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### Angaben zur Veröffentlichung / Publication details:

Oeschler, Niels, Micha Deppe, Philipp Gegenwart, Christoph Geibel, and Frank Steglich.  
2006. "Thermal expansion of  $\text{CeCu}_2(\text{Si}_{1-x}\text{Gex})_2$ ." *AIP Conference Proceedings* 850: 1165–61.  
<https://doi.org/10.1063/1.2355118>.

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# Thermal Expansion of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$

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**Abstract.** We report low-temperature thermal expansion measurements on  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  single crystals with  $0.01 \leq x \leq 0.45$ . The results evidence a new magnetic phase diagram more complex than previously obtained. In addition to the antiferromagnetic transition  $T_N(x)$ , a first order transition  $T_1(x)$  within the magnetic state is observed in the whole doping range studied. For  $x = 0.25$ , a tetracritical point is found at which  $T_N(x)$  and  $T_1(x)$  merge together. Furthermore, the analysis of the Grüneisen parameter suggests that at this concentration a transition from rather localized  $f$  electrons for  $x > 0.25$  to composite heavy fermions for  $x < 0.25$  occurs. This strongly supports the itinerant spin-density-wave scenario for the quantum-critical point observed in  $\text{CeCu}_2\text{Si}_2$ .

**Keywords:** heavy fermion, quantum critical point,  $\text{CeCu}_2\text{Si}_2$

**PACS:** 71.27.+a

The study of quantum critical phenomena is one of the most fascinating topics in strongly correlated electron physics. Especially, heavy fermion (HF) systems have turned out to be an appropriate class of systems to study this unconventional state. HF systems that are located close to a magnetic instability show pronounced deviations from Fermi-liquid behavior.

HF systems can be driven towards the quantum critical point (QCP) at which the magnetic order temperature vanishes continuously by applying hydrostatic pressure or by chemical substitution. Many compounds as  $\text{CePd}_2\text{Si}_2$  and  $\text{CeIn}_3$  have been found to behave as described. Currently, two scenarios are discussed which describe the properties of a system at the QCP. In the spin-density-wave scenario, the composite heavy fermions formed by the  $f$  electrons and the conduction electrons exist throughout the QCP. They are stable not only on the paramagnetic side, but also on the magnetically ordered side. A spin-density-wave type of antiferromagnetic order occurs through an instability of the Fermi surface of the heavy quasiparticles. Thus, the HF state is expected to evolve within the magnetic phase by approaching the QCP. In the locally QCP the composite fermions only exist on the paramagnetic side. At the QCP, they break up into rather localized  $f$  electrons and light conduction electrons. The magnetic order is formed by localized moments.

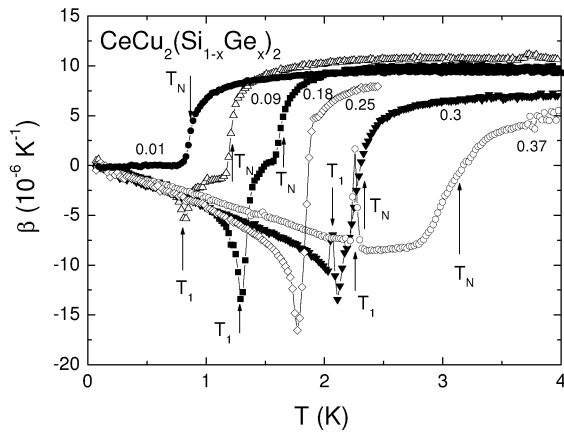
In order to distinguish between both scenarios a study of the development of composite heavy quasiparticles within the magnetic phase is an adequate approach. The absolute value of the Grüneisen parameter being related to the hybridization strength gives insight to the HF state. Determining the Grüneisen parameter of a system located at a QCP in dependence of a tuning parameter allows an unambiguous determination of the nature of

the QCP.

The first discovered HF superconductor at ambient pressure is  $\text{CeCu}_2\text{Si}_2$  [1]. The superconducting phase was found to exist within the magnetic phase called “A phase” for stoichiometric  $\text{CeCu}_2\text{Si}_2$ . The magnetic phase was suggested to be a spin-density wave type of antiferromagnetic order [2]. By substituting Si by the isoelectronic Ge the magnetic ordering is stabilized since the hybridization between the  $f$  electrons and the conduction electrons is decreased. A complex phase diagram has been developed based on results on polycrystalline  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  samples [3].

Recently, large, high-quality  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  single crystals became available [4] to study the magnetic phase and the development of the HF state. Neutron diffraction measurements [5], specific heat [4] and thermal expansion [6] were performed on samples of the same batch. Here, we present thermal expansion results on the  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  single crystals with  $0.01 \leq x \leq 0.45$ . The crystals were grown by a modified Bridgman technique using Cu flux. The thermal expansion measurements were performed utilizing an ultrahigh resolution capacitive dilatometer.

