Observation of two critical points linked to the high-field phase B in CeCu₂Si₂

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We present thermal expansion and magnetostriction measurements on a CeCu₂Si₂ single crystal of A/S type up to 17.9 T magnetic field applied along the crystallographic a direction ($\Delta L \parallel a \parallel H$) and down to 0.015 K temperature. We identify clear thermodynamic anomalies at the superconducting transition T_c and at two second-order transitions $T_{A,B}$ into ordered phases A and B. Our measurements establish the boundary of phase B at high field and low temperature. No evidence for additional high-field phases above B is found up to the maximum field. We speculate based on our experimental results that (i) phase B is similar to phase A of spin-density wave type and (ii) the first-order phase transition between A and B is caused by Fermi-surface reconstruction. We furthermore identify a quantum critical point at $H_c \simeq 17$ T, where T_B is suppressed to zero, and a bicritical point at (0.35 K, 7.0 T), where phase lines $T_A(H)$ and $T_B(H)$ meet.

I. INTRODUCTION

CeCu₂Si₂ is one of the most intriguing heavy-fermion superconductors since its discovery in 1979 [1]. For decades, it is strongly believed that superconductivity and magnetism in CeCu₂Si₂ do not only coexist side by side, but that superconductivity is actually caused by magnetic fluctuations associated with a nearby quantum critical point [2–4]. Strong evidence for this scenario was found by Stockert *et al.* in inelastic neutron scattering (INS) experiments [5], where gapped spin excitations inside the superconducting (SC) state were observed. Usually, it is assumed that because of strong onside Coulomb repulsion between f electrons, magnetic mediated superconductivity in heavy-fermion systems should result in a SC order parameter with either d-wave or p-wave symmetry. While this simple approach has been supported by results on CeCoIn₅ and CeIrIn₅ systems [6], recent experiments probing the symmetry of the superconducting order parameter in CeCu₂Si₂, such as thermal conductivity, specific heat, and magnetization partially conducted under rotational fields find no evidence for nodes in the energy gap [7–9]. A recent theoretical study favors a nodeless $s^{\pm}$-wave function while taking into account intra- as well as strong interband magnetic quantum critical scattering [10]. On the other hand, Pang *et al.* propose an effective two-band d-wave model [11], which also explains fully gapped behavior at very low temperatures and is in accordance with a sign change of the SC order parameter as found in INS experiments [5]. Needless to say, the discussion is ongoing and the relationship between superconductivity and magnetism remains a highly topical area of research, 39 years after the discovery of superconductivity in magnetic CeCu₂Si₂.

The ground state of *homogeneous* CeCu₂Si₂ is sensitive to the precise stoichiometry of the sample, because minimal Cu excess or deficiency changes the hybridization between f electrons and conduction electrons [4]. Small deviations from the 1:2:2 ratio produce either an antiferromagnetically ordered A-type (A) ground state, an only superconducting (S) ground state, or an A phase that undergoes a transition into superconductivity at lower temperatures (A/S) as depicted schematically in the inset of Fig. 1 [4]. A/S-type single crystals are closest to the nominal 1:2:2 ratio [12]. Common to all three types of single crystals is the occurrence of a second field-induced phase B. The phase transition into phase B has been found so far in measurements of the elastic constants [13], the resistivity [14], and in magnetization experiments [15]. Lang *et al.* detected a clear anomaly at the onset of phase B in magnetostriction measurements in magnetic fields to 8 T and temperatures down to 0.25 K [16], which promotes dilatometric measurements as a suitable probe to precisely track the phase boundary in even higher magnetic fields.

In the following, we map out the T-H phase diagram of an A/S-type single-crystal CeCu₂Si₂ with magnetic fields applied along the crystallographic a direction ($\Delta L \parallel a \parallel H$). Our work aims at gaining a better understanding of the field-induced phase B, which is currently widely unknown. We furthermore want to explore if additional phases emerge in higher magnetic fields. Note, the here presented phase diagram is already mentioned in two review articles [17,18], but without showing the actual experimental data.

