

Field dependence of the magnetic propagation vector of the heavy fermion compound CeCu_2Ge_2 studied by neutron diffraction

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Abstract

CeCu_2Ge_2 , the counterpart of the heavy-fermion superconductor CeCu_2Si_2 , exhibits an incommensurate antiferromagnetically long-range ordered ground state with $\tau = (0.28 \ 0.28 \ 0.54)$ below $T_N = 4.15$ K. The magnetism is strongly affected by a Kondo screening of the Ce 4f-moments by conduction electrons. The similar energy scale of both, Kondo and exchange interactions, results in a complex magnetic phase diagram and gives rise to potential quantum critical phenomena at very low temperatures. We present elastic neutron diffraction data obtained on a CeCu_2Ge_2 single crystal employing the cold triple axis spectrometer PANDA at MLZ and the diffractometer D23 at ILL.

The field dependence of the magnetic propagation vector was measured at $T \leq 400$ mK in the $[110]/[001]$ plane with vertical magnetic fields applied along $[1\bar{1}0]$. We observe a low-field incommensurate magnetic phase AF1, a first order phase transition around 7.8 T with the coexistence of two phases AF1 and AF2 with slightly different propagation vectors, the disappearance of AF1 at 8 T and the existence of AF2 up to 12 T with a possible modification at 10 T. At 12.6 T, yet still well below the value of 26 T of the saturation for magnetic fields in $[110]$ direction, the AF2-type magnetic order is lost and magnetic intensities are not to be found at incommensurate positions in the $[110]/[001]$ plane any more. These new results contradict a previously suggested scenario with a QCP located at 8 T and contribute new information to the $B - T$ phase diagram of CeCu_2Ge_2 in $[110]$ direction.

Keywords: Heavy fermions, magnetic ordering, neutron diffraction, CeCu_2Ge_2

1 Introduction

CeCu₂Ge₂ is a heavy fermion system crystallizing into the ThCr₂Si₂-type structure (space group I4/mmm). Below $T_N = 4.15$ K, antiferromagnetic order with Kondo partly compensated moments is established with an incommensurate propagation vector of $\tau_1 = (0.283 \ 0.283 \ 0.538)$ at $T = 40$ mK [1]. It shares several properties with the archetypical heavy fermion superconductor CeCu₂Si₂: Both exist in the ThCr₂Si₂-type structure, the energy scales of RKKY interaction and Kondo temperature $T_K \approx (5-10)$ K [2] are in the same range yet with CeCu₂Ge₂ lying more on the localized moment side while in CeCu₂Si₂ the Kondo compensation dominates the moment ordering that sets in only at much lower temperatures. The similarities of both compounds were already exploited by Si/Ge substitution and pressure studies [3] with the aim to understand the physical origin of heavy fermion superconductivity. Recently, the superconductivity in CeCu₂Si₂ could be unambiguously linked to an antiferromagnetic quantum critical point (QCP) [4]. It is known for other heavy fermion antiferromagnets, notably YbRh₂Si₂ [5] and CeCu_{6-x}Au_x ($x \geq 0.1$) [6], that also a magnetic field can be used as a control parameter to tune the system towards a QCP. This motivated us to carry out a neutron scattering study on the influence of a magnetic field on the magnetic correlations in CeCu₂Ge₂. Here, we present elastic neutron scattering data in a magnetic field up to 12.6 T at low temperature, which indicates the presence of multiple phase transitions. We will then compare these data to previously reported thermodynamic measurements and discuss the implications on quantum criticality.

Finally we also want to refer to results obtained on single crystalline samples of CeCu₂Ge₂ for the magnetic phase diagrams in [100] and [110] directions determined from thermodynamic properties [7] and to the magnetic field dependent neutron spectroscopy of the low-energy spin wave excitations [8].

2 Experimental Methods

We used the CeCu₂Ge₂ single crystalline sample, which was already employed in Ref. [9] in connection with inelastic neutron experiments to determine the crystal field level schemes. We oriented the crystal in the plane spanned by [110]/[001] applying the magnetic field in [1 $\bar{1}$ 0] direction.

