

Complex antiferromagnetic order of CeCuSn

H. Nakotte, E. Brück, K. Prokes, J.H.V.J. Brabers and F.R. de Boer

Van der Waals-Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam (Netherlands)

L. Havela*

Department of Metal Physics, Charles University, Ke Karlovu 5, 12116 Prague 2 (Czech Republic)

K.H.J. Buschow

Philips Research Laboratories, 5600 AJ Eindhoven (Netherlands)

Yang Fu-ming

Institute of Physics, Academia Sinica, P.O. Box 603, Beijing (China)

Abstract

CeCuSn, which forms in the hexagonal CaIn_2 -type of structure, orders antiferromagnetically at 8.6 K. A second maximum at about 7.35 K seen in the temperature dependence of the specific heat indicates a rearrangement of the antiferromagnetic structure. The antiferromagnetic ordering is also reflected in the temperature dependence of the electrical resistivity. Upon application of magnetic fields up to 5 T, a complex development of magnetic phases can be followed by monitoring the temperature dependencies of the magnetization and the specific heat. In the B - T diagram, three different magnetic phases were detected and their boundaries could be determined.

1. Introduction

Within the framework of a systematic research program of CeTX compounds (T =transition metal and X =metal from the p-block of the Periodic Table), we have recently reported long-range magnetic order in compounds with T =Cu and X =Si, Ge and Sn [1]. Ferromagnetic order was detected for CeCuSi and CeCuGe, whereas for CeCuSn, a complex antiferromagnetic behaviour was indicated by the occurrence of two distinct maxima in the specific heat. In order to gain more insight on the nature of these two transitions in CeCuSn, we have performed more elaborate studies on the field dependence of the specific heat and the temperature dependencies of the electrical resistivity and the magnetization.

2. Experimental details

A polycrystal of CeCuSn was prepared by arc-melting stoichiometric amounts of the pure elements in a Ti-gettered argon atmosphere. The button obtained was wrapped into Ta foil and sealed in quartz tubes under

300 mbar of argon. The button was annealed for 10 days at 800 °C. All lines of the X-ray powder pattern could be indexed in the proper CaIn_2 structure [2]. However, our X-ray results did not conclude whether the Cu and Sn atoms are randomly distributed over the In positions in this structure, or whether an ordered variant of the CaIn_2 structure is formed. Although the annealing temperature and time differ slightly from those used in ref. 1, the lattice parameters of the two samples were found to be in good agreement. The homogeneity of the sample was checked by microprobe analysis. For the matrix, a homogeneous distribution with the proper 1:1:1 composition was found. Nevertheless, small amounts of impurity phases (in total less than 2%) were detected. Besides Ce_2O_3 , which is the major impurity phase, very small amounts of precipitates with compositions 2:1:1 and 2:3:3 were found.

We performed measurements of: the electrical resistivity between 4.2 and 300 K using the standard ac four-point probe method; specific heat in the temperature range between 1.3 and 40 K in various applied magnetic fields up to 5 T using the standard semi-adiabatic method; temperature (between 1.5 and 20 K) and field (between 0.05 and 6 T) dependencies of the magnetization by means of a SQUID and a pendulum magnetometer.

*Author to whom correspondence should be addressed.

3. Results and discussion

The temperature dependence of the electrical resistivity is shown in Fig. 1. Between 270 and 150 K, an almost linear decrease of the electrical resistivity with decreasing temperature is found, followed by a development towards the T^5 dependence at somewhat lower temperatures, which emphasizes the main scattering mechanism to be the electron phonon scattering. At low temperatures, an additional scattering mechanism, which is connected with the evolution of magnetism, is seen. As can be seen in the inset in Fig. 1, a distinct drop in the resistivity is observed at about 7.8 K, which is intermediate between the two maxima found in the zero-field specific heat [1]. Most likely, the occurrence of only one distinct anomaly and the slight difference in the ordering temperature originate from the texture in the sample. Measurements on single crystalline compounds crystallizing in anisotropic crystal structure reveal strongly anisotropic behaviour for different current directions (see *e.g.* [3]).

