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Low-energy magnetic excitations of $CeCu_2Ge_2$ investigated by inelastic neutron scattering

P Geselbracht[1], E Faulhaber[1], M Rotter[2], K Schmalzl[3], D Quintero-Castro[4], O Stockert[5], M Loewenhaupt[6], and A Schneidewind[7]

[1] Heinz Maier-Leibnitz Zentrum (MLZ), Technische Universität München, Garching, Germany
[2] McPhase Project, Dresden, Germany
[3] Jülich Centre for Neutron Science at ILL, Forschungszentrum Jülich GmbH, Grenoble, France
[4] Helmholtz-Zentrum Berlin, Berlin, Germany
[5] Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Germany
[6] Institut für Festkörperphysik, Technische Universität Dresden, Dresden, Germany
[7] Jülich Centre for Neutron Science at MLZ, Forschungszentrum Jülich GmbH, Garching, Germany

Abstract

$CeCu_2Ge_2$, the magnetic counterpart of the heavy-fermion superconductor $CeCu_2Si_2$, exhibits an antiferromagnetic ground state below $T_N = 4.15$ K with an incommensurate propagation vector of $\tau = (0.28\ 0.28\ 0.54)$. The magnetism is determined by local Ce 4f-moments and the ordering RKKY interaction as well as the onset of Kondo screening by conduction electrons. Tuning the energy scale of the Kondo effect and RKKY interaction towards enhanced Kondo screening may result in quantum critical phenomena at low temperature. While the existence of quantum critical phenomena induced by external pressure or chemical substitution is well known, the situation is less clear for magnetic field tuning. We will present an investigation of the spin excitation spectrum in magnetic field by inelastic neutron scattering and compare it to mean-field (McPhase) simulations of the antiferromagnetic spin wave excitation spectrum. We argue that our data can be described by the presence of spin wave excitations.

Keywords: Heavy fermion, Inelastic neutron scattering, mean-field calculations, McPhase, $CeCu_2Ge_2$

1 Introduction

The heavy-fermion compound $CeCu_2Ge_2$ crystallizes in the $ThCr_2Si_2$-type structure (space group I4/mmm). Below $T_N =$ 4.15 K long range antiferromagnetic order is established. The propagation vector of $\tau_1 = (0.284\ 0.284\ 0.543)$ for $T = 1.5$ K [1] is determined by the nesting properties of the Fermi surface [2]. As this propagation vector is close to (2/7 2/7 7/13), a lock-in transition is suggested which is observed at $T_L = 1.5$ K. The $4f^1$ (J = 5/2) state of the Ce^{3+} ions is split by crystal electric field (CEF) into one ground-state doublet and doublets at higher energy (> 10 meV) [3, 4, 5, 6]. The formation of magnetic order is determined by

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the interplay of the CEF split $4f^1$ states, the long range ordering RKKY interaction and the onset of Kondo screening by conduction electrons. As a result, if the exchange interaction that controls the strength of RKKY interaction and Kondo effect is tuned by either external pressure or Si substitution, a quantum critical point (QCP) occurs surrounded by a superconducting dome [7, 8]. It is known for other heavy fermion antiferromagnets, notably $YbRh_2Si_2$ [9] and $CeCu_{6-x}Au_x$ [10], that also a magnetic field can be used as a control parameter to tune the system towards a QCP. Such a possibility for $CeCu_2Ge_2$ was first investigated using inelastic neutron scattering (INS), where a QCP was claimed to exist at 8 T for $B \parallel [1\bar{1}0]$ [11, 12]. This interpretation, however, becomes dubious when including new data on the field dependence of the propagation vector [13]. It was found that a first order transition occurs between 7.7 T and 7.9 T where the propagation changes from $\tau_1 = (0.285\ 0.285\ \ 0.538)$ to $\tau_2 = (0.310\ 0.310\ 0.543)$, followed by another slight shift at around 10 T to $\tau_3 = (0.317\ 0.317\ 0.543)$. At 12.6 T magnetic satellites of the type (hhl) have vanished.

