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Investigation of magnetic and magnetoelastic properties of the unconventional heavy-fermion compound $CeCu_2Ge_2$

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Abstract
Heavy-fermion compounds, high-T_c cuprates or iron pnictides are characterized by adjacent properties and a rich magnetic phase diagram with unconventional or competing states at low temperatures. $CeCu_2Ge_2$, the counterpart of the heavy-fermion superconductor $CeCu_2Si_2$, exhibits an incommensurate antiferromagnetically long-range ordered ground state with $\tau_1 = (0.28\ 0.28\ 0.54)$ up to $T_N = 4.1$ K which is strongly affected by a screening of the Ce 4f-moments by conduction electrons expressed by a Kondo temperature of $T_K = (\ 6–10)$ K.

The similar energy scales of the magnetic exchange and the Kondo effect result in a complex magnetic phase diagram with the possibility of quantum criticality at very low temperatures. The magnetic properties of $CeCu_2Ge_2$ were studied by magnetization measurements and, using the strong magnetoelastic coupling, by magnetostrictive investigations over a wide temperature range and in fields up to 50 T. Especially, the magnetostriction curves reveal clearly the details of the temperature and field dependence of the phase transitions. Here we present experimental results used for a refinement of the $H-T$ phase diagram in [100] and [110] direction.

Keywords: Heavy fermions, magnetization, magnetostriction, $H-T$ phase diagram, $CeCu_2Ge_2$

1 Introduction

The relation between local Kondo screening and magnetic exchange interactions determines the ground-state behaviour of lanthanide-based heavy-fermion compounds. Influenced by the interplay of 4f-electrons and conduction electrons, magnetically ordered or paramagnetic ground

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states exist on one side, a normal metallic or superconducting character of electric transport occurs on the other. The electronic properties can be varied by suitable changes of chemical composition, by pressure or by magnetic field. By the latter one, the spatial orientation of magnetic moments and the magnetic fluctuations are changed remarkably leading to magnetic phase transitions which can be detected directly by magnetization measurements or indirectly by magnetostrictive investigations due to the existing strong magnetoelastic coupling.

Prominent examples of this scenario are the rare-earth based heavy-fermion $CeCu_2(Si,Ge)_2$ compounds. Samples of this isostructural tetragonal ($ThCr_2Si_2$ structure) series have been extensively studied during the last decades. At the beginning of this process, the superconducting character of a heavy-fermion compound was found at $CeCu_2Si_2$ for the first time [1]. Later on, superconductivity was verified for $CeCu_2Ge_2$ with maximum T_c of about 2 K, but due to the higher unit cell volume only under pressure (8–18 GPa). Additionally, deviations from Fermi liquid behaviour were concluded from resistivity and thermopower measurements [2, 3]. The coexistence of a heavy-fermion low-temperature peak in $\gamma(T)$ and long-range antiferromagnetic order in $CeCu_2Ge_2$ was shown by specific heat and magnetization data [4] with an ordering temperature of $T_N = 4.1$ K, slightly lower than the Kondo temperature $T_K = (6–10)$ K [5]. This fact demonstrates the intermediate character between competing local Kondo behaviour and long-range magnetic order.

More details about the electronic properties and the phases in magnetic field were analyzed in the past few years. The zero-field magnetic structure below T_N determined by neutron diffraction of poly-crystalline samples of $CeCu_2Ge_2$, published already in [5], is of incommensurate character with a propagation vector $\tau_1 = (0.28\ 0.28\ 0.54)$. Subsequent neutron diffraction experiments on single-crystalline $CeCu_2Ge_2$ [6] resulted in a sinusoidal spin-density wave. The propagation vector is proposed to shift slightly with decreasing temperature and lock-in below 1.5 K with (0.283 0.283 0.538) which is close to (2/7 2/7 7/13) [6].

A band structure calculation [7] was used to model the Fermi surface of $CeCu_2Ge_2$. The incommensurate magnetic structure can be explained by the nesting properties of the calculated Fermi surface taking into account an itinerant component of the 4f-moments of Ce^{3+} in addition to their local character.