Elastic neutron scattering experiments were performed at the PANDA cold triple-axis spectrometer at the Heinz Maier-Leibnitz Zentrum and the D23 thermal diffractometer at the Institut Laue-Langevin. The setup used at PANDA was $\lambda = 4.8$ Å with PG (002) monochromator and analyzer and with a cooled Be filter to suppress $\lambda/2$ contributions. On D23 $\lambda = 2.4$ Å with PG (002) monochromator and PG filter was used.

At both instruments the sample was cooled using a dilution insert in a vertical cryomagnet, hence the magnetic field was applied along the [1 $\bar{1}$ 0] direction. Cooling to base temperature was done without applied field and then the sample was kept at $T \leq 300$ mK for both experiments.

Elastic rocking scans (sample rotation at fixed scattering angle) were performed at PANDA covering the surrounding of the magnetic Bragg peak at (H H L) with $H = 0.28$ and $L = 1.45$ that corresponds to a propagation vector (h h l) with $h = H$ and $l = 2-L$. At D23, the evolution of the same magnetic Bragg peak with magnetic field was followed performing scans over H and L, respectively. To check for field induced changes in the nuclear structure or magnetic contributions on the positions of the nuclear Bragg peaks, rocking scans over the (004), (006), (110) and (222) nuclear Bragg peaks were performed. These rocking scans for each magnetic field were fitted using one Gaussian, except for the (110) peak, where two Gaussians were used to account for the presence of a small grain. In a similar way, the H and L scans over the

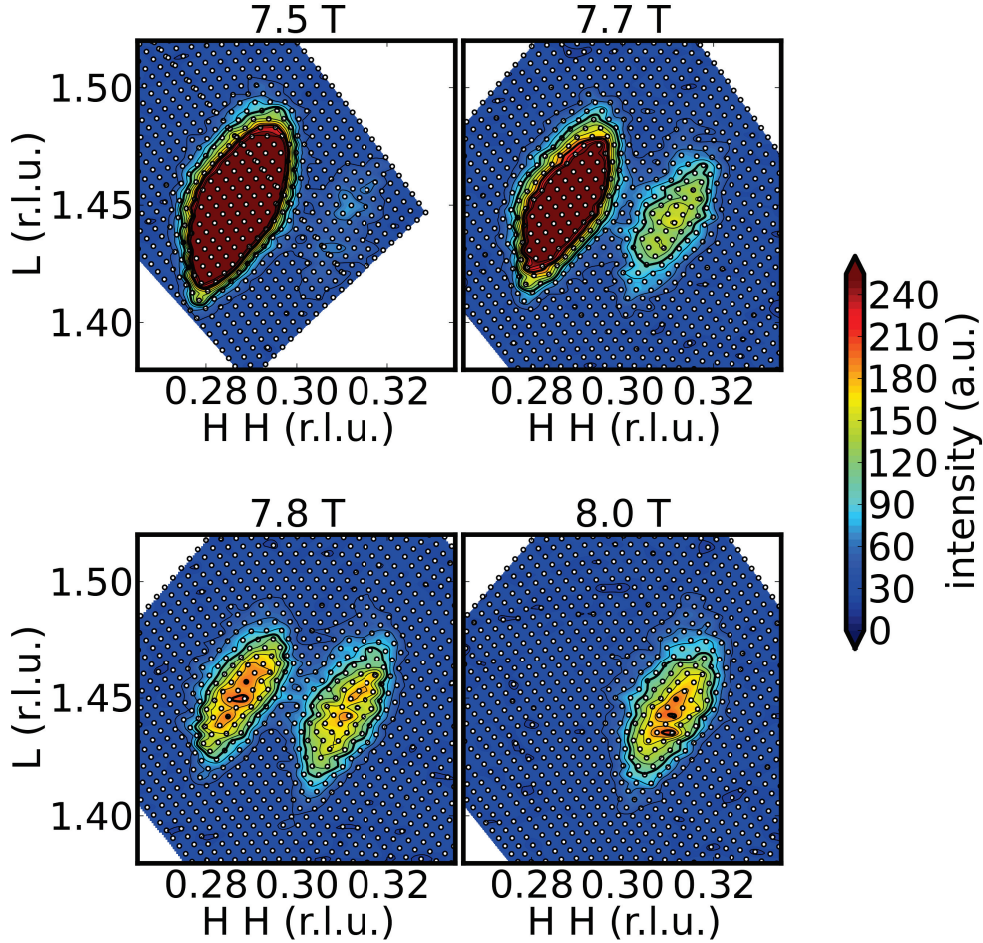


Figure 1: Magnetic field dependence of the magnetic Bragg peak at $(H\ H\ L)$ in a narrow field range around the phase transition at B_1 . Data taken on PANDA.

magnetic Bragg peak were fitted with one Gaussian. At 12.6 T, rocking scans were performed over a large part of the Q -space to search for further magnetic order.