The magnetic behavior in all three main crystallographic directions was investigated by magnetic and magnetoelastic measurements and presented in [14]. The magnetic phase diagrams for fields in [100] and [110] directions differ only marginally exhibiting two phase transitions at 8 T and 12 T and a transition at 26 T into a magnetically saturated state. In contrast, the magnetoelastic properties for fields in [001] direction show only a continuous decrease in length for increasing fields which can be attributed to a steady rotation of the magnetic moments into the [001] direction with no indication of a phase transition. Zeng et al. [15] present a much more complicated structure of the phase diagram in [100] direction. However, the observations for the [001] direction are consistent in [14] and [15].

The aim of the present paper was to investigate the magnetic excitation spectra as a function of magnetic field at the lowest temperatures, well in the magnetically ordered state. The highest currently available magnetic field for INS is 15 T for vertical magnets where the field has to be applied perpendicular to the scattering plane. Additionally, mean field simulations were used to test to which extent linear response theory and random phase approximation can account for the observed excitation spectrum and are discussed in the last part of the present paper.

2 Experimental: Sample and inelastic neutron scattering

Inelastic neutron scattering (INS) was performed on the cold triple-axis spectrometers PANDA (Heinz Maier-Leibnitz Zentrum), V2 FLEXX (Helmholtz-Zentrum Berlin). The same $CeCu_2Ge_2$ single crystalline sample as in Ref. [6] was mounted at PANDA in a dilution insert in a vertical cryomagnet and on V2 FLEXX in a ^{3}He insert and a vertical cryomagnet. As a scattering plane spanned by [110]/[001] is needed to measure at the gamma point of the magnetic unit cell, the magnetic field direction results along $[1\bar{1}0]$ for a vertical cryomagnet. In both experiments the sample was cooled below $T_L = 1.5$ K before the field was ramped up. All INS spectra, were obtained in fixed $k_f = 1.3$ Å^{-1} mode, with PG (002) monochromator. To eliminate $\lambda/2$ contaminations, a cooled Be filter was used on PANDA and a velocity selector on V2 FLEXX. Both instruments and their sample environments were run under similar conditions to ensure compatibility of the obtained data. Typical measuring times were 10 min per data point for PANDA and V2 FLEXX. Both instruments show somewhat comparable energy resolution, but the nondispersive elastic line is approximately two times broader for V2 FLEXX (FWHM $< 60\ \mu$eV (PANDA), $< 120\ \mu$eV(V2 FLEXX)). The feature at 1.5 meV is less pronounced for V2 FLEXX most likely due to a lower Q-resolution. The dispersion presented in Fig. 2 was measured at IN12 (Institut Laue Langevin) in similar conditions without magnetic field [16].

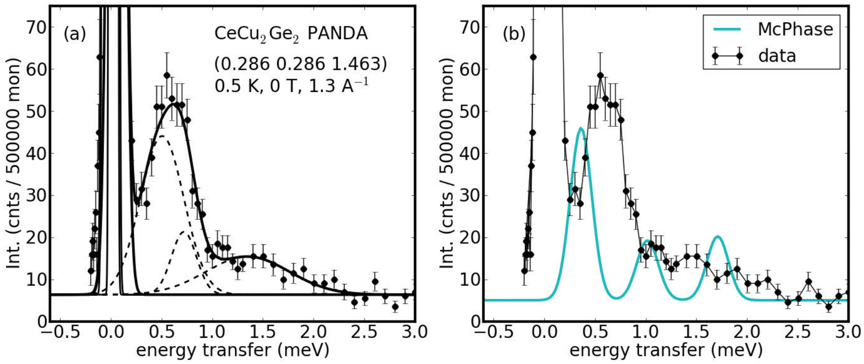

Figure 1: (a) Constant-Q spectra at the Γ-point of the magnetic unit cell for the low-field magnetic structure AF1 at zero field. The solid line is a fit using three Gaussians (dashed lines) for the inelastic part. (b) Same INS data as in (a) together with the McPhase calculation for the zero field spin wave spectrum.

3 Results and Discussion

3.1 INS spectra

The excitation spectrum at the Γ-point of the magnetic unit cell for the low-field magnetic structure AF1 in zero magnetic field (Fig. 1 (a)) was fitted using three Gaussian shaped contributions: a dominant excitation at 0.5 meV that nearly covers the smaller excitation at 0.75 meV and a much broader one at 1.4 meV.