The field dependence of the specific heat in the temperature range between 1.3 and 15 K is shown in Fig. 2. In 0 T, the specific heat curve exhibits four anomalies at 2.7, 5.6, 7.35 and 8.6 K. These features were also observed in the sample used in ref. 1. However, the hump at 2.7 K and the shoulder seen at 5.8 K are weaker compared to the values in ref. 1. Therefore, these anomalies have to be ascribed to different amounts of impurity phases present in both samples. The latter anomaly is most likely due to Ce_2O_3 , which orders

antiferromagnetically at about 6 K [4]. Only the two maxima at 7.35 and 8.6 K are thought to be intrinsic for CeCuSn. In ref. 1, the field dependencies of the specific heat, which were performed in magnetic fields of 1, 2, 3, 4 and 5 T, led to the conclusion that the maximum at 8.6 K is connected with antiferromagnetic interactions (shifting to lower temperatures upon application of a magnetic field), whereas the maximum at 7.35 K reflects ferromagnetic interactions (shift to higher temperatures). Assuming a crossover of the opposite shifts at 1 T, this did explain the sharp single-line feature in 1 T and the subsequent doubling for 2 T. However, the present results obtained in intermediate fields (Fig. 2) yield a different picture. While the maximum at 7.35 K is suppressed in rather low fields of about 1 T, the maximum at 8.6 K splits into two peaks for higher fields. The latter two peaks both shift to lower temperatures with further increase in the field. Therefore, the two maxima at 2 T both originate from the 8.6 K maximum at 0 T.

The temperature dependence of the magnetization in various magnetic fields is shown in Fig. 3. Clearly, CeCuSn shows qualitatively different behaviour for different field ranges. For magnetic fields above 4.5 T, no distinct change in the temperature dependence of the magnetization can be seen. Between 3.0 and 4.5 T, a distinct change in the slope indicates the phase boundary between the paramagnetic and the magnetically ordered region. Between 1.25 and 2.5 T, some kind of maximum develops. The inflection points (indicated by the arrows in Fig. 3 for the 2.5 T result) indicate two magnetic

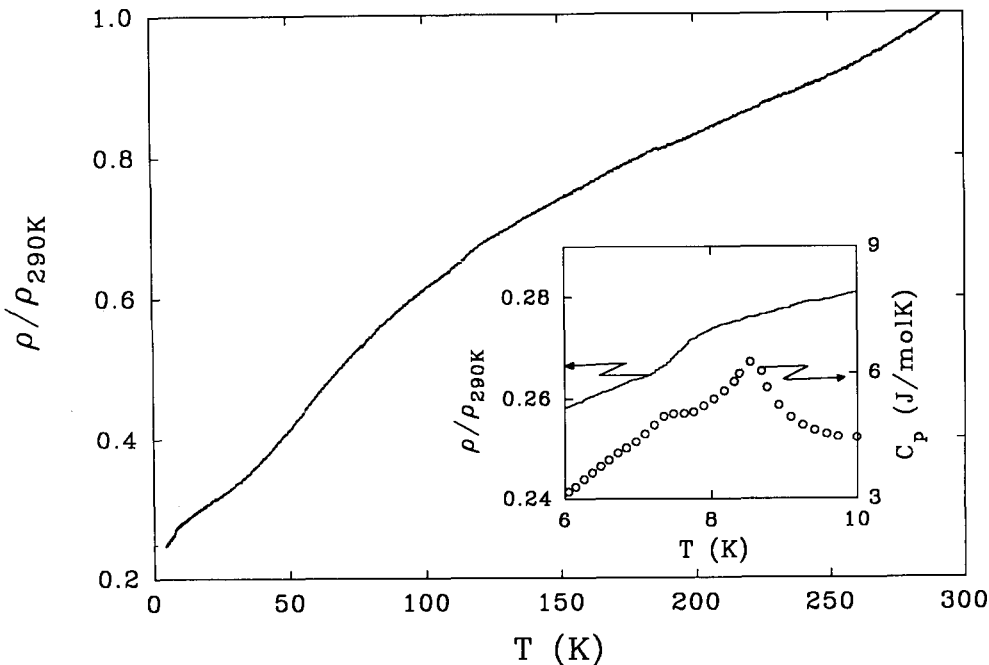


Fig. 1. Temperature dependence of the electrical resistivity of CeCuSn normalized to the resistivity value at 290 K. The inset shows the low temperature part together with the zero-field specific heat.