3 Results for phases AF1 and AF2

In Fig. 1 intensity maps taken on PANDA around the $(H\ H\ L)$ magnetic Bragg peak are shown in the narrow field region of the phase transition at $B_1 = 7.8$ T. This has to be compared with the results of our D23 experiment displayed in Fig. 2 covering the full field range from zero field to 12.6 T. From the PANDA data of Fig. 1 it is evident that at 7.5 T the sample is in the AF1

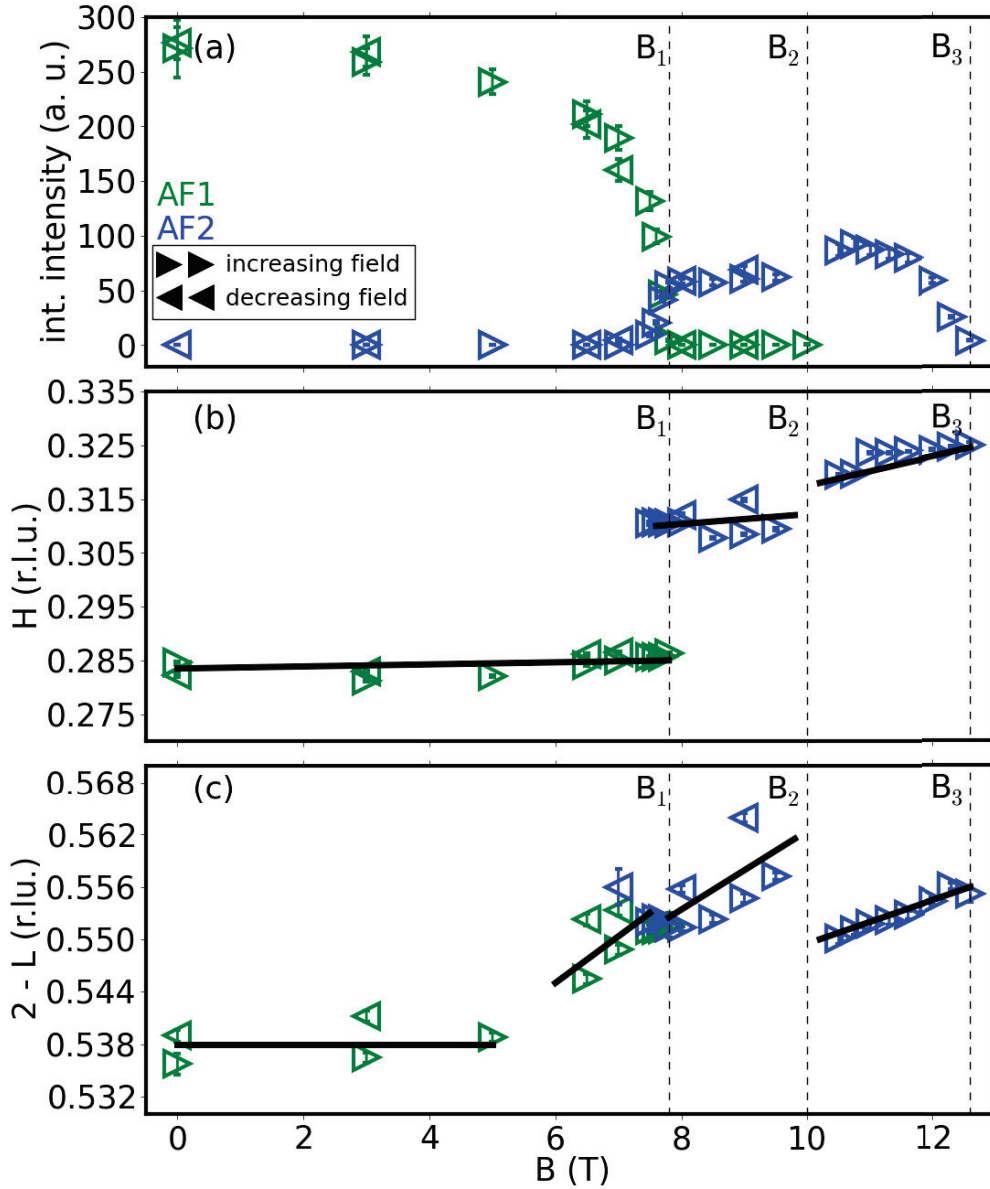


Figure 2: Magnetic field dependence of the $(H\ H\ L)$ magnetic Bragg peak of the AF1 phase and AF2 phase. Indicated are also the positions of the magnetic transition fields B_1 , B_2 and B_3 . The region between B_1 and B_3 may be divided into a low-field region with AF2 and a region with a modified AF2 from B_2 and B_3 . Data taken on D23. The solid lines are guides for the eye.

phase and at 8 T it is in the AF2 phase with a slightly modified incommensurate propagation vector. In the field region between these two magnetic fields both phases coexist, indicative of a first order phase transition. The complete determination of both magnetic structures (moment direction, squaring-up, single-q, multiple-q etc.) requires further experiments. However, we have some evidence that the moments in the low-field phase AF1 point along $[1\bar{1}0]$ direction. This is in contrast to the expected anisotropy due to the crystalline electric field and has therefore to be due to an anisotropic exchange interaction.

In Fig. 2 we present the evolution with magnetic field in the whole examined field range of the integrated intensities of the (H H L) magnetic Bragg peak as well as the $h = H$ and $l = L/2$ component of the propagation vectors determined from the D23 experiment. The h -component of the propagation vector is constant for AF1 for all fields up to $B_1 = 7.8$ T ($h = 0.285$), then jumps to the other value for AF2 ($h = 0.310$) and stays again constant up to $B_2 = 10$ T. This is followed by another minor jump above B_2 and a slight increase between B_2 and B_3 . The behaviour of the l -component of the propagation vector is different: in the AF1 phase it is constant up to about 6 T ($l = 0.538$), then starts to increase to $l = 0.55$ (D23, Fig. 2) and has about the same value for the AF1 and AF2 