All three excitations show dispersive behavior in zero magnetic field if measured along the [110] direction (Fig. 2). A detailed analysis of the dispersion will be published in a forthcoming paper [16], here we only refer to the results. Due to the observed dispersion we can consider these excitations as spin waves. The peak shapes are well described by Gaussian lines. For increasing magnetic fields, but still within the AF1 phase, the three-peaked structure of the spin wave excitations becomes more clearly visible as the mode in the center gains in intensity and the mode at lowest energy shifts towards lower energies (Fig. 3 (a) to (c)). There is also a rearrangement of integrated intensities of all three spin wave excitations.

Between 7.7 T and 7.9 T, a first order phase transition occurs, where the propagation vector changes from $\tau_1 = (0.285\ 0.285\ 0.538)$ to $\tau_2 = (0.310\ 0.310\ 0.543)$ with the coexistence of both phases at $B_1 = 7.8$ T [13]. The INS spectra at the Γ point of both magnetic phases are shown for this magnetic field in Fig. 4. In both spectra the intensity distribution can be fitted using three Gaussian shaped spin wave excitations as for lower fields. No further fluctuations emerge, as expected for a first order phase transition.

Upon further increasing the magnetic field we first reproduce partly the results of our neutron diffraction experiments [13] and of the magnetic phase diagram in [110] direction [14] in Fig. 5 (a) and (b), respectively.

In the AF2 phase between B_1 and B_2 and in the slightly modified phase AF2(mod) between B_2 and B_3 we present the excitation spectra at 10.5 T measured at V2 FLEXX in Fig. 5 (c) and at 11.5 T measured at PANDA in Fig. 3 (d), respectively. Fig. 5 (d) shows the V2 FLEXX spectra in a region where the intensity of magnetic Bragg related to the modified AF2 (mod) structure starts to decrease. Finally, for the high-field region beyond 12.6 T with yet unknown

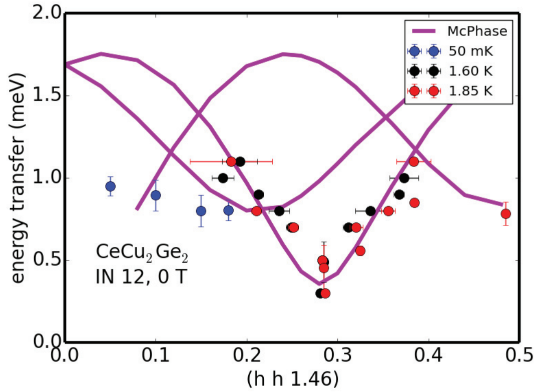

Figure 2: With McPhase for 1.85 K calculated spin wave dispersion (solid lines) compared to data obtained by INS experiments performed on IN12 (ILL) at comparable conditions as the spectra shown in this work, data at 50 mK, 1.60 K and 1.85 K [16].