In Fig. 1 the volume thermal expansion  $\beta$  of a selection of the  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  samples studied is shown. The thermal expansion results show clear anomalies at the antiferromagnetic ordering transition  $T_N$  as well as at the first order transition within the magnetic phase  $T_1$  that was previously not detected for all concentrations. A tetracritical point was observed for  $x = 0.25$  at which  $T_N$  and  $T_1$  merge. In  $B = 0$ , both transitions temperatures seem to be degenerated. Only by applying a magnetic field the first order transition is shifted stronger towards lower temperatures than the antiferromagnetic order temperature. A more detailed analysis of the phase



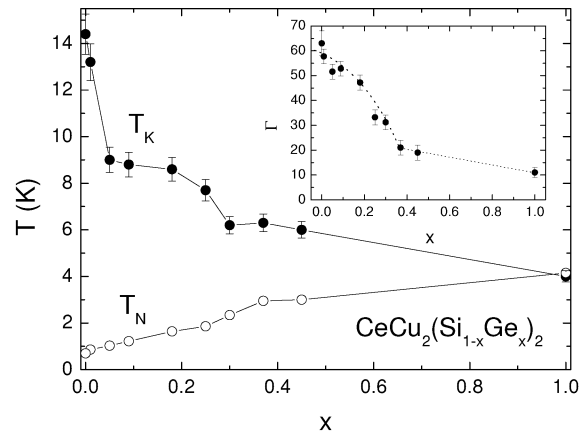
**FIGURE 1.** Volume thermal expansion coefficient  $\beta$  of  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  vs.  $T$ .

diagram is given in [6].

In the inset of Fig. 2 the Grüneisen parameter  $\Gamma$  vs.  $x$  is plotted. The Grüneisen parameter is calculated according to  $\Gamma = V_{\text{mol}}/\kappa_T \cdot \beta/C$ , where  $V_{\text{mol}}$  depicts the molar volume and  $\kappa_T$  the isothermal compressibility. The specific heat data of samples of the same batch were taken from [4]. The Grüneisen parameter is obtained within the paramagnetic state for a temperature of  $T = 1.1 \cdot T_N$ . The physical meaning of the Grüneisen parameter is given by the following expression:  $\Gamma \propto -\ln T^*/\ln V$ , the volume dependence of the characteristic temperature. For a Kondo system the characteristic temperature  $T^*$  is linked to the hybridization strength. Therefore, the Grüneisen parameter is also a measure of the hybridization strength. The Grüneisen parameter of HF systems is known to be orders of magnitude higher than in ordinary metals.

For  $\text{CeCu}_2\text{Ge}_2$  the Grüneisen analysis yields a value of around 10 which is slightly enhanced indicating a weak hybridization of the  $f$  electrons with the conduction electrons. Upon decreasing the Ge content in  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  the magnetic phase becomes weaker and the hybridization grows. A small increase of  $\Gamma$  is found between  $x = 1$  and  $x = 0.37$ . Around  $x = 0.25$  an abrupt rise in  $\Gamma$  is found and for lower Ge concentration  $\Gamma$  levels in at a high value of around 50 marking a fully developed HF state.

The same concentration dependence is observed for the Sommerfeld coefficient which is also related to the hybridization strength [4]. Also the Kondo temperature calculated by the magnetic entropy shows a similar behavior, cf. Fig. 2. For pure  $\text{CeCu}_2\text{Ge}_2$  the Kondo temperature is in the same order as the magnetic ordering temperature. Only a slight change is observed by lowering the Ge concentration. Around  $x = 0.25$ ,  $T_K$  increases strongly. In the same concentration range a



**FIGURE 2.** Kondo temperature  $T_K$  and Néel temperature  $T_N$  of  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  vs.  $x$ . Inset: Grüneisen parameter  $\Gamma$  of  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  vs.  $x$ .

slight decrease of  $T_N$  is observed.

All these findings suggest a abrupt increase of the hybridization strength around  $x = 0.25$ . For  $x > 0.25$ , the  $f$  electrons exhibit rather localized character whereas for  $x < 0.25$  they form composite heavy fermions. It might be coincidence that the tetracritical point is detected for the same concentration at which the drastic change of the hybridization strength is found.

Quantum critical behaviour is observed for pure  $\text{CeCu}_2\text{Si}_2$ . Neutron diffraction experiments on  $\text{CeCu}_2\text{Si}_2$  reveal an itinerant spin-density-wave type of magnetic ordering [5]. This is in agreement with previous measurements of the resistivity [7] and magnetization [2]. The presented study of the thermal expansion and the Grüneisen parameter supports the existence of the itinerant QCP and excludes the locally QCP. For the case of the locally QCP one would expect the formation of the composite heavy fermion state only around the QCP. The Grüneisen study, by contrast, suggests that the heavy electrons form already far away from the QCP at the magnetic side of the QCP.

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