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Magnetic phase diagram of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ measured with low-temperature thermal expansion

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We investigated the evolution of the magnetism in the alloy $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ by means of low-temperature thermal expansion measurements on large single crystals with $0.01 \leq x \leq 0.45$. The results evidence a new magnetic phase diagram more complex than that obtained in previous studies on polycrystals. The two main features are a second order transition from a paramagnetic to an antiferromagnetic state with a transition temperature $T_N(x)$ continuously increasing with Ge content and a first order transition corresponding to some change in the magnetically ordered structure at $T_1(x) \leq T_N(x)$. $T_1(x)$ and $T_N(x)$ seem to merge at $x \approx 0.25$ leading to a tetracritical point at this concentration. An analysis of the Grüneisen parameter suggests that at this critical 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 scenario for the quantum-critical point observed in pure CeCu_2Si_2 .

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I. INTRODUCTION

The discovery¹ of superconductivity in the heavy fermion (HF) system CeCu_2Si_2 initiated the field of heavy fermion superconductivity. The large Sommerfeld coefficient ($\gamma \approx 0.8 \text{ J/mol K}^2$) and the pronounced jump in the specific heat at the superconducting (SC) transition temperature T_c ($\Delta C/T_c \approx 1 \text{ J/mol K}^2$) led to the conclusion that the SC Cooper pairs are formed by the heavy quasiparticles whose large effective masses indicate a strong involvement of the magnetic $4f$ electrons.¹ Although the discovery was made 25 years ago, the nature of this SC state and of the SC pairing mechanism is still not clarified. Ten years after the discovery of the SC state, several groups found that a further ground state, the so-called A phase, competes with the SC state.^{2,3} Although the first studies gave evidence for a magnetic character of this phase, its true nature remained unclear. Investigations on CeCu_2Si_2 (Ref. 4) and CeCu_2Ge_2 under pressure (Ref. 5) as well as of the alloy system^{6,7} $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ showed that CeCu_2Si_2 is located very close to a quantum-critical point (QCP), at which the transition temperature of the A phase vanishes. Since in the past years many other examples of Ce-based systems have been found, in which the disappearance of the magnetic phase coincides with the onset of superconductivity, it was speculated that heavy fermion superconductivity is caused by antiferromagnetic (AF) spin fluctuations associated with the disappearance of the AF ordered state at a QCP.⁸ However, in order to get a better insight into the mechanism, one needs to get a deeper understanding of the associated magnetic phase.

In the case of CeCu_2Si_2 , it turned out that a fruitful approach to get a better knowledge of this anomalous A phase was the study of the alloy system $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$.^{6,7} Since Ge is isoelectronic, but larger than Si, replacing Si by Ge merely corresponds to applying negative chemical pressure. This leads to a weakening of the hybridization between the f and conduction electrons and therefore to a stabilization of the magnetically ordered state. First investigations of the alloy were performed by Knebel *et al.*⁶ and Trovarelli *et al.*⁷

on polycrystals. Both observe a monotonous, although non-linear increase of the Néel temperature T_N with x . Inside the AF phase, several phase transitions are detected. The authors report on a first order transition in the concentration range $0.3 \leq x \leq 0.6$. For higher x , the development of this phase boundary line differs in both phase diagrams. Also the low-concentration region shows discrepancies and unspecified phase transition lines. The characterization of the various phases does not go beyond the level of speculation.

Recently, large single crystals of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ became available. These single crystals allow a much more specific and deeper investigation of the magnetism in this system. Using these single crystals it was possible to observe magnetic reflections with neutron scattering in the whole concentration range down to $x=0.05$.⁹ They revealed an incommensurate propagation vector which changes only slightly with composition, while the size of the AF ordered moment decreases strongly with decreasing Ge content, from $1.05\mu_B$ in pure CeCu_2Ge_2 (Ref. 10) to $\approx 0.1\mu_B$ at $x=0.05$.⁹ Eventually, this led to the discovery of magnetic reflections in the A phase of pure CeCu_2Si_2 .¹¹ It was found that the propagation vector $\tau=(0.215, 0.215, 0.530)$ corresponds to a nesting wave vector of the heavy quasiparticle Fermi surface, strongly supporting a spin density wave nature of this A phase.