High-field magnetization measurements were carried out at 1.3 K and discussed in [8]. Weak metamagnetic transitions in fields parallel to the [110] and [100] axes were observed at 7.8 T and 10.5 T, respectively, followed by a magnetic saturation at 27 T with an absolute moment $\mu_S \simeq (0.6\text{-}0.7)\ \mu_B$. This value is much smaller than 1.5 μ_B estimated from the crystal-field splitting of the ground state and demonstrates again the effective Kondo screening of the Ce 4f-moments. Another attempt to obtain a complete phase diagram was initiated extrapolating specific heat and resistivity features measured in moderate fields up to 14 T with a quadratic extrapolation to very high fields. This results in a saturation field of 35 T along [100] and 31 T along [001] [9]. Other authors [10] suggest a phase transition at about 8 T for fields in [110] direction. Their interpretation of the neutron data contains the idea of a field-tuned quantum critical phenomenon (QCP) at which quantum critical spin fluctuations dominate local Kondo fluctuations and induce a transition between an antiferromagnetic and a quantum magnetic instability phase. By another (inelastic) neutron scattering experiment, the single-ion properties of Ce^{3+} were studied [11]. The crystal field splitting was found to produce three doublets at 0, 17.0 and 18.0 meV.

A more general magnetic $H-T$ phase diagram was constructed by the authors of [12] using angle-dependent transport measurements. This phase diagram shows a number of new phases, especially a modification of the antiferromagnetic ground state at about 8 T and some intermediate phases at higher fields. Additionally, the reported suppression of thermal fluctuations at very low temperatures leads to interesting aspects of quantum criticality. All these experiments give a deep insight into special properties of $CeCu_2Ge_2$.

It is the aim of this paper to use existing and new data derived from thermodynamic investigations (magnetostriction, magnetization) in order to complete the phase diagram for the main [100] and [110] crystallographic directions.

2 Samples and experimental methods

The single crystalline $CeCu_2Ge_2$ samples used in the experiments were grown from Cu flux using a modified Bridgman technique, compare [13]. The crystal used for macroscopic experiments was a 3.0 x 3.0 x 2.5 mm^3 cuboid with a mass of 171 mg. The crystal quality was proved by x-ray and neutron Laue scattering. The structural properties and lattice constants are already given in the publications above.

The macroscopic properties of the $CeCu_2Ge_2$ samples were studied at first by magnetization measurements in the temperature range of 1.2–300 K and in magnetic fields up to 14 T using an Oxford Instruments vibrating sample magnetometer. Additionally, the magnetic ac-susceptibility was measured down to 0.4 K. The magnetic measurements, *i.e.* measurements of electronic properties, were accompanied by magnetostrictive investigations detecting lattice effects caused by a strong magneto-elastic coupling. Both methods, magnetization and magnetostriction often are complementarily in order to find anomalies. The striction experiments were performed in a high-resolution capacitive dilatometer [14] up to fields of 15 T. This field range was extended up to 50 T afterwards in a pulsed field magnet at Helmholtz-Zentrum Dresden-Rossendorf. To avoid the influence of mechanical vibrations, these measurements were done by the optical FBG method [15] providing a resolution of 10^{-6} in relative length changes.

3 Magnetization and magnetostriction: Results

In order to construct the magnetic phase diagram in [100] direction from thermodynamic data it is instructive to compare the field dependencies of magnetostriction and magnetization at several temperatures (Fig. 1) and of the temperature dependencies of both properties at several fields (Fig. 2).

The left part of Fig. 1 shows the forced magnetostriction for different temperatures between 1.5 K and 10 K. For 1.5 K, 1.7 K and 2 K two distinct transitions for fields near 8 T and at 11 T are well pronounced. The transition at the lower field is characterized by a sample expansion due to fluctuation effects. It is followed by a maximum and then by a strong shrinking of the sample. Beyond the second transition at 11 T the sample expands again. For 2.7 K both transitions seem to merge into one transition. The effects due to the above mentioned transitions are only very week in the field dependent magnetization curves (right part of Fig. 1) . The curve at 1.7 K shows a tiny step at 8 T with some small hysteresis. Some of the curves measured at temperatures below 2 K have one more small magnetization anomaly at 11.5 T, but it is only to be seen in the derivatives.

