not clear at present. In simple metals the magnetoresistance usually has a positive sign due to the Lorentz force. According to Yamada and Takada [10,11], the magnetoresistance in the metallic systems with an electron-spin scattering has a negative sign for the paramagnetic and ferromagnetic cases, which comes from the suppression of fluctuation of the localized spins by the magnetic field. The magnetoresistance in an antiferromagnetic material has a positive sign. In a complex metallic system, such as CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$, the presence of all these cases are possible. Thus the behaviour of the magnetoresistance of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ system is a result of the competition of various types of scattering mechanism in the magnetic fields applied.

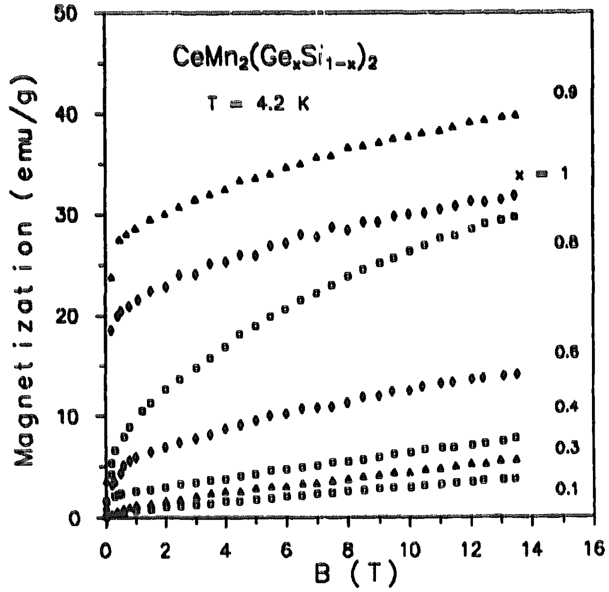

Fig. 3. The magnetic-field dependence of the magnetization for samples of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions.

In a metallic system the change of the electrical resistivity in an applied magnetic field is determined by both the change of the charge carrier mobility and concentration. The behaviour of the Hall effect in some investigated samples from the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions has shown that applied magnetic fields lead to the strong localization of charge carriers and to the appearance of a narrow energy gap.

It is known that in the Ce based heavy fermion systems the magnetoresistance has a negative sign. In the Kondo lattice compound CeCu$_2$Si$_2$ at 4.2 K, the magnetoresistance has a value of − 4% in a magnetic field of 13 T [12]. All samples from the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ solid solutions and the CeCu$_2$Si$_2$ compound have the same crystal structure as CeGa$_2$Al$_2$ (ThCr$_2$Si$_2$) type. Thus, much larger values of the magnetoresistance in some of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ samples is connected with the presence of the magnetic order of the Mn sublattice.

The RMn$_2$X$_2$ compounds with the CeGa$_2$Al$_2$ (ThCr$_2$Si$_2$) type structure have the layer structure of the R-(X-Mn-X)-R*-(X-Mn-X)-R type. One of the reasons for the large galvanomagnetic effects in a composition based on the CeMn$_2$Si$_2$ compound may be the polarization of the Ce ions by the Mn sublattice due to the Ce–Mn interlayer interactions. The possibility of polarization of the paramagnetic R sublattice by the Mn sublattice in RMn$_2$(Ge,Si)$_2$, due to the R–Mn interactions has been shown by Brabers et al. [13].

4. Conclusion

The investigation of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ system demonstrates large galvanomagnetic effects, a maximum value of which was found in the CeMn$_2$Si$_2$ compound ($x = 1$) at 4.2 K. Magnetization of the CeMn$_2$(Ge$_x$Si$_{1-x}$)$_2$ samples depends on composition and is connected to a type of magnetic ordering of the

Mn sublattice. However, the value of the magnetization and character of its magnetic-field dependence can not be the main reason for the large galvanomagnetic effects. These galvanomagnetic effects are a result of the coexistence of the valence instability of Ce ions and the anti-ferromagnetic ordering of the Mn sublattice, due to the strong electron–electron interactions and polarization of the Ce ions by the Mn sublattice.

Acknowledgements

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