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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 for the first time 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 new quantum critical point at $H_c \simeq 17$ T, where T_B is suppresssed to zero, and a bicritical point at $(0.35 \text{ K}, 7.0 \text{ T})$, where phase lines $T_A(H)$ and $T_B(H)$ meet.

I. INTRODUCTION

 $CeCu₂Si₂$ is one of the most intriguing heavy fermion (HF) superconductors since its discovery in 1979¹. Since 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) experiments⁵ , 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 $CeCoIn_5$ and $CeIrIn_5$ systems⁶, 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 node-less s^{\pm} -wave function while taking into account intra as well as strong inter-band magnetic quantum critical scattering¹⁰. On the other hand, *Pang et al.* propose an effective 2-band d -wave model¹¹, 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 experiments⁵. Needless to say, the discussion is ongoing and the relationship between superconductivity and magnetism stays 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 electrons⁴. 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.1⁴. A/S-type single crystals are closest to the nominal $1:2:2$ ratio¹². 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 resistivity¹⁴, and in magnetization experiments¹⁵. Lang et al. detected a clear anomaly at the onset of phase B in magnetostriction measurements in magnetic fields to 8T and temperatures down to 0.25 K^{16} , which promotes dilatometric measurements as 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 $\ar{tice}^{17,18}$, but without showing the actual experimental data.

II. METHODS

 $CeCu₂Si₂$ crystalizes in the tetragonal $ThCr₂Si₂$ structure with space group I4/mmm. Large single crystals were grown by crucible free cold boat technique¹⁹. We observe antiferromagnetic (AFM) order into the Aphase at $T_A = 0.7 \text{ K}$ and the onset of superconductivity at $T_c= 0.51$ K, which places the A/S sample right at the

FIG. 1. a) Length change $\Delta L/L_0$ normalized to the initial value L_0 versus temperature T from 0.08 K to 0.8 K for $\Delta L \parallel a \parallel H$ between 0 and 17.9 T in CeCu₂Si₂. 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 $CeCu₂Si₂$ 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 $T_c = 0.51 \text{ K}$ and at the onset of AFM order at $T_A = 0.7 \text{ K}$.

spot in the phase diagram, where superconductivity and magnetism compete^{4,20}.

Magnetostriction and thermal expansion of the $L_0(0 \text{ T}, 300 \text{ K}) = 2.26 \text{ mm}$ 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 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 K 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 step-like anomaly at

FIG. 2. Length change $\Delta L/L_0$ normalized to L_0 versus applied magnetic field H || ΔL || a between zero and 17.9 T is shown for temperatures in the range 0.015 K to 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.

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 order is thermally suppressed. We want to emphasize that phases A and B can not 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 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. Fig. 1b) 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. 1b) and indicated by shaded areas. The inset in Fig. 1b) 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

FIG. 3. The magnetostriction coefficient $\lambda(H)$ is shown for 4 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.

magnetic field H for temperatures $T \leq 0.7$ 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 ~ 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) to d) in Fig. 3 show the evolution of shape and position of the 3 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 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 A and B phase 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

Fig. 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 $CeCu₂Si₂$. 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

FIG. 4. $T - H$ phase diagram of an A/S single crystal $CeCu₂Si₂$ 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 grey phase boundary and second order phase lines are black, giving rise to a BCP at about $(0.35 \text{ K}; 7.0 \text{ T})$ and a QCP at $(0 \text{ K};$ 17 T).

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 phase A and B are of first order, marked as grey 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. Neverteless, 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 Fermisurface (FS) nesting²² of the heavy bands. The ordered moment is about $0.1 \mu_B$. μ SR experiments in zero field suggest phase separation of AFM and SC regions at the transition T_c with increasing SC volume for decreasing $temperatures²⁰$.

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 can not 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 to note 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 ($H \parallel$ α) 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 later 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 $\sim 0.1 \mu_B$.

Renomalized band structure calculations in zero field 24 estimate separate sheets of Fermi surface for light and heavy quasiparticles. Relatively light quasiparticles exhibiting 5 times the bare electron mass m_e are verified in dHvA experiments.²⁵ Heavy quasiparticles $(500m_e)$ are expected on a quasi 2-dimensional 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 B is a Lifshitz transition and that the FS topology changes from quasi 2D to 3D. Unfortunately, it is impossible to test the change of dimensionality by quantum oscillation measurements, because of the heavy mass of the quasi-particles. *Hunt et al.* reported only a small change in the dHvA-frequency and mass of the light bands from 171 T, 4.62 m_e inside the A-phase to 162 T, 5.15 m_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 $transition²⁶$.

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 do be 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 didnt 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. Fig. 1a) 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 3 dimensions²¹ and the quadratic term typical for (non-critical) Landau-Fermi liquid contributions. While this observation is only a first hint for quantum critical behavior occurring close to this newly discovered 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 laws²¹. This approach has been applied i.e. to classify the zero pressure QCP in $CeNi₂Ge₂$ as 3-dimensional QCP of AFM-SDW type²⁷ with same $\Delta L/L \sim T^{3/2}$ critical contribution to the length change as observed in this new field-induced QCP in $CeCu₂Si₂$. 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 CeCu₂Si₂ 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 new fieldinduced 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 $H \geq H_c$. 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.

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