phase in the field region of the first order phase transition at B_1 (compare Fig. 1: the PANDA data seem to indicate a tiny increase of the l value when going from AF1 to AF2). In the AF2 phase the l -component further increases with field up to B_2 , it then jumps to lower values at B_2 followed by another increase up to B_3 . The behaviour around $B_2 = 10$ T may indicate the possible presence of another phase transition into a slightly modified AF2 phase. Similar observations are made when comparing the longitudinal and transversal magnetostriction for fields in $[110]$ direction to map out the corresponding phase diagram with macroscopic measurements [7].

4 Results at 12.6 T

At 12 T the integrated intensity related to the incommensurate structure AF2 starts to decline and at $B_3 = 12.6$ T it vanishes completely. Fig. 3 shows the intensity map at 12.6 T covering the entire Brillouin zone in the $[1\ 1\ 0]/[0\ 0\ 1]$ scattering plane. One can see that no sign of magnetic satellites are recognizable at 12.6 T. This demonstrates that above B_3 there is no magnetic order of type AF1 or AF2. Thus the nature of the phase beyond 12.6 T is still a puzzle.

5 Results on the nuclear Bragg peaks

The magnetic field dependence of the integrated intensity and sample rotation ω of the nuclear Bragg peaks is shown in Fig. 4. Compared to the uncertainty of the measurement, a small but systematic increase in the integrated intensity with magnetic field is observed for all four investigated nuclear Bragg peaks. This increase is compatible with a ferromagnetic component of $0.35\ \mu_B$ in agreement with the magnetization data.

An approximately linear shift in ω occurs from zero field up until $B_1 = 7.8$ T. From there on, up to the highest magnetic field at 12.6 T, ω stays constant. For (004) as well as (006) a jump in ω is present at B_1 while for (110) and (222) one observes a kink. The change in rotation angle is practically the same in value and direction for all four nuclear Bragg peaks in the AF1 phase and may therefore be an artefact of the experiment (rotation of the sample rod due to magnetic forces?). However, there is no sample rotation in the AF2 phase. At the moment, it is not possible to give an unambiguous explanation of this behaviour.