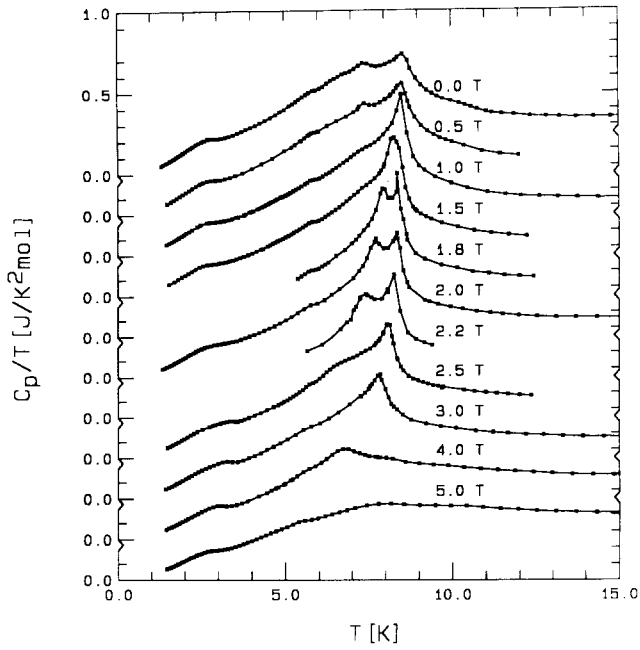


Fig. 2. Temperature dependence of the specific heat of CeCuSn in various applied magnetic fields in the representation C_p/T versus T .

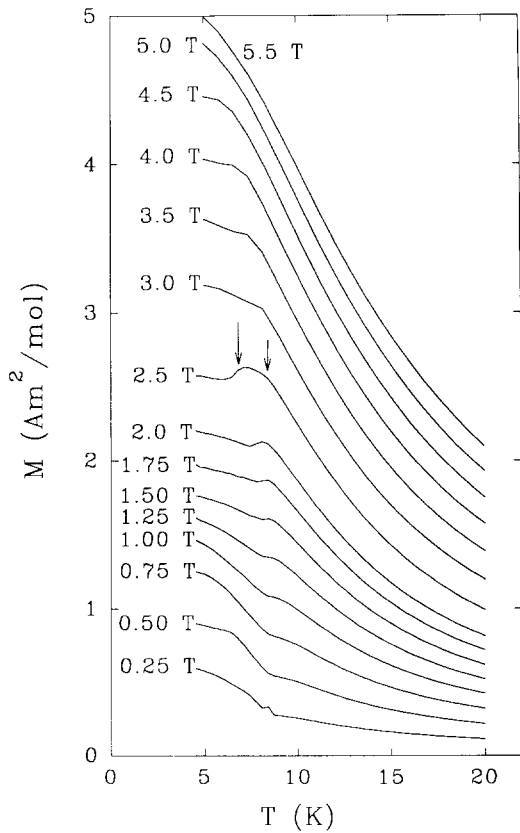


Fig. 3. Temperature dependence of the magnetization of CeCuSn in various magnetic fields. The arrows correspond to the maximum values of dM/dT for the 2.5 T result.

transitions for this field range. Below 1.25 T, there is no distinct maximum, but some kind of plateau followed by an increase at lower temperatures. In all cases, the phase boundaries can be derived from the derivatives/inflection points of the magnetization curves.