II. METHODS

CeCu₂Si₂ crystallizes in the tetragonal ThCr₂Si₂ structure with space group $I4/mmm$. Large single crystals were grown by crucible free cold boat technique [19]. We observe
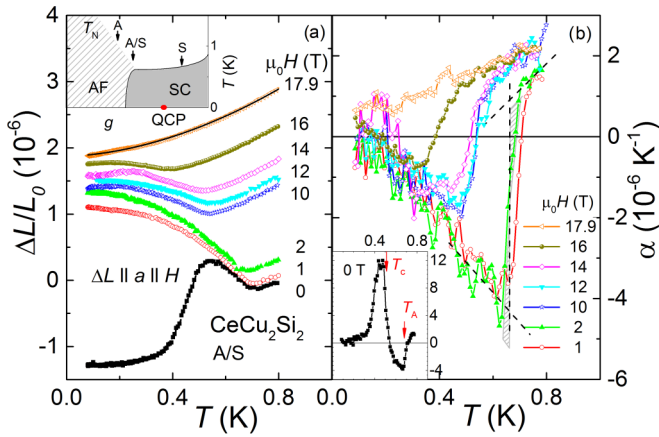
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FIG. 1. (a) Length change $\Delta L/L_0$ normalized to the initial value L_0 versus temperature T from 0.08 to 0.8 K for $\Delta L \parallel a \parallel H$ between 0 and 17.9 T in $CeCu_2Si_2$. The zero-field measurement shows two phase transitions, whereas measurements in magnetic field only reveal one transition into either phase A or B. The solid line represents a fit to the data at 17.9 T as expected for a QCP of AFM SDW type in 3D [21] (for details, see Discussion). The corresponding thermal expansion coefficient $\alpha(T)$ for all measurements in magnetic field is shown in (b). We estimate the precise transition temperatures $T_{A,B}$ by equal area construction in $\alpha(T)$ as illustrated for the 2 T data with broken lines and shaded areas. The inset in (a) shows the schematic phase diagram T versus g. The hybridization g defines the ground state of $CeCu_2Si_2$ as A, A/S, or S type as indicated by arrows. The inset in (b) displays $\alpha(T)$ in zero field with anomalies at the SC transition $T_c = 0.51$ K and at the onset of AFM order at $T_A = 0.7$ K.

antiferromagnetic (AFM) order into the A phase at $T_A = 0.7$ K and the onset of superconductivity at $T_c = 0.51$ K, which places the A/S sample right at the spot in the phase diagram where superconductivity and magnetism compete [4,20].

Magnetostriction and thermal expansion of the $L_0(0 \text{ T}, 300 \text{ K}) = 2.26$-mm-long sample is measured inside the vacuum chamber of a dilution refrigerator with a capacitive dilatometer. The dilatometer is manufactured from high resistive CuBe alloy to avoid heating effects due to eddy currents during increasing and decreasing magnetic-field sweeps. The sample itself was thermally decoupled from the dilatometer with a graphite disk and anchored directly to the mixing chamber with a braid made out of individual silver wires. This design allows one to bypass cooling difficulties caused by a large nuclear Schottky contribution to the specific heat of copper in high magnetic fields.

III. RESULTS

At first, we concentrate on thermal expansion measurements between 0.08 and 0.8 K in zero and constant magnetic fields as shown in Fig. 1. The sample length $\frac{\Delta L}{L_0}(T)$ measured in 0 T expands with increasing temperature inside the SC phase, shows a steplike anomaly at the entrance to phase A with negative slope inside A and a minimum at the transition into the paramagnetic (PM) state followed by an increase of the sample length for $T > 0.7$ K. Experimental data taken in magnetic fields up to 16 T show as well a negative $\frac{\Delta L}{L_0}(T)$ behavior inside phases A and B and positive length changes when the
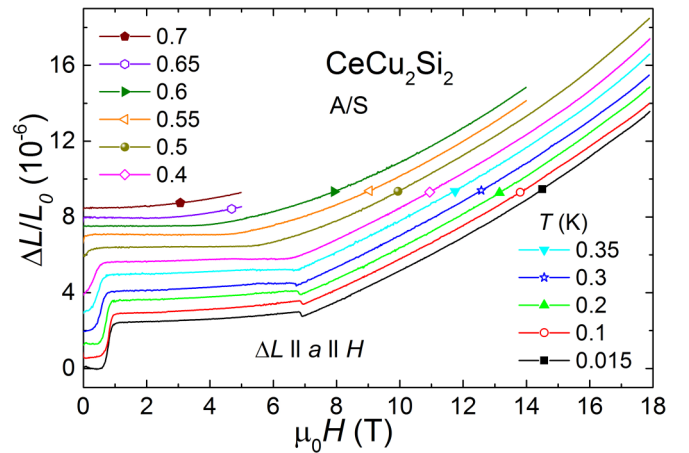

FIG. 2. Length change $\Delta L/L_0$ normalized to L_0 versus applied magnetic field $H \parallel \Delta L \parallel a$ between zero and 17.9 T is shown for temperatures in the range 0.015–0.7 K. We observe clear steps at the SC transition at 1 T and around 7 T separating phase A from B for lowest temperatures. The anomaly at 1 T shifts to lower fields with increasing T, whereas the step between phase A and B is clearly visible to 0.3 K and gets less pronounced at higher temperature.

order is thermally suppressed. We want to emphasize that phases A and B cannot be distinguished by the temperature dependence of the thermal expansion. A closer look at the data collected at 14 T reveals a broad maximum at around 0.25 K inside phase B. This additional anomaly only occurs in one of the measurements and further investigations are necessary to clarify its origin. The sample length increases monotonically with no obvious anomalies in the highest field of 17.9 T.