The left part of Fig. 2 shows the thermal expansion curve at zero field and two longitudinal magnetostriction curves at 3 T and 6 T (i.e. magnetostriction measurements parallel to the field direction). The transition temperatures and fields were again determined from anomalies, here kinks or bends, which could be analyzed in detail by the derivative of the curves with respect to temperature. The temperature dependence of the sample length along [100] presents only one critical temperature at around $T_N \simeq 4.3$ K in the zero field curve. This point stays

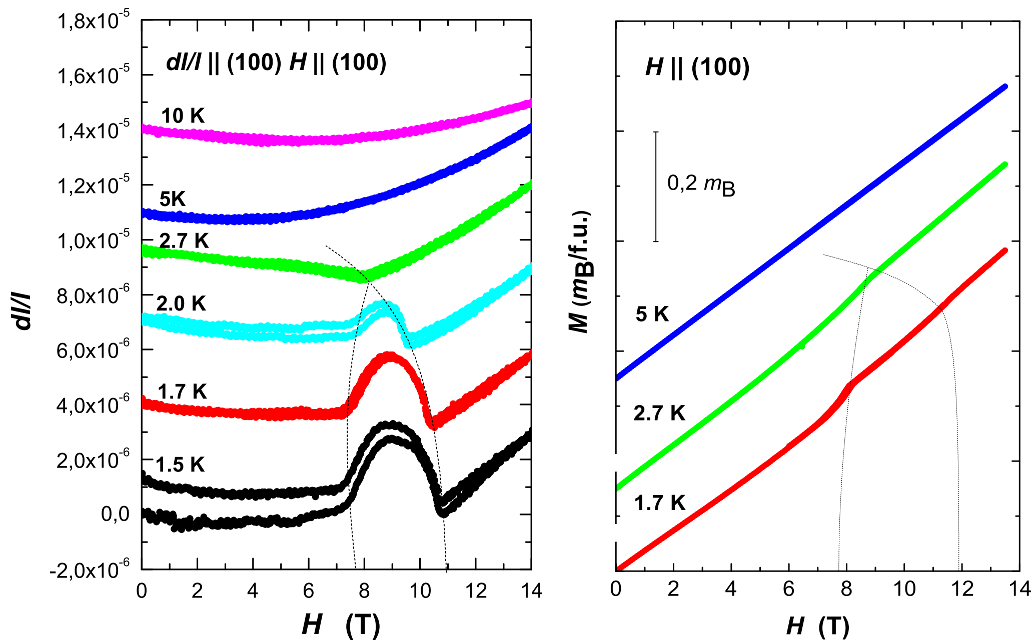

Figure 1: Field dependency of the isothermal longitudinal forced magnetostriction (left) and of the magnetization (right) of single-crystalline $CeCu_2Ge_2$ for magnetic fields parallel to the crystallographic [100] axis. Phase transitions are indicated by dotted lines.

stable at about 4 K for the curves at 3 T and 6 T. However, a second distinct kink develops which shifts to lower temperatures for increasing fields. Below 2 K a strong downturn is visible in all curves presumably related to an artefact of the set-up.

The results agree well to those of the magnetization measurements as shown in the right part of Fig. 2. The low-field curves below 1 T (not shown here) are characterized by a single sharp peak at the (anitferromagnetic) ordering temperature $T_N = 4.3$ K. The strong increase around this point suggests enhanced fluctuations. At higher fields above 5 T, the peak becomes more broadened with a significant internal structure and clearly splits into a double-peak structure at higher fields. The peak at the lower temperatures decreases to even lower temperatures for increasing fields. Above 10 T this decrease in temperature gets smaller and the peak vanishes at about 13 T.

The [100] saturation field is not accessible by moderate fields and had to be determined by isothermal high pulsed field magnetostriction measurements up to 50 T at temperatures between 2 K and 40 K. The low-temperature saturation field at 2 K in [100] direction was found at about 26 T and extrapolated to 27 T for 0 K. Again, the high field magnetostriction curves are characterized by anomaly features at low fields of about 9 T, approximately, decreasing in field with increasing temperature as found before by investigations of spontaneous and forced magnetostriction in steady fields.

Additional curves were measured at the $CeCu_2Ge_2$ single crystal in [110] and [001] oriented