We present here an investigation of the magnetism in this system by means of thermal expansion measurements. Linear thermal expansion measurements are especially appropriate to study phase diagrams due to their high sensitivity to detect phase transitions and the ability to distinguish between first and second order transitions. It is a thermodynamic method which detects the physical properties along different crystallographic axis.

After reporting thermal expansion measurements on single crystalline $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ with $0.01 \leq x \leq 0.45$, a detailed phase diagram of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ is presented which gives new information about the different phases and their phase boundary lines compared to the former published diagrams. An analysis of the thermal expansion data using

the Grüneisen ratio gives new hints about the nature of the magnetic phases and the evolution of the magnetism in this alloy.

II. EXPERIMENT

The $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ single crystals were grown by a modified Bridgman technique using Cu flux. With this technique, large gram-size (0.3–1 g) single crystals were obtained. The powder x-ray diffraction measurements demonstrated that $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ crystallizes in the tetragonal ThCr_2Si_2 structure. All single crystals were oriented by x-ray Laue backscattering. The structural properties of these single crystals are identical to those published for the polycrystalline samples,^{6,7} cf. the volume of the unit cell increases from $x=0$ to $x=1$ linearly by 6.2% while the c/a ratio remains almost unaffected. Additional energy-dispersive x-ray spectroscopy confirmed the samples of the $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ series to be single phase. The growth method and the characterization of the single crystals are described in detail in Ref. 12.

We performed thermal expansion measurements on $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ crystals with $x=0.01, 0.05, 0.09, 0.18, 0.25, 0.3, 0.37$, and 0.45 . Samples of the same batches were studied by specific heat measurements.¹³ The same crystals with $x=0.05, 0.25$, and 0.45 were also studied by neutron diffractometry.⁹ All measurements along the basal plane of the tetragonal structure were performed along the $[100]$ axis, except for $x=0.18$ and 0.3 whose linear thermal expansion coefficients were measured along $[110]$.

The linear thermal expansion was measured in an ultrahigh resolution capacitive dilatometer in the temperature range between 50 mK and 6 K in magnetic fields up to 8 T which is applied along the measurement direction. The thermal expansion coefficient was calculated from the differential quotient of the change of the length, l , with respect to a temperature interval. The linear thermal expansion coefficient $\alpha(T)$ is defined as $l^{-1} \cdot \partial l / \partial T$. The volume expansion coefficient $\beta(T)$ for a tetragonal system is $\beta(T) = 2 \cdot \alpha_a(T) + \alpha_c(T)$, where α_a and α_c are the linear thermal expansion coefficients along $[100]$ and $[001]$, respectively. A second order phase transition is evidenced by a step in the thermal expansion coefficient. The transition temperature is determined by using the equal-areas construction. By contrast, a first order transition is characterized by a step in the length. The transition temperature is then taken to be at the corresponding peak position in β .

All presented measurements except for a few cases explicitly mentioned in the text were performed by cooling down the sample and simultaneously measuring the length change.

III. RESULTS

Figure 1 shows the linear thermal expansion coefficients α_a and α_c as well as the volume thermal expansion coefficient β for the $x=0.37$ sample at $B=0$. The step at $T_N=3.04$ K corresponds to the second order transition from the paramagnetic to the AF state. One observes a negative