The thermal expansion coefficient is defined as $\alpha(T) = L_0^{-1} \times \frac{\partial \Delta L_a(T)}{\partial T}$ along one crystallographic direction $a \parallel \Delta L \parallel H$. It measures directly the uniaxial pressure p_i dependence of the entropy S via the Maxwell relation $\frac{\partial S}{\partial p_i} = -\frac{\partial L_i}{\partial T}$ and is therefore well suited to investigate phase transitions and related phenomena with enhanced entropy contributions. Figure 1(b) shows the thermal expansion coefficient $\alpha(T)$ for the corresponding measurements in (a). The transition from paramagnetism into phase A or B is of second order and characterized by a step in the thermal expansion coefficient. We use an equal area construction to get precise values of $T_{A,B}$ as demonstrated for the 2 T data in Fig. 1(b) and indicated by shaded areas. The inset in Fig. 1(b) shows $\alpha(T)$ at zero field with a first-order-like anomaly at 0.51 K that marks the onset of superconductivity.

The change of sample length $\frac{\Delta L}{L_0}(H)$ as a function of magnetic field H for temperatures $T \leqslant 0.7$ K is presented in Fig. 2. We observe almost no sample expansion for 1 T $< \mu_0 H < 7$ T and small quadratic field dependence above 7 T at lowest temperature. A positive step at 1 T indicates the suppression of superconductivity and a negative step at $\sim$7 T marks the phase boundary between phases A and B.

The magnetostriction coefficient $\lambda(H) = (L_0 \mu_0)^{-1} \times \frac{\partial \Delta L_a(T)}{\partial H}$ is the first derivative of the sample length in respect to field. Panels (a)–(d) in Fig. 3 show the evolution of shape and position of the three anomalies in $\lambda(H)$ at 1 T (SC to phase A), 7 T (phase A to B), and 17 T (suppression phase B) upon increasing temperature in detail. The sharp positive
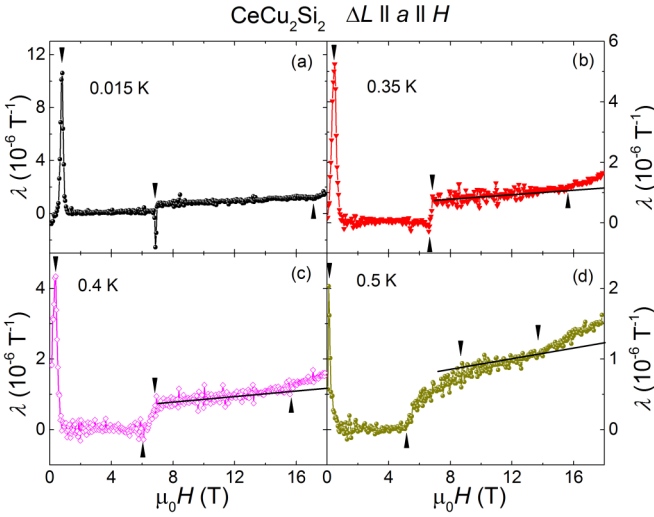
FIG. 3. The magnetostriction coefficient $\lambda(H)$ is shown for four selected temperatures: (a) 0.015 K, (b) 0.35 K, (c) 0.4 K, and (d) 0.5 K illustrating how signatures at the phase transitions (marked with arrows) change with increasing temperature. Please see text for details.

FIG. 4. T-H phase diagram of an A/S single-crystal $CeCu_2Si_2$ for $\Delta L \parallel a \parallel H$ as estimated by thermal expansion (open symbols) and magnetostriction (solid symbols) experiments. First-order discontinuities are marked with a gray phase boundary and second-order phase lines are black, giving rise to a BCP at about (0.35 K; 7.0 T) and a QCP at (0 K; 17 T).

delta peak in (a) that indicates a first-order transition between SC ground state and phase A becomes broader and moves to zero field below 0.5 K. The phase boundary between phases A and B is a sharp negative delta anomaly at lowest temperature suggesting a first-order type, too. It develops into two separated kinks at 0.35 K [(panel (b)] that get more distant with increasing temperature as seen in panels (c) and (d). The suppression of phase B can be inferred from a change of slope in $\lambda(H)$ for fields close to 17 T [panel (a)] with slightly reduced critical fields in higher temperatures.

