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# Magnetic properties of CeCuX compounds

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The temperature dependence of the magnetic susceptibility, the magnetization at 4.2 K in fields up to 35 T, and the specific heat between 1.3 and 40 K in fields up to 5 T of the ternary Ce intermetallic compounds CeCuX (X = Si, Ge, Sn) have been studied. All three compounds form in ordered ternary structures related to the hexagonal $AlB_2$ type. Ferromagnetic order is found below 14.9 K for CeCuSi and below 10.2 K for CeCuGe. The magnetic order of CeCuSn below 8.6 K is of antiferromagnetic type but probably complex, as indicated by a double peak in the specific heat and the occurrence of a very low zero-field moment.

## I. INTRODUCTION

Within the framework of a research program on ternary Ce intermetallics of the type CeTX with T a transition metal and X an element from the *p*-block in the periodic table, we have prepared compounds with $T$ = Cu and $X$ = Si, Ge and Sn, all belonging to the tetravalent metals of group IVB of the periodic table.

All three compounds, CeCuSi, CeCuGe amd CeCuSn, crystallize in structures derived from the hexagonal $AlB_2$ type of structure with Ce on the Al positions. CeCuSi has been reported to form at high temperatures in the $AlB_2$ structure,[1] but upon annealing at 750 °C an ordered ternary phase with doubled $c$ axis has been reported to form.[2] Also CeCuGe has been reported to crystallize in the $AlB_2$ structure.[3] In the case of CeCuSn, the Cu and Sn atoms form a zigzag arrangement, leading to the $CaIn_2$ type of structure[4].

## II. EXPERIMENT

Polycrystalline samples were prepared by arc-melting stoichiometric amounts of the pure metals in a Ti-gettered argon atmosphere. The obtained buttons were wrapped into Ta foil and sealed in quartz tubes under 300 mbar argon. CeCuSi and CeCuGe were annealed for 1 week at 800 °C and CeCuSn for 2 weeks at 750 °C. The quality of the samples was checked by x-ray powder diffraction. All compounds were found to have crystallized in the hexagonal structure with doubled $c$ axis indicating an ordered ternary structure. The lattice parameters are for CeCuSi $a = 4.238$ Å and $c = 7.988$ Å, for CeCuGe $a = 4.311$ Å and $c = 7.933$ Å, and for CeCuSn $a = 4.583$ Å and $c = 7.865$ Å. According to the x-ray pattern, the compounds CeCuGe and CeCuSn were single phase, whereas CeCuSi was found to contain a second $AlB_2$-type phase with slightly smaller lattice parameters. The composition of this phase was determined by electron microprobe to correspond to $CeCu_{0.8}Si_{1.2}$.

## III. RESULTS AND DISCUSSION

The temperature dependence of the inverse susceptibility is displayed in Fig. 1. Above 100 K, for all three compounds Curie–Weiss behavior is observed with effective moments of about 2.5 $\mu_B$, almost the full Ce free-ion value, and with negative paramagnetic Curie temperatures of about − 20 K. At 15 K the susceptibility of CeCuSi diverges, which is also the case for CeCuGe at 10 K. In CeCuSn, below 10 K, slightly above the magnetic ordering temperature of 8.6 K as observed in the specific-heat measurement (see below), the magnetization curves no longer pass through the origin. The value of $M(0)$, obtained by extrapolating the magnetic isotherms which are almost linear in the field range between 0.1 and 1 T to zero field, increases when the temperature is lowered reaching a value of 0.07 $\mu_B$ at 1.6 K. At present, it is not clear whether the existence of $M(0)$ is an intrinsic property of CeCuSn or whether this small contribution is due to an impurity phase. The observed differential susceptibility below 10 K is approximately temperature independent.

In Fig. 2 the magnetization at 4.2 K in fields up to 35 T is shown, measured on free-powder particles free to orient themselves in the applied field, simulating a single crys-

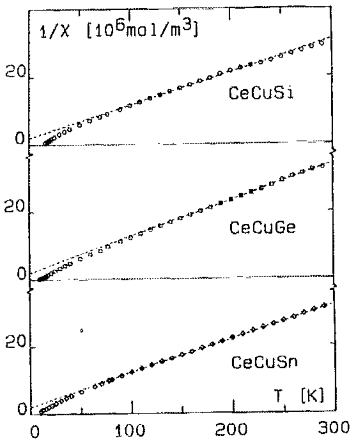

FIG. 1. Temperature dependence of the inverse magnetic susceptibility of CeCuSi, CeCuGe, and CeCuSn. The lines are guides to the eye.

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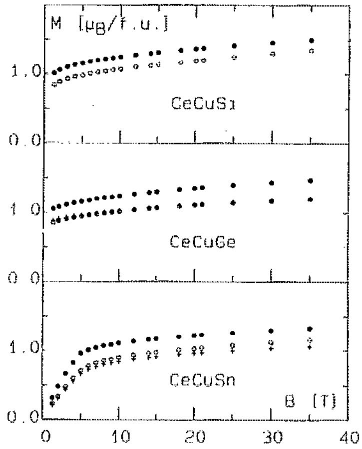

FIG. 2. Magnetization vs applied magnetic field at 4.2 K for CeCuSi, CeCuGe, and CeCuSn. The measurements have been carried out on powder free to be oriented by the applied magnetic field (●), randomly oriented powder fixed by frozen alcohol (○), and bulk pieces (+).

tal along the easy axis, and on powder particles fixed in alcohol, representing an ideal polycrystal. For comparison, for CeCuGe and CeCuSn also the results of magnetization measurements on samples consisting of bulk pieces have been included. The results are very similar to those obtained on the fixed-powder particles, showing preferential orientation of the grains in the samples to be relatively unimportant. The difference of about 15% between the free- and fixed-powder magnetizations, which is found for all three compounds and which is still present in fields as high as 35 T, shows that the magnetocrystalline anisotropy in these compounds is considerable. The value of 15% may be indicative for an easy magnetization direction within the basal plane. Extrapolation of the magnetization from high fields to zero field gives moments of about 1.2 $\mu_B$, indicating that we are dealing with a mixture of crystal-field doublets.