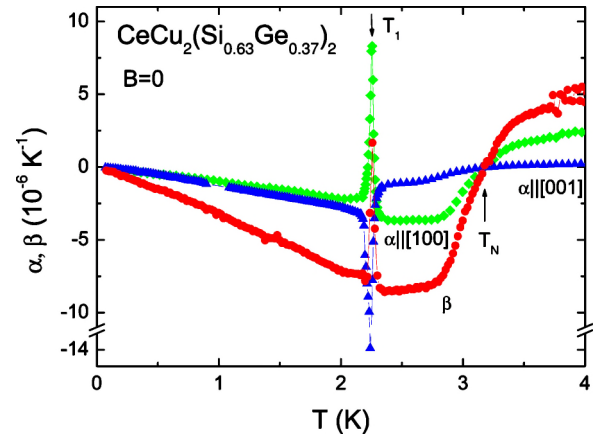


FIG. 1. (Color online) Linear and volume expansion coefficients of $\text{CeCu}_2(\text{Si}_{0.63}\text{Ge}_{0.37})_2$ vs T .

step along both directions. The peak at $T_1=2.25$ K $< T_N$ clearly marks a first order transition. Here, the sign of the peak differs along both directions. A positive sign is observed for $\alpha_{\parallel}[100]$, whereas α is negative for $\alpha_{\parallel}[001]$. The volume thermal expansion coefficient β shows only a less pronounced positive peak at T_1 .

In Fig. 2, the evolution of the volume thermal expansion coefficient $\beta(T)$ with decreasing Ge content from $x=0.37$ to $x=0.01$ is shown. The step in $\beta(T)$ at T_N shifts continuously to lower temperature with decreasing Ge content. By contrast, T_1 decreases only slightly between $x=0.37$ and $x=0.3$ being very close to T_N for the latter concentration. For $x=0.25$, it is not possible anymore to separate the first order and the second order transition from each other. But the pronounced peak at the bottom of the second order step suggests that both transitions have merged. In order to test this hypothesis, a magnetic field is applied. The results for $\alpha_{\parallel}[100]$ for $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ are shown in Fig. 3 for $B=0$ T, 4 T, and 8 T, respectively. In an external field the first order peak is obviously separated from the second order step: At 8 T, $T_1=1.38$ K is well below $T_N=1.65$ K. At this field, a further transition of apparently magnetic origin is observed at

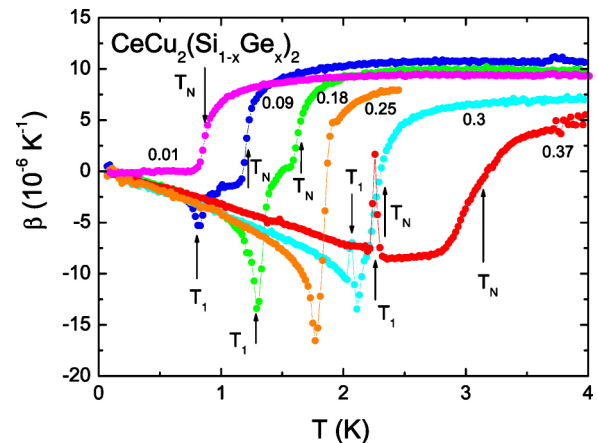


FIG. 2. (Color online) Volume thermal expansion coefficient β of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ vs T for $0.01 \leq x \leq 0.37$ measured upon cooling.

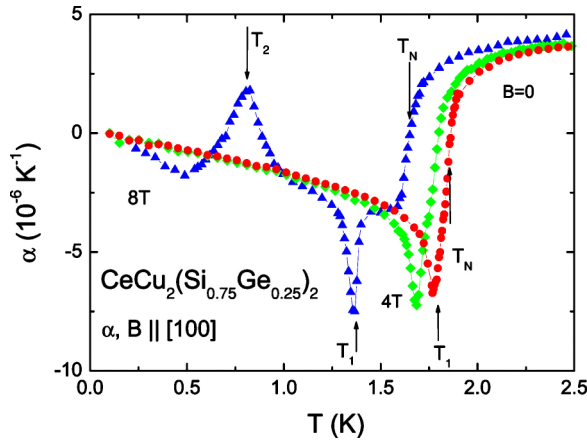


FIG. 3. (Color online) Thermal expansion coefficient α of $\text{CeCu}_2(\text{Si}_{0.75}\text{Ge}_{0.25})_2$ along the basal plane vs T at different fields $B=0$ T, 4 T, and 8 T, respectively.