IV. DISCUSSION

Figure 4 summarizes the results of our thermal expansion and magnetostriction measurements up to 18 T for magnetic fields applied parallel a in A/S-type $CeCu_2Si_2$. We find superconductivity below 0.51 K in zero field, which is suppressed by a magnetic field in excess of 1 T. The SC ordered phase is surrounded by phase A that evolves into phase B at about 7 T at lowest temperatures. Phase B is suppressed in fields of 17 T and higher. A careful inspection of the data did not reveal any hint for additional phases above B, at least up to 17.9 T. The SC transition and the transition between phases A and B are of first order, marked as gray lines in Fig. 4. All other transitions are of second order (black lines). The phase diagram exhibits a bicritical point (BCP) at about (0.35 K, 7 T), where two second-order and one first-order phase boundaries converge.

In the following, we discuss the nature of phase B. To our knowledge, no experimental data are published probing the local microscopic environment above 7 T. Nevertheless, certain conclusions on the order in the B phase can be drawn from macroscopic quantities and similar temperature dependence as found in phase A. Neutron diffraction experiments carried out inside phase A verify incommensurate antiferromagnetic (AFM) order of spin-density wave (SDW) type with
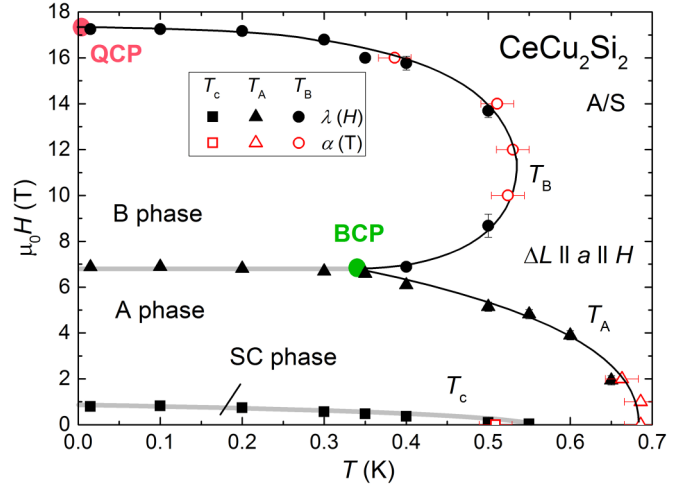
(0.215, 0.215, 0.530) ordering vector, which is caused by renormalized Fermi-surface (FS) nesting [22] of the heavy bands. The ordered moment is about $0.1\mu_B$. Muon spin relaxation experiments in zero field suggest phase separation of AFM and SC regions at the transition T_c with increasing SC volume for decreasing temperatures [20].

The anomalies in $\alpha(T)$ are very similar at the phase boundaries $T_A(H)$ and $T_B(H)$ and of second-order type. This suggests a change of symmetry between PM state and both phases A and B, respectively. It is microscopically proven for phase A [22]. Thermal expansion inside phase B has a similar temperature dependence [negative $\alpha(T)$] as in A; compare Fig. 1. The transition between phase A and B is most likely a first-order discontinuity as indicated by the step in the magnetostriction. Unfortunately, we have only measured at a certain temperature in one field direction, either increasing field or decreasing field. Therefore, we cannot identify possible hysteretic behavior, which is often observed at first-order transitions. The magnetostriction coefficient $\lambda(H)$ is almost zero in phase A and therefore slightly different from the small linear field dependence as found in the B phase. It is worth noting that no universal field dependence of $\lambda(H)$ for AFM ordering is expected. It rather depends on the shape and anisotropy of the energy dispersion function for a specific type of magnetic order.

Now, we will have a look at other thermodynamic quantities. Magnetization experiments up to 11.5 T ($H \parallel a$) carried out by Tayama *et al.* [15] reveal a linear increase of the magnetization $M(H)$ inside phase A as well as inside B with similar slope. A step is observed in $M(H)$ at the transition between both phases at lowest temperatures, which is also an indication for its first-order nature. There is only a tiny increase of the magnetization by $(2 \times 10^{-3})\mu_B$ on going from phase A to B. This can be caused either by a small change of the magnetic moment or by a large moment change that is predominantly screened by the AFM order. The latter case seems to be less likely, because

large ordered moments were not observed in neutron-scattering experiments inside phase B [23].