Figure 3 shows the temperature dependence of $C_p/T$ of CeCuSi between 1.3 and 30 K at 0, 1, 3, and 5 T. At 0 T a sharp peak is observed at 14.9 K, which is broadened and shifted to higher temperatures by an applied magnetic field, as expected for a ferromagnetic material. The shoulder around 5 K is probably due to some impurity. The entropy under the peak after subtraction of an estimated phonon contribution equals about $R \ln 2$.

In Fig. 4 $C_p/T$ vs $T$ is shown for CeCuGe at 0, 1, 3, and 5 T. Similar to CeCuSi, the zero-field peak at 10.2 K is smeared out and shifted to higher temperatures upon applying an external field. The anomalies at about 6 K are due to Ce oxide. Again, for the entropy a value of about $R \ln 2$ is found.

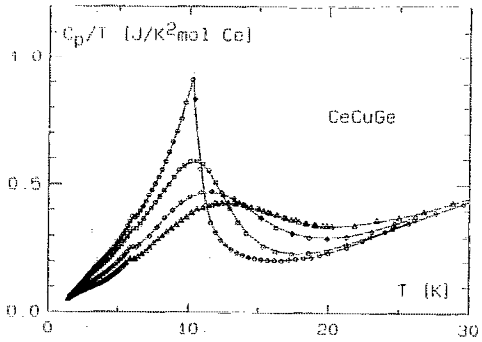

FIG. 4. $C_p/T$ vs $T$ for CeCuGe in applied magnetic fields of 0 T (○), 1 T (□), 3 T (◇) and 5 T (△). Lines are guides to the eye.

The temperature and field dependence of $C_p/T$ of CeCuSn are displayed in Figs. 5 and 6, and are much more complex than for the other two compounds. The broad double peak appearing in zero field below 8.6 K becomes a sharp feature in an applied field of 1 T. In an applied field of 2 T the peak becomes double again, becoming slightly more pronounced and shifted to lower temperatures, at 3 T it is again a single peak and at 5 T only a broad shoulder is left. The feature at about 2.5 K can not be ascribed to Ce oxide. It is not yet clear whether it is an intrinsic feature of CeCuSn. The magnetic entropy under the peak is estimated to be somewhat less than $R \ln 2$. The general tendency of $C_p/T$ to shift to lower temperature in an applied field is consistent with an antiferromagnetic type of ordering. The very complex structure of the specific heat will be subject of further study.

## IV. CONCLUSIONS

From the temperature dependence of the magnetic susceptibility and the field dependence of the specific heat it is concluded that CeCuSi and CeCuGe order ferromagneti-

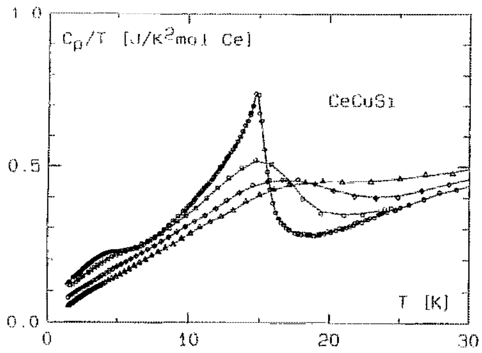

FIG. 3. $C_p/T$ vs $T$ for CeCuSi in applied magnetic fields of 0 T (○), 1 T (□), 3 T (◇), and 5 T (△). Lines are guides to the eye.

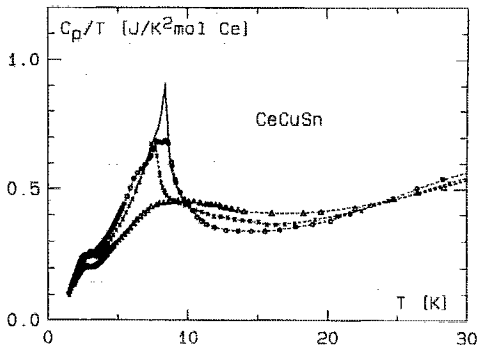

FIG. 5. $C_p/T$ vs $T$ for CeCuSn in applied magnetic fields of 0 T (○), 3 T (×), and 5 T (△). The 1-T result is represented by the solid line. The dashed lines are guides to the eye.

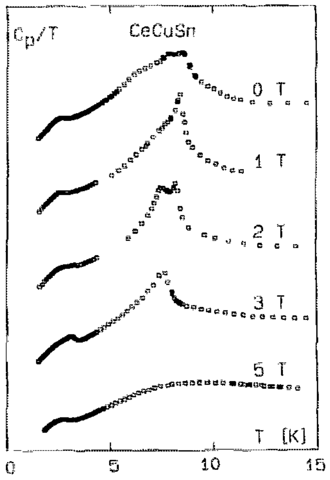

FIG. 6. $C_p/T$ vs $T$ in arbitrary units for CeCuSn in applied magnetic fields of 0, 1, 2, 3, and 5 T.

cally at 14.9 and 10.3 K, respectively. The anisotropy, probably of easy-plane type, persists in fields as high as 35 T. At this moment, it is difficult to decide about the magnetic ground state of CeCuSn. The results indicate a complex antiferromagnetic structure, which needs further investigation by microscopic methods.

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