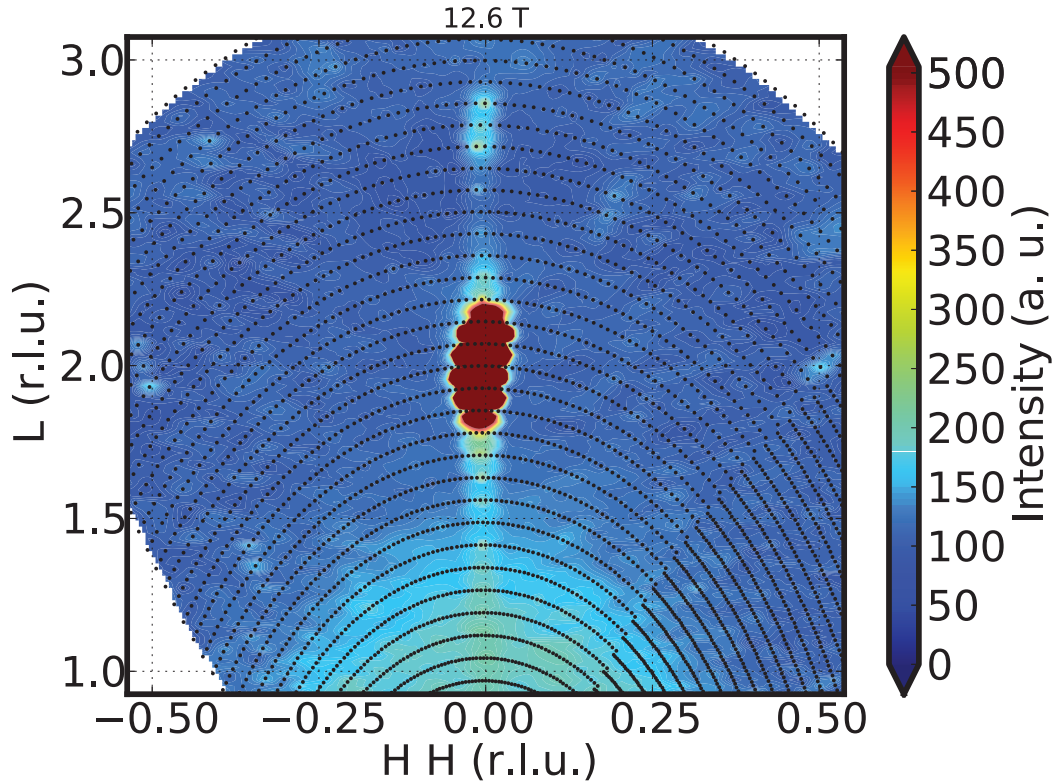


Figure 3: Intensity map at 12.6 T obtained from rocking scans. Note: Since only rocking scans were performed, small deviations from the $[1\ 1\ 0]/[0\ 0\ 1]$ scattering plane occurred, however these are smaller than the resolution for the used setup. The very weak intensity at $(0\ 0\ 2.78)$ does not correspond to magnetic order as it is of nuclear origin. Data taken on D23.

6 Discussion

The most obvious reasons for the phase transitions at B_1 and B_3 , first a spin-flop transition at B_1 and then saturation of the magnetization above B_2 , can be ruled out. A spin-flop transition would not explain the change of the propagation vector at B_1 . For saturation of the magnetization, a magnetic field much larger than B_2 would be required [7], [10]. Also the additional magnetic intensity on the positions of the nuclear Bragg peaks that is expected for a saturation moment of $0.7\ \mu_B$ is not present.

The presence of a QCP has been suggested at 8 T [11] close to B_1 from the temperature scaling of the dynamic susceptibility measured by inelastic neutron scattering. A fundamental property of a QCP either of the Hertz-Millis-Moriya (HMM) type [12, 13, 14] or the local type [15] is that it destroys the magnetic order. This is not the case at B_1 , since the magnetic order is not destroyed but undergoes a first-order phase transition from the AF1 into the AF2 phase. Therefore, the presence of a QCP at B_1 can be ruled out. Since the state above B_3 is most likely disordered, a QCP might be located at B_3 . However, from the elastic neutron scattering data presented here, no further statements can be made about this possibility.