$T_2 \approx 0.8$ K. This anomaly was not investigated further.

For Ge concentrations $x < 0.25$, we again observe a first order transition at $T_1 < T_N$, well separated from T_N . In Fig. 4, the linear and volume expansion coefficients are shown for $x=0.09$. The transition into the AF phase manifests itself as a second order transition at $T_N=1.25$ K with a negative jump for both crystallographic directions. However, the first order transition at $T_1=0.8$ K is now marked by a much smaller, broader peak which is negative for both directions, in contrast to the situation found for $x > 0.25$. Since the same behavior is observed for $x=0.18$, this indicates a change in the nature of this phase transition from $x > 0.25$ to $x < 0.25$. A further anomaly is observed for $T_c=0.12$ K. It is assumed to be associated to the SC transition as indicated by a drop to zero of the resistivity at 0.2 K.¹⁴

For $x=0.09$, we could observe clear thermal hysteresis effects at T_1 . In Fig. 5, we compare α upon cooling and warming. While no hysteresis can be resolved at T_N and T_c , a clear shift between cooling and warming can be found at T_1 , giving direct evidence for the first order character of this transition. Even larger hysteretic effects were observed at lower Ge concentration. The inset of Fig. 5 shows the linear

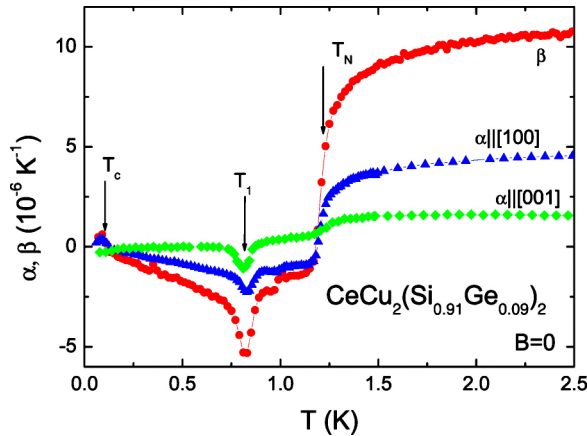


FIG. 4. (Color online) Linear and volume expansion coefficients of $\text{CeCu}_2(\text{Si}_{0.91}\text{Ge}_{0.09})_2$ vs T .

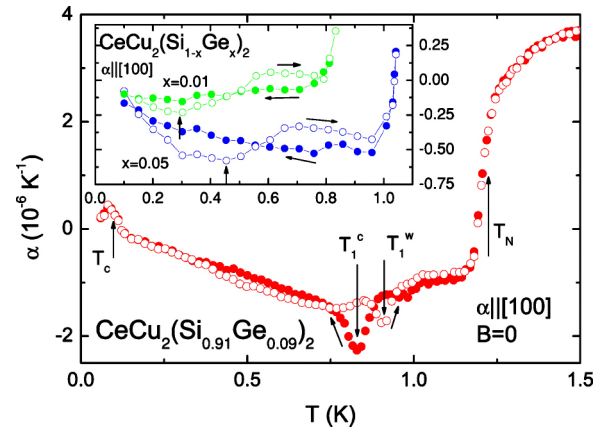


FIG. 5. (Color online) Thermal expansion coefficient α of $\text{CeCu}_2(\text{Si}_{0.91}\text{Ge}_{0.09})_2$ along [100] vs T upon cooling (open symbols) and warming (closed symbols). Arrows label the transition temperature T_1 for cooling (c) and warming (w). Inset, α vs T for $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ with $x=0.01$ and 0.05 along [100] upon cooling (open symbols) and warming (closed symbols). Arrows mark the transition temperature T_1 , see text.