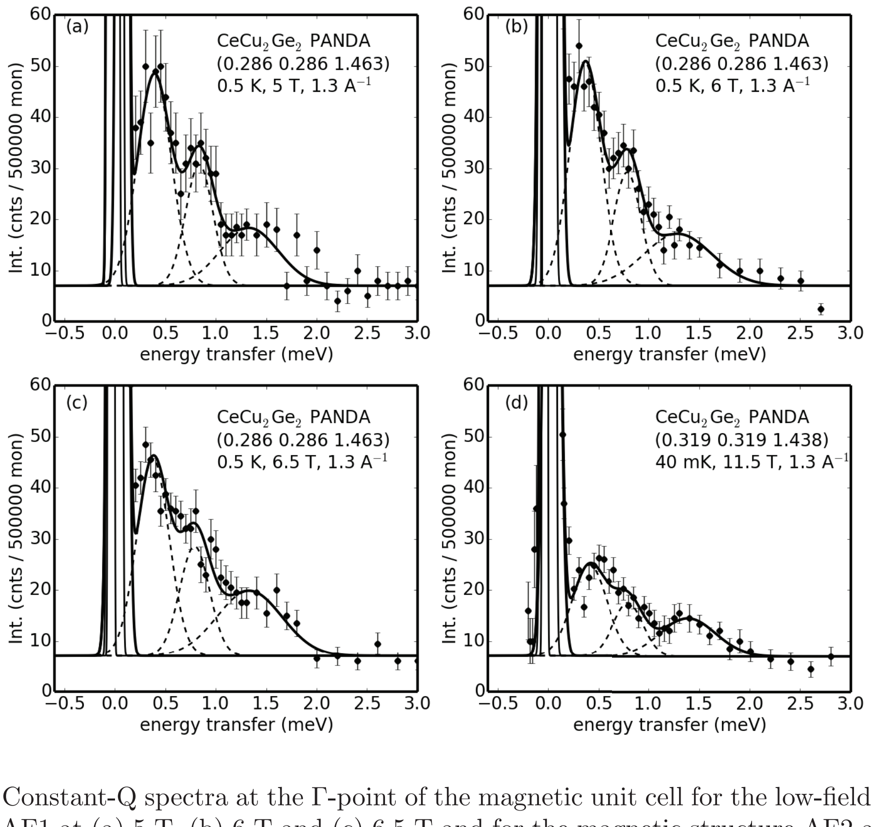

Figure 3: Constant-Q spectra at the Γ-point of the magnetic unit cell for the low-field magnetic structure AF1 at (a) 5 T, (b) 6 T and (c) 6.5 T and for the magnetic structure AF2 at (d) 11.5 T. Data taken on PANDA.

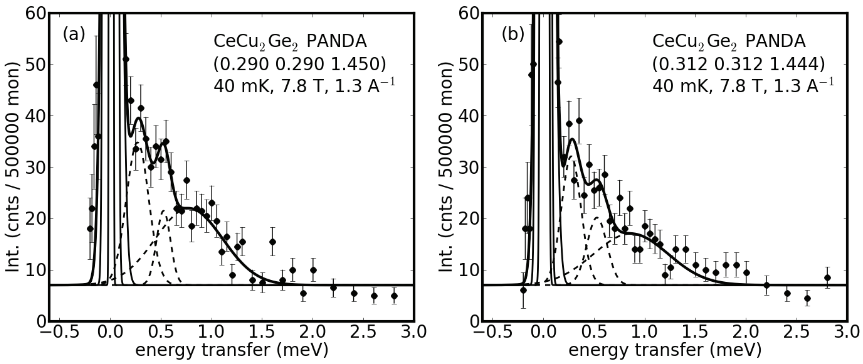

Figure 4: Constant-Q spectra for a magnetic field of 7.8 T where both magnetic structures coexist [13] mesured at the corresponding Γ-points of the magnetic unit cells (a) of the zero field structure AF1 at (0.29 0.29 1.45) and (b) of the new magnetic structure AF2 at (0.31 0.31 1.44). Data taken on PANDA.

magnetic phase we show the spectra at 13 T and 13.5 T in Fig. 5 (e) and (f), respectively. The gap of the spin wave excitations is (0.39 ± 0.07) meV for 10.5 T and increases to (0.49 ± 0.1) meV at 11.5 T. From there on it stays constant even above 12.6 T.

Due to the somewhat different conditions for resolution and intensity between PANDA and V2 FLEXX we have analyzed the spectra shown in Fig. 5 only with two Gaussians as in the V2 FLEXX spectra no sign of a well resolved third spin wave excitation could be identified. Also in the PANDA data at 11.5 T the centered mode is hardly resolvable as it is superposed by the edge of the spin wave at lowest energy.

3.2 Mean-field calculations using McPhase

Mean-field calculations were performed using the McPhase software package [17]. The treated Hamiltonian $\mathcal{H}$ is given by

$$\mathcal{H} = \sum_i B_l^m O_l^m(\mathbf{J}_i) - g_J \mu_B \mathbf{J}_i \mathbf{H} - \frac{1}{2} \sum_{ij} J_i^\alpha \mathcal{J}_{\alpha\beta}(ij) J_j^\beta \qquad (1)$$