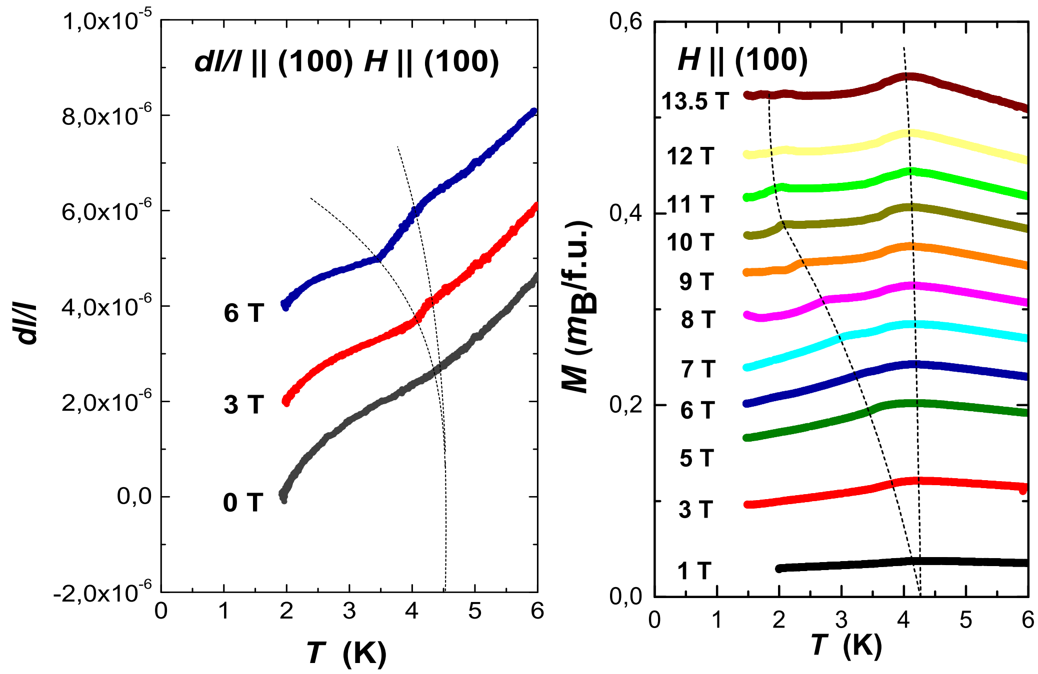

Figure 2: Temperature dependency of the spontaneous longitudinal magnetostriction (left) and of the magnetization (right) of single-crystalline $CeCu_2Ge_2$ for magnetic fields parallel to the crystallographic [100] axis. Phase transitions are indicated by dotted lines.

fields (not shown here).The curves along the [110] direction resemble those for the [100] direction. Only the transition fields differ slightly between both field direction and the absolute value of 10^{-5} of the contraction around 8 T is a bit larger in [110] direction. This may indicate some evidence for a small anisotropy (calculations point to an anisotropy of the two-ion interaction) in the basal plane.

The data measured in fields along the [001] direction do not show any anomaly up to a saturation field of 15 T. In contrast to the behaviour in the basal plane, the forced magnetostriction in fields parallel to [001] has a continuous decrease in length up to the highest available field of 15 T and the sample seems to have no tendency to saturate. This experimental fact is attributed to a steady-going rotation of the moments into the [001] direction.

All of the identified effects amount to less than 10^{-5} in relative changes of the sample length so that the crystallographic structure is unchanged in field and the magnetic structures can probably be considered as variations of the zero field structure.

4 Magnetic phase diagrams for [100] and [110] directions

The experimental data concluded from measurements of magnetization and magnetostriction together with the information from other publications enable us to construct a more specified