thermal expansion data for $x=0.01$ and 0.05 along [100] measured upon cooling and warming. Whereas the curves for cooling show a monotonous temperature dependence, the curves for warming up exhibit an S-like shape. This behavior seems to be caused by a less pronounced first order transition. We define somehow arbitrarily the transition temperature T_1 as the minimum of the warming curve as no indications for a clear transition below T_N is found in the data for cooling. The reason for choosing this definition for T_1 is that at higher Ge content T_1 also corresponds to the minimum in $\alpha(T)$. With this definition T_1 can be followed down to $x=0.01$. The origin of the pronounced difference between cooling and warming is not clear to us. Large hysteretic effects might lead to the disappearance or the smearing out of the transition at T_1 .

We also investigated a single crystal with a larger Ge content $x=0.45$. The very large size of this single crystal (~ 1 g) lead to some difficulties with the reproducibility of the measurements, likely due to thermalization problems and sample inhomogeneity. The most reliable results (as deduced from a comparison with the specific heat results¹³) were obtained for the thermal expansion along the [001] direction, which are shown in Fig. 6. The second order transition from the paramagnetic to the AF state is again marked by a negative step in $\alpha(T)$. However, the behavior below T_N becomes more complicated. There is still a well defined first order transition at $T_1=2.14$ K with a signature similar to that observed for $x=0.37$. But at lower temperature a further pronounced anomaly appears at $T_{LI}=1.28$ K. This anomaly is broader and shows some substructure probably due to the large crystal size. Huge hysteretic effects identify this anomaly as first order transition, too. Similar hysteretic effects were already observed in the same temperature range in pure CeCu_2Ge_2 .¹⁷ Both first order transitions at T_1 and T_{LI} were also observed in neutron scattering experiments.⁹ A preliminary analysis of the neutron data indicates a change of the orientation of the moments at T_1 , while at T_{LI} both a

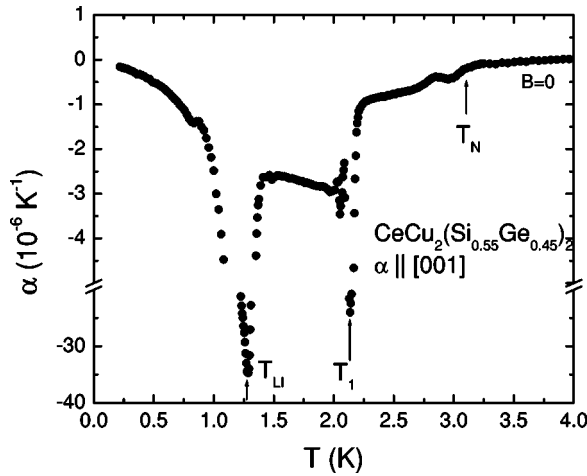


FIG. 6. Thermal expansion coefficient α of $\text{CeCu}_2(\text{Si}_{0.55}\text{Ge}_{0.45})_2$ along [001] vs T .

reorientation of the moments and a lock in of the propagation vector seem to occur simultaneously.⁹

IV. DISCUSSION

Our thermal expansion measurements on large $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ single crystals reveal new information about the magnetism in the system. The T - x magnetic phase diagram deduced from our results is shown in Fig. 7. It is supported by results from specific heat,¹³ and neutron diffraction measurements,⁹ which are also included.

The first main feature of this phase diagram is the continuous decrease of T_N , the transition temperature from the paramagnetic to the AF phase, with decreasing Ge content. Its development is consistent with previous results on polycrystalline samples.^{6,7} The second main feature is the observation of a first order transition at $T_1 \leq T_N$ in the whole concentration range $0.01 \leq x \leq 0.45$. For $0.3 \leq x \leq 0.45$ the $T_1(x)$ phase boundary is in good agreement with that deduced for polycrystals.^{6,7} However, for $x \leq 0.3$ this phase transition could not be observed in the previous experiments. Only one of the published phase diagrams shows a single point at $x=0.1$ related to a first order transition.⁷ By contrast, in our measurements this transition is very well marked by a peak in $\beta(T)$ for $x \geq 0.09$. Only at lower Ge concentration the determination of the transition temperature becomes difficult due to hysteretic effects as described in the previous paragraph. Recently, a similar anomaly was observed at $T_1 \approx 0.35$ K in $\alpha(T)$ and $C(T)$ measurements in a pure A-type CeCu_2Si_2 single crystal.¹¹ Evidence for a transition at around 0.3 K was also found in earlier thermal expansion measurements of polycrystalline CeCu_2Si_2 .¹⁵