The results in thermal expansion, magnetostriction, as well as magnetization experiments lead us to the conclusion that phase B is similar to phase A, i.e., being of SDW AFM order with a small ordered moment in the order of $\sim 0.1 \mu_B$.

Renormalized band-structure calculations in zero field [24] estimate separate sheets of Fermi surface for light and heavy quasiparticles. Relatively light quasiparticles exhibiting five times the bare electron mass m_e are verified in de Haas–van Alphen (dHvA) experiments [25]. Heavy quasiparticles ($500m_e$) are expected on a quasi-two-dimensional (quasi-2D) Fermi surface in the shape of warped cylinders along the c axis and small pockets. Zwicknagl *et al.* [18,24] suggest that the transition between phase A and phase B is a Lifshitz transition and that the FS topology changes from quasi-2D to three-dimensional (3D). Unfortunately, it is impossible to test the change of dimensionality by quantum oscillation measurements, because of the heavy mass of the quasiparticles. Hunt *et al.* reported only a small change in the dHvA frequency and mass of the light bands from 171 T, $4.62m_e$ inside the A phase to 162 T, $5.15m_e$ inside phase B [25]. We wish to emphasize that our observation of a first-order phase transition between phases A and B would be consistent with a Lifshitz transition [26].

A possible alternative scenario to a Lifshitz transition occurring at 7 T is domain reorientation. Stockert *et al.* find, indeed, in neutron-diffraction experiments at $H = 0$ symmetry, equivalent peak positions that point to the existence of AFM domains inside phase A [22]. Assuming the transition at 7 T is solely due to domain reorientation, clear magnetic Bragg peaks are expected inside phase B at or close to some of the Bragg peaks observed in phase A. However, elastic neutron-scattering experiments in phase B did not reveal any magnetic Bragg peaks [23]. This excludes the transition at 7 T to be merely due to domain reorientation.

Finally, we comment on the second-order phase line $T_B(H)$ that is suppressed in magnetic fields and gives rise to a QCP with critical field $H_c \cong 17$ T (see Fig. 4). The thermal expansion measurement that comes closest to H_c is the one carried out in 17.9 T. Figure 1(a) shows a power-law fit $\Delta L/L = a_0 + a_1 T^{3/2} + a_2 T^2$ (solid line) with reasonable agreement within scattering of the experimental data and a_1, $a_2 > 0$. The

$T^{3/2}$ term is hereby expected for an AFM SDW QCP in three dimensions [21] and the quadratic term typical for (noncritical) Landau Fermi-liquid contributions. While this observation is only a first hint for quantum critical behavior occurring close to this magnetic-field-induced QCP, it is likely to stimulate further experimental investigations of other bulk as well as microscopic properties.

Note, the combination of thermal expansion measurements together with specific heat data $C(T)$ is an extremely successful method to detect and classify quantum critical behavior, because the Grüneisen ratio $\Gamma \sim \alpha(T)/C(T)$, a measure of the relevant energy scale, diverges at QCPs with certain power laws [21]. This approach has been applied, i.e., to classify the zero pressure QCP in $CeNi_2Ge_2$ as three-dimensional QCP of AFM-SDW type [27] with the same $\Delta L/L \sim T^{3/2}$ critical contribution to the length change as observed in this field-induced QCP in $CeCu_2Si_2$. In contrast, the SDW description was excluded based on this analysis for other heavy-fermion systems such as $CeCu_{5.8}Ag_{0.2}$ [28] or $YbRh_2Si_2$ [27,29].

V. SUMMARY

In conclusion, we have used thermal expansion and magnetostriction experiments to establish the complete H-T phase diagram of A/S-type $CeCu_2Si_2$ up to 17.9 T. We confirm the existence of three different types of ordering; superconductivity, and magnetic phase A and B, with the SC phase occurring below 0.51 K and in magnetic fields up to 1 T. Phase A is stable up to 7 T showing a weak first-order transition into phase B. Our dilatometric measurements support the picture that phase B is of similar SDW type as phase A. We identify two field-induced critical points in the phase diagram. A QCP is observed at $H_c \cong 17$ T with hints of quantum critical behavior in the thermal expansion for $H \geqslant H_c$. We furthermore identify a bicritical point at finite temperature at 0.35 K and 7 T, where two second-order phase lines $T_A(H)$ and $T_B(H)$ and one first-order phase line merge.

ACKNOWLEDGMENTS

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