The first term corresponds to the crystal field parametrised by B_2^0 = -10.26 K, B_4^0 = -0.056 K and B_4^4 = +2.67 K in Stevens notation [18] determined by INS [6], the second term corresponds to the Zeeman energy for the applied magnetic field $\mathbf{H} = \mu_0 \mathbf{B}$ with g_J being the Landé factor for the Ce^{3+} ion. In the third term, J_j^α (α = a, b, c) are the three components of the angular momentum operator of the jth Ce^{3+} ion. The tensor $\mathcal{J}_{\alpha\beta}(ij)$ includes the dipole interaction of the Ce^{3+} ion (in the range of 20 Å), a self interaction $J_{cc}(i, j = i)$ = -0.25 meV ($J_{aa}(i, j = i) = J_{bb}(i, j = i) = 0$) to account effectively for Kondo screening and the RKKY interaction as in Eq. 2.

$$J_{aa}(\mathbf{r}) = J_{bb}(\mathbf{r}) = A \frac{\sin(2\rho) - 2\rho\cos(2\rho)}{(2\rho)^4} \qquad (2)$$

with $\rho = \sqrt{(k_a r_a)^2 + (k_b r_b)^2 + (k_c r_c)^2}$ and the parameters A = -500 meV, $k_a = k_b$ = 0.9335 Å^{-1} and k_c = 1.9712 Å^{-1}. r_α with α = a, b, c are the components of the vector $\mathbf{r}$