Another new result in this phase diagram is the presence of a tetracritical point at $x \approx 0.25$, connected with the merging of the $T_N(x)$ and the $T_1(x)$ phase boundaries. A direct evidence for the merging of both lines is the thermal expansion of the $x=0.25$ single crystal for which the two transitions cannot be resolved at $B=0$, but are well separated at $B=8$ T. The specific heat measurements on the same samples

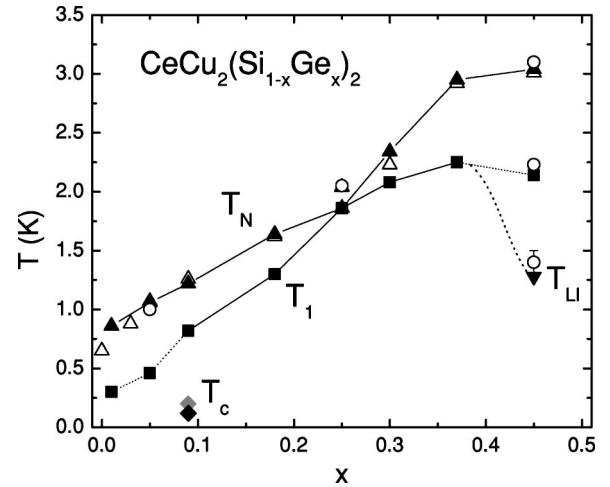


FIG. 7. Magnetic phase diagram of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ for $x \leq 0.45$. Triangles mark the second order transition at T_N , squares the first order transition at T_1 , diamonds the superconducting transition at T_c , and the down-side triangle the anomaly at T_{LI} . Dotted lines indicate possible interconnection of different phase boundary lines. The closed symbols present the results from thermal expansion, the open triangles from specific heat measurements (Ref. 13), the open circles from neutron scattering measurements (Ref. 11). The closed diamond marks the position of the superconducting jump in resistivity (Ref. 14).

show a broad transition whose jump height is enlarged compared to that for other concentrations. This also hints to a merging of both transitions.¹³ Further evidence for this comes from the observation that the signature of the transition is different for $x > 0.25$ than for $x < 0.25$. While the peak at T_1 in $\alpha(T)$ along the basal plane is negative in the whole concentration range, along [001] it changes from negative for $x < 0.25$ to positive for $x > 0.25$.

The microscopic nature of the transition at T_1 is yet not clear. Neutron scattering experiments⁹ suggest that for $x \leq 0.25$ the dominant effect is a lock-in of the incommensurate propagation vector, while for $x \geq 0.25$ the situation is even less clear. It might be a combination of a lock-in transition and a reorientation of the moments. Then one would expect that the transition observed at T_{LI} in the $x=0.45$ single crystal should merge at lower Ge content with the T_1 transition, as tentatively indicated by a dotted line in the phase diagram of Fig. 7 and as suggested in one of the phase diagrams obtained from polycrystals.⁷ This has, however, to be confirmed by further neutron scattering experiments.