Below 7.35 K, the magnetization as a function of applied magnetic field exhibits a very weak metamagnetic transition, which reflects the destruction of the zero-field phase. The result at 1.5 K is shown in Fig. 4. A very small hysteresis is seen in the magnetization curve. For low temperatures, the strong curvature in very low fields, which originates from the saturation of the Ce^{3+} impurities, indicates a complete compensation of the moments in the zero-field structure of CeCuSn. Between 7.35 and 8.6 K, no metamagnetic transition could be resolved.

Combining the anomalies of the specific heat and magnetization results (Figs. 2–4), we can draw a tentative magnetic phase diagram of CeCuSn. As seen in Fig.

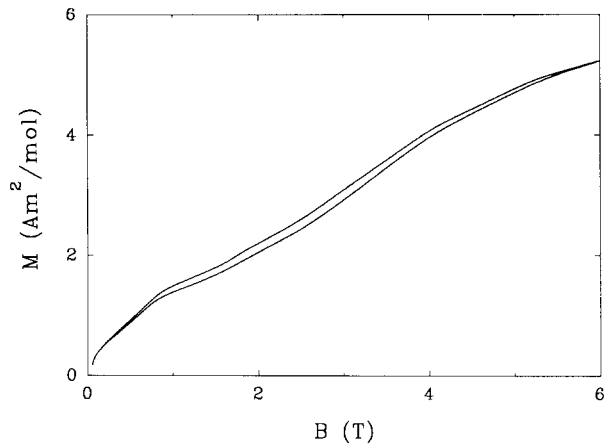


Fig. 4. Field dependence of the magnetization of CeCuSn at 1.5 K. Note the weak metamagnetic transition around 2.5 T.

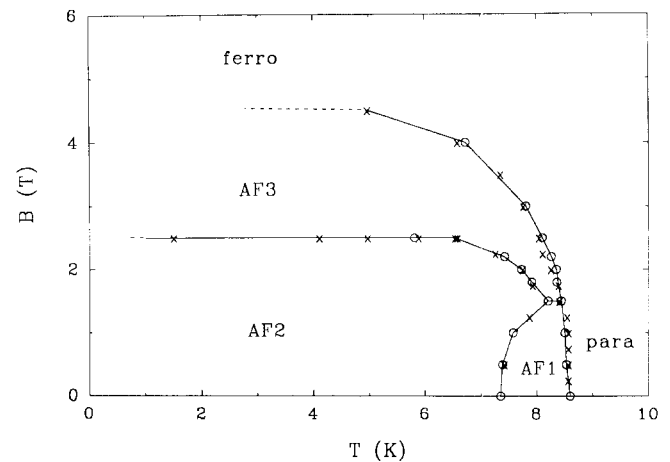


Fig. 5. Magnetic phase diagram of CeCuSn. The circles denote the values obtained from the specific heat results, whereas the crosses denote the values obtained from magnetization measurements.

5, the results imply three different magnetic phases. Below 8.6 K, CeCuSn orders antiferromagnetically (AF1). Below 7.35 K, this structure is rearranged to phase AF2. At low temperatures, application of a magnetic field most probably yields a step-wise transformation from the antiferromagnetic phase AF2 to phase AF3 at 2.5 T and then to a forced ferromagnetic aligned phase for fields above 4.5 T. The phase AF3 is most likely a phase with strong antiferromagnetic interactions, but incomplete compensation. The phase AF1 exists only in a rather small temperature interval between 7.35 and 8.6 K for magnetic fields below 1.5 T. However, the boundaries in this phase diagram should be taken with caution as they were obtained on a polycrystal. It is well known that, for anisotropic materials, the phase boundaries are very sensitive to the direction of the magnetic field applied (see e.g. CePtSn [3]).