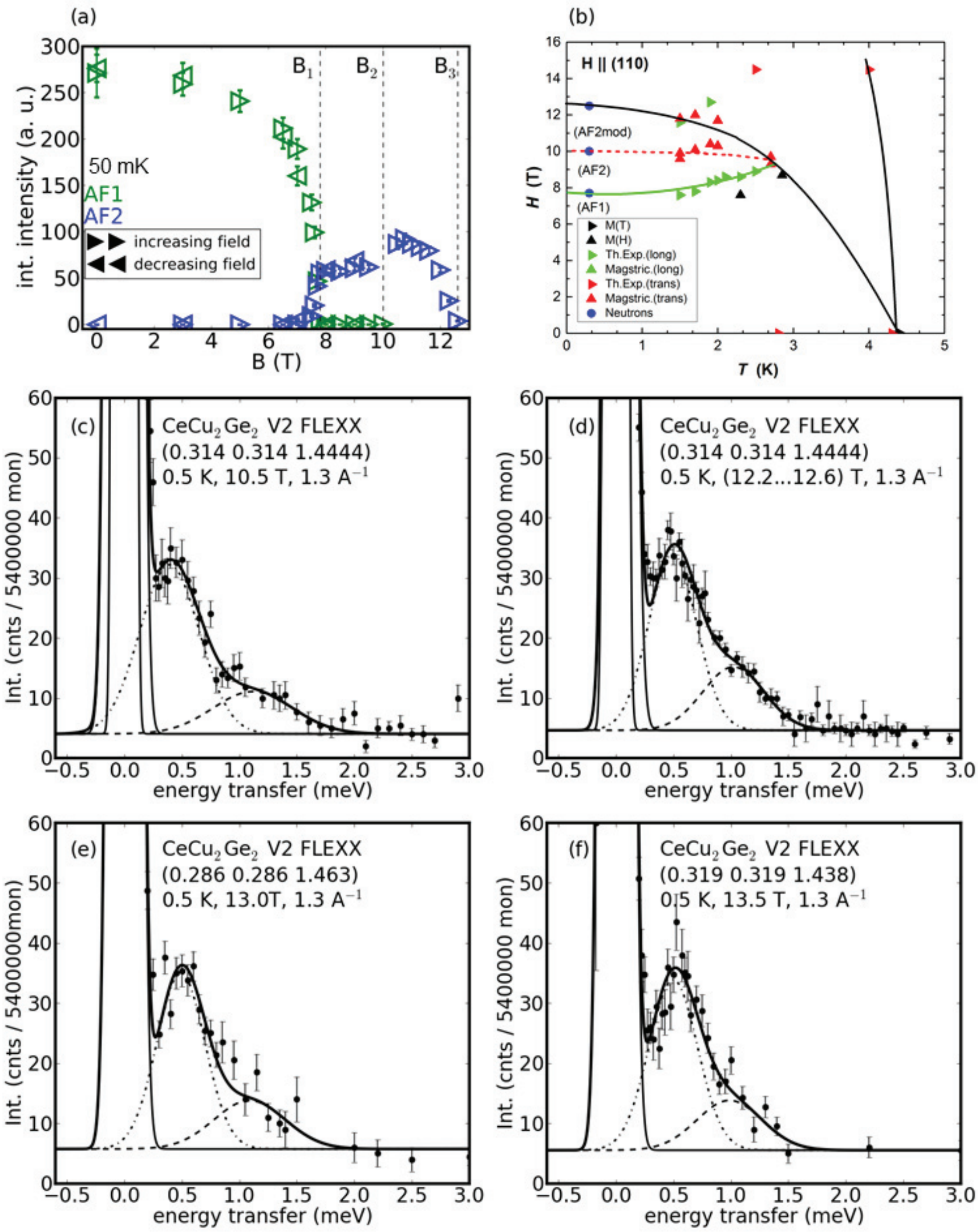

Figure 5: (a): Field dependence of the intensity of the magnetic Bragg peaks in the phases AF1 and AF2. Data are taken from Ref. [13]. (b): Magnetic phase diagram of $CeCu_2Ge_2$ in $[1\bar{1}0]$ direction from Ref. [14] (c) and (d): Constant-Q spectra at the Γ-point of the magnetic unit cell corresponding to the propagation vector of the phase AF2 in the field region between B_2 and B_3. In (d) data of spectra obtained at 12.2 T, 12.5 T and 12.6 T are integrated to achieve better statistics. (e) and (f): Constant-Q spectra for the yet unknown phases above 12 T at magnetic fields of 13.0 T and 13.5 T. Data taken V2 FLEXX as indicated in the figures.

connecting two ions. A maximum distance of 10.652 Å was considered and $J_{cc}(i \neq j) = 0$.

A super cell 7 x 7 x 9 (441 atoms) to maintain a propagation vector (hkl) = (0.286 0.286 0.54) was chosen to create a magnetic structure. As $J_{cc}(i \neq j) = 0$, the moments order non collinear in the basal plane. Magnetic excitations were then calculated from this structure using the dynamical matrix diagonalisation (DMD) methode of the *mcdisp* module for different Q-points and the obtained results were convoluted with a Gaussian with a full-width-half-maximum of 0.25 meV to enable comparison with the INS data.

The so obtained magnetic excitations spectrum for 0 T at the AF1 Bragg peak position (0.285 0.285 1.46) is shown in Fig. 1 (b) together with the INS data. The calculations yield a strong spin wave mode at low energy and and two less intense modes at 1 meV and 1.8 meV. The calculations reproduce a three peak feature, which is also found in the measured INS spectra at zero field: there are three spin wave modes with one strong mode at lowest energy and two less intense modes at higher energy. Note that the DMD formalism does not take into account lifetime effects which lead to increased line width for some modes. Such broadening can be clearly seen in the experimental data. Calculations of the spin wave dispersion in (hh1.46) direction for zero field are also consistent with the measured spin wave dispersion (Fig. 2) [16].

4 Conclusions

In summary, we investigated the magnetic excitation spectra of $CeCu_2Ge_2$ by INS and compared the measured spectra with mean field (McPhase) simulations of spin wave excitations. We found that the INS spectra over the whole accessible field range (0 T to 13.5 T) can be described by the presence of two or three spin wave modes.

The main features in the measured INS spectra could be qualitatively reproduced by the calculations. No signs of additional, possibly quantum critical, fluctuations were observed for 8 T. Also the existence of a QCP at 8 T as suggested in Refs. [11, 12] can be ruled out by the existence of the newly discovered magnetic order above 8 T [13].

The situation for the phase transition around 12 T is less obvious as no further magnetic order could be detected above 12.6 T [13]. The intensity corresponding to the AF2 phase vanishes almost linearly with increasing field as seen in Fig. 5 (a) starting at 12.0 T. This might qualify the transition at 12.6 T for a (quantum) fluctuation driven phase transition. On the other hand, Ref. [15] reported for the corresponding transition in [100] field direction a first order transition. By analogy with La doped $CeRu_2Si_2$ [19] and $CeCu_2Si_2$ [20] one may expect that quantum criticality manifest itself in a quasielastic response with a FWHM of 0.2 meV. Although this is larger than the energy resolution of the experimental data, such a contribution to the excitation spectrum, if present, might be covered under under the spin wave excitations. Therefore, for the transition at 12.6 T we have to leave the question for quantum criticality unanswered.

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