The concentration region $x \approx 0.25$ is not only marked by the tetracritical point and the change in the nature of the T_1 transition, also pronounced changes in other physical properties are found which give hints to a change in the nature of the magnetic state. It seems that the nature of the f electrons evolves from a rather localized state for $x > 0.25$ to itinerant heavy electrons for $x < 0.25$. This is evidenced by the evolution of the Grüneisen parameter Γ which relates the volume thermal expansion coefficient β and the specific heat C : $\Gamma = V_{\text{mol}} / \kappa_T \cdot \beta / C$, where κ_T is the isothermal compressibility. Pressure experiments¹⁶ on CeCu_2Si_2 give $\kappa_T = 8.3 \times 10^{-12} \text{ Pa}^{-1}$. The molar volume V_{mol} was calculated

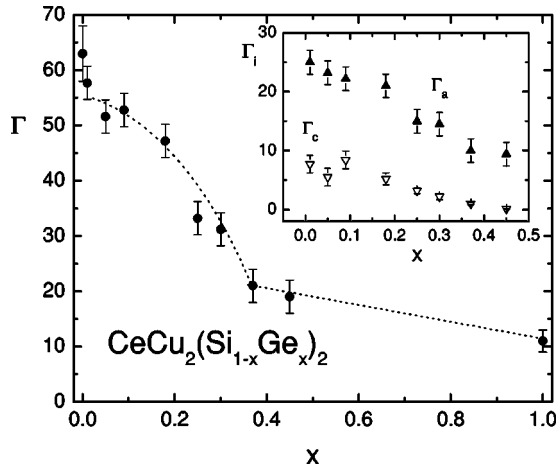


FIG. 8. Grüneisen parameter Γ of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ taken at $T=1.1 \cdot T_N$ for $0 \leq x \leq 1$. Lines are guides to the eyes. Inset, uniaxial Grüneisen parameter Γ_i of $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ along the basal plane (Γ_a , up-side triangles) and along the c -axis (Γ_c , down-side triangles) in the paramagnetic state vs x .

for each concentration using its specific lattice parameters. The specific heat data were taken from measurements on the same single crystals or on single crystals from the same batch.¹³ Figure 8 shows the dependence of Γ on the Ge concentration taken at $T=1.1 \cdot T_N$ in the paramagnetic state. We include the result for pure CeCu_2Ge_2 deduced from thermal expansion data of Ref. 17 and the specific heat data of Ref. 18. For pure CeCu_2Ge_2 , $\Gamma \approx 10$ is already enhanced compared to that of ordinary metals ($\Gamma \approx 1$) suggesting that even in CeCu_2Ge_2 f electrons are slightly hybridized with the conduction electrons. Decreasing the Ge content in the alloy leads at first to only a minor increase of Γ , to $\Gamma \approx 20$ for $x=0.37$. Then however, further decreasing x results in an abrupt increase of Γ , leading to a value of $\Gamma \sim 48$ at $x=0.18$ and eventually to $\Gamma \sim 58$ at $x=0.01$. This value is very close to $\Gamma=63$ reported for pure CeCu_2Si_2 .¹⁵ Thus, for $x < 0.25$ we observe Γ values which are typical for heavy fermion systems.

The Grüneisen parameter describes the volume dependence of the characteristic energy $k_B T^*$: $\Gamma \propto -\ln T^* / \ln V$. For a Kondo system the characteristic energy is given by the Kondo-lattice energy which is related to the hybridization of the f electrons and the conduction electrons. It is experimentally accessible through the single-ion Kondo temperature T_K . The weakly enhanced Grüneisen parameter characterizes CeCu_2Ge_2 and $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ with $x \geq 0.37$ as a Kondo lattice with weak hybridization of the f electrons. The hybridization seems to change abruptly in the concentration range $x \approx 0.25$. At lower Ge concentration, the f electrons become more itinerant and form a typical HF state. This is also supported by a similar concentration dependence of the Sommerfeld coefficient γ .¹³ The strong increase of both Γ and γ for $x < 0.25$ indicates a pronounced increase of the mass renormalization of the heavy electrons in this concentration range.