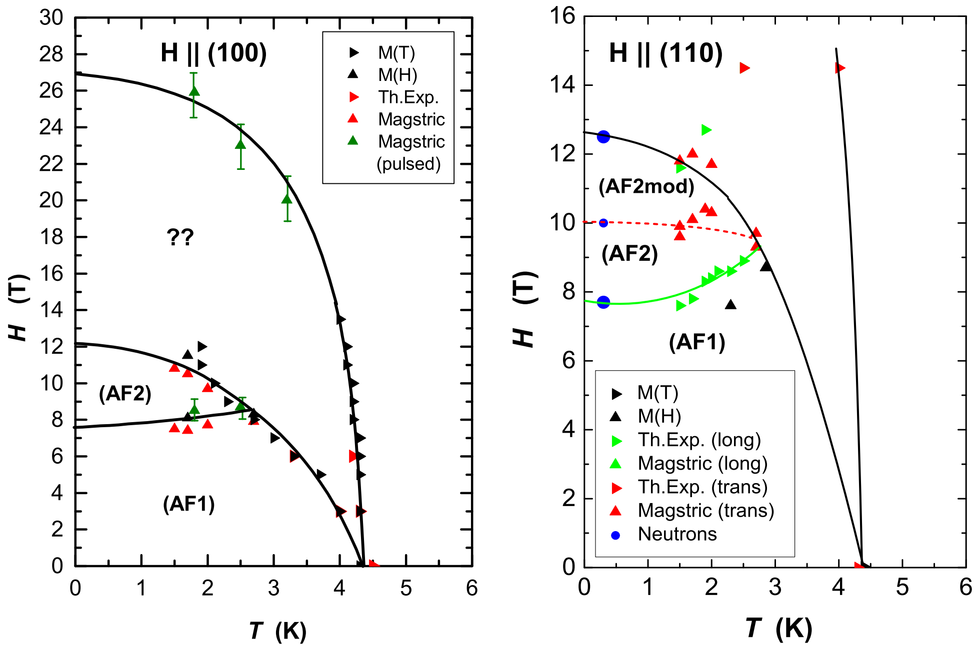

Figure 3: Magnetic phase diagram of $CeCu_2Ge_2$ as a function of applied magnetic field along [100] (left) and [110] (right). The data points are taken from magnetization and magnetostriction as marked in the figure (transversal: sampleA, longitudinal: sampleB). The three data points at low temperature in the phase diagram in [110] direction are taken from neutron diffraction presented in Ref. [16].

magnetic phase diagram of $CeCu_2Ge_2$ for magnetic fields applied in the main directions of the basal plane as presented in Fig. 3). The transition lines were constructed with respect to an uniform character of the detected anomalies in the relevant measurements. The phase diagram contains the incommensurate antiferromagnetic phase (AF1) with $\tau_1 = (0.28\ 0.28\ 0.54)$ below the ordering temperature $T_N = 4.3$ K. This structure undergoes a lock-in transition at $T_L \cong 1.5$ K with h = k = 0.283 or 2/7 and with l = 0.538 or 7/13 [6]. These findings are confirmed by our neutron diffraction experiments at low temperature and expanded to finite magnetic fields up to 13 T applied in [110] direction [16].

The marginal differences between both, [100] and [110] phase diagrams (compare the slightly different region of the existence of the AF2 phase) hint to a very small or even absent anisotropy in the basal plane as already mentioned above. The different properties of the AF1 and AF2 phases are mainly due to a modification of the propagation vector of the spin-density wave at the transition at 7.8 T from τ_1 to $\tau_2 = (0.31\ 0.31\ 0.54)$ [16]. This may be attributed to a change of the Fermi nesting properties. The transition itself is of first order as observed from the coexistence of both, AF1 and AF2 phases at 7.8 T (see corresponding figure in [16]). The gapped spin wave excitations seem to soften slightly at the 7.8 T transition but recover again for further increasing fields [17]. Transition fields of $B_1 = 7.8$ T, $B_2 = 10$ T and $B_3 = 12.6$ T in [110] direction have been identified from our neutron diffraction experiments [16] and added in the right-hand phase diagram of Fig. 3.

At higher fields of (12-13) T the long-range ordered AF2 phase vanishes and the extended range of a yet unknown (marked: "??") phase is opened. Our neutron study of this unconventional "cross-over behaviour" reveals a vanishing of the antiferromagnetic Bragg peaks related to propagation vectors of type (hhl). This may either be interpreted (i) as a possible total lack of long-range magnetic order accompanied by a stepwise increase of the magnetic moment component in field direction and strongly fluctuating moments perpendicular to it due to a modified Kondo screening of the Ce^{3+} moments or (ii) as a long-range magnetic structure with a yet unidentified propagation vector. The latter explanation is more favoured in the light of our field dependent inelastic neutron scattering experiments [16]. If or how in addition lattice instabilities which were detected in the field range directly above the phase boundary around (12-13) T influence the formation of the unknown phase via magnetoelastic coupling is still under consideration. Finally, $CeCu_2Ge_2$ comes to (induced) saturation at an in-plane field of about 26 T with a moment $\mu_S \simeq 0.7\ \mu_B$ much lower than the saturation moment 2.5 μ_B of the free Ce^{3+} ion or 1.5 μ_B of the Ce^{3+} crystal field ground state doublet.

5 Conclusions

In summary, we have mapped out the magnetic phase diagram of $CeCu_2Ge_2$ for magnetic field directions along [100] and [110] in the basal plane and perpendicular, *i.e.* along [001]. The basal plane anisotropy is confirmed to be rather small or even absent. The results suggest that the antiferromagnetic order with the known propagation vectors τ_1 and τ_2 survives up to 12-13 T followed by a yet unidentified phase. This open question has to be solved by further microscopic investigations. Finally, the induced ferromagnetism is realized at about 26 T. Attempting to model the magnetic properties within a meanfield approach clearly demonstrate the need to include the Kondo effect in order to reproduce the experimental susceptibility, magnetization and transition fields.

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