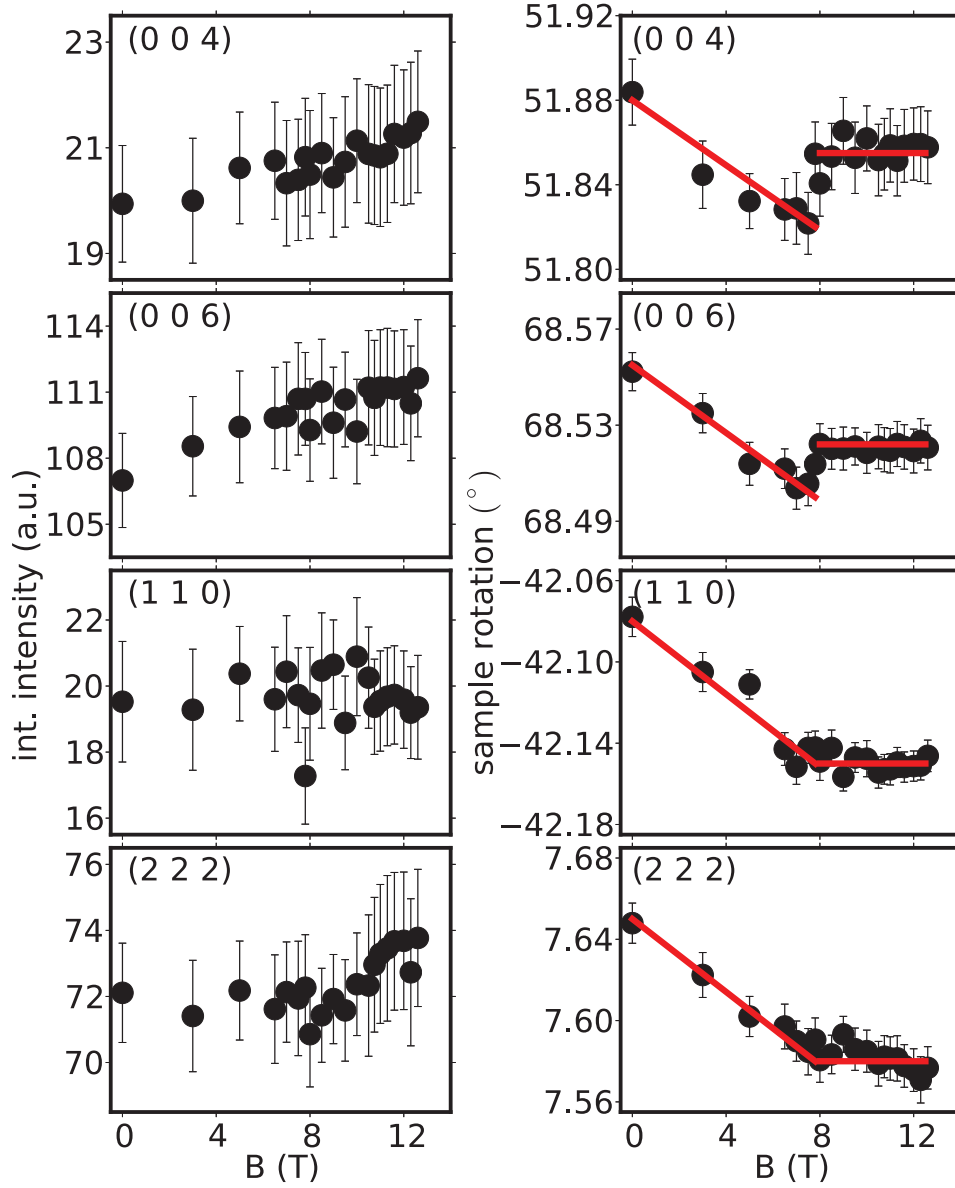


Figure 4: Magnetic field dependence of the (004), (006), (110) and (222) nuclear Bragg peaks measured at D23. Left: integrated intensities, right: sample rotation angles.

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