For CeCu_2Ge_2 , the Kondo temperature $T_K \approx 4$ K and the AF ordering temperature $T_N=4.2$ K are very close.¹⁹ Therefore, the onset of the AF ordered state prevents the full de-

velopment of the Kondo screening at low temperatures and the formation of really heavy quasiparticles. Decreasing the Ge content leads to an increase of T_K and simultaneously to a somewhat stronger decrease of T_N . At a given ratio T_K/T_N which seems to be reached around $x \approx 0.25$, the onset of AF ordering cannot anymore suppress the Kondo mechanism in an efficient way allowing the formation of heavy electrons. This fact is reflected in the strong increase of Γ and γ .

The AF ordering temperature T_N in CeCu_2Si_2 can be completely suppressed to $T=0$ by either applying a slight hydrostatic pressure or replacing part of Si by Cu. Presently, two scenarios are discussed for the description of a QCP: the local scenario^{20,21} and the itinerant scenario.²²⁻²⁴ The basic difference is related to the behavior of the heavy electrons at the QCP. In the local QCP scenario the heavy quasiparticles break up at the QCP.^{20,21} They exist only on the paramagnetic side of the QCP. On the magnetically ordered side, they disintegrate into light conduction electrons and localized f electrons which order magnetically.²⁵ By contrast, in the itinerant scenario the heavy electrons remain stable also on the magnetically ordered side of the QCP.²²⁻²⁴ In this case, the magnetic order corresponds to a spin density wave (SDW) connected with an instability of the Fermi surface of the heavy electrons. In this scenario, the disintegration of these heavy electrons into light electrons and localized f electrons occurs further away from the QCP within the AF ordered region. On the magnetic side close to the QCP, one expects significant differences in the magnetic state between the two scenarios. In $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$, the enhancement of both the Grüneisen parameter and the Sommerfeld coefficient for $x < 0.25$ point to the existence of heavy quasiparticles also in the AF ordered region away from the QCP. This gives further support in favor of the itinerant (SDW) scenario for CeCu_2Si_2 as already inferred from the non-Fermi-liquid power laws in $\Delta\rho(T)$ and $\gamma(T)$ following $T^{3/2}$ and $(\gamma_0 - aT^{1/2})$ dependence, respectively, in the normal state of “S-type” CeCu_2Si_2 close to the QCP.^{4,26}

The measurements of the linear thermal expansion coefficient allow a study of the uniaxial Grüneisen parameter, Γ_i , where Γ_a indicates the uniaxial Grüneisen parameter along the basal plane and Γ_c along [001]. The deduced values are shown in the inset of Fig. 8. The Grüneisen parameter along the basal plane exceeds that along the tetragonal c -axis by roughly a factor of 3. Apparently, the hybridization is more important along the basal plane. This behavior is in good agreement to that found for CeCu_2Si_2 in previous studies.¹⁵

V. CONCLUSION

The focus of our thermal expansion study on large high-quality $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ single crystals is to establish a more detailed phase diagram than previously reported.^{6,7} The composition dependence of the antiferromagnetic order transition at T_N is in good agreement with these published results. However, the measurements enable to define the first order transition at T_1 within the magnetic phase in the whole concentration range studied. Furthermore, a tetracritical point at $x \approx 0.25$ at which the $T_1(x)$ and $T_N(x)$ phase boundary lines merge marks a change in the nature of the transition

at T_1 between $x > 0.25$ and $x < 0.25$ and thus in the magnetic nature of the phases.

The Grüneisen ratio which reflects the hybridization strength of the f electrons increases only slightly between pure CeCu_2Ge_2 and $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ with $x=0.37$, but rises rapidly in the region of the tetracritical point around $x \approx 0.25$. This suggests that the formation of the heavy fermions out of the localized f electrons and the light conduction electrons occurs in this concentration range, well before the QCP located slightly beyond the true stoichiometry point,

i.e., in the Cu-rich side of the homogeneity range (“S-type” CeCu_2Si_2).²⁷ The heavy quasiparticles are found to exist on the magnetic side of the QCP. A local QCP can therefore be excluded. These results give a strong support for the itinerant SDW scenario describing the QCP in CeCu_2Si_2 .

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