4. Conclusions

Our studies of the field and temperature dependencies of the specific heat, the electrical resistivity and magnetization of CeCuSn yield a tentative magnetic phase diagram with three different magnetic phases. Whereas the phase AF3 is only stable in an applied magnetic field, two magnetic phases AF1 and AF2 are formed in zero field, similar to the magnetic phase diagram for the compound CePtSn [3]. However, the latter compound forms in the orthorhombic TiNiSi-type of structure, which complicates a direct comparison. More insight can be gained by comparing the results with the findings for UTSn (T = Pd, Au, Cu) compounds, which also form in the CaIn₂-type of structure [5]. It is interesting to note that UPdSn and UCuSn form in the ordered variant of this structure (GaGeLi-type) with the Pd/Cu and the Sn atoms ordered, whereas in UAuSn the Au and the Sn atoms are randomly distributed [5]. In ref. 7, the crystallographic differences of these compounds were discussed in terms of the relative differences in atomic radius. Of course, these arguments should be valid also for CeCuSn. Therefore, we may expect CeCuSn also to form in an ordered variant of the CaIn₂-type of structure. Comparing now with the bulk properties of the UTSn compounds, we note that UAuSn has one single magnetic transition, whereas UPdSn exhibits two transitions [5]. The situation for UCuSn is not yet solved, although two distinct anomalies were observed in the electrical resistivity [6]. As to the magnetic phase diagram, we find a quite similar phase diagram for UPdSn, where a third magnetic phase also occurs upon application of a magnetic field [8]. Although the critical temperatures and fields

for UPdSn are much higher, we may speculate that the magnetic structures in CeCuSn and UPdSn are similar. This is partly supported by the in-plane type of anisotropy found in CeCuSn [1], which is also found in UPdSn [5]. Nevertheless, microscopic tools (e.g. neutron diffraction, Mössbauer spectroscopy) have to be applied in order to establish the magnetic structures in CeCuSn. These experiments will be performed in the near future.

Acknowledgments

The present investigation is part of the scientific exchange program between China and The Netherlands. The work has been supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse organisatie voor Wetenschappelijk Onderzoek (NWO). Part of the work has been supported by the Magnetism Laboratory, Institute of Physics, Academia Sinica, Beijing.

References

- 1 Fuming Yang, J.P. Kuang, Jingyuan Li, E. Brück, H. Nakotte, F.R. de Boer, Xiucheng Wu, Zhenxiao Li and Yupeng Wang, *J. Appl. Phys.*, **69** (1991) 4705.
- 2 A.E. Dwight, in C.E. Lundin (ed.), *Proc. 12th Rare Earth Research Conf.*, Denver Research Institute, Denver, CO, 1976, p. 480.
- 3 T. Takabatake, H. Iwasaki, G. Nakamoto, H. Fujii, H. Nakotte, F.R. de Boer and V. Sechovsky, *Physica B*, **183** (1993) 108.
- 4 M.J. Besnus, J.P. Kappler and A. Meyer, *J. Phys. F: Metal Phys.*, **13** (1983) 597.
- 5 F.R. de Boer, E. Brück, H. Nakotte, A.V. Andreev, V. Sechovsky, L. Havela, P. Nozar, C.J.M. Denissen, K.H.J. Buschow, B. Vaziri, M. Meissner, H. Maletta and P. Rogl, *Physica B*, **176** (1992) 275.
- 6 H. Fujii, H. Kawanaka, T. Takabatake, E. Sugiura, K. Sugiyama and M. Date, *J. Magn. Magn. Mater.*, **87** (1990) 235.
- 7 R.A. Robinson, J.W. Lynn, V. Nunez, K.H.J. Buschow, H. Nakotte and A.C. Lawson, *Phys. Rev. B*, **47** (1993) 5090.
- 8 H. Nakotte, E. Brück, F.R. de Boer, P. Svoboda, N.C. Tuan, L. Havela, V. Sechovsky and R.A. Robinson, *J. Appl. Phys.*, **73** (1993) 6551.
- 9 J. Sakurai, K. Kegai, Y. Ishikawa and K. Mori, *Physica B*, **186-188** (1993) 583.

Note added in proof

In the course of this study we became aware of the results of Sakurai *et al.* [9]. They prepared two samples of CeCuSn by means of different methods. A strong dependency of magnetic properties on the preparation method was concluded. Nevertheless, the results on one of these samples is perfectly consistent with our data.