Nature of the *A* Phase in $CeCu_2Si_2$

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Neutron diffraction experiments have been performed on a magnetically ordered $CeCu_2Si_2$ single crystal exhibiting *A*-phase anomalies in specific heat and thermal expansion. Below $T_N \approx 0.8$ K antiferromagnetic superstructure peaks have been detected. The propagation vector of the magnetic order appears to be determined by the topology of the Fermi surface of heavy quasiparticles as indicated by renormalized band-structure calculations. The observation of long-range incommensurate antiferromagnetic order as the nature of the *A* phase in $CeCu_2Si_2$ suggests that a spin-density-wave instability is the origin of the quantum critical point in $CeCu_2Si_2$.

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The heavy-fermion metal $CeCu_2Si_2$ is still of considerable interest for both theorists and experimentalists due to its intriguing low temperature properties. Since the observation of superconductivity in $CeCu_2Si_2$ in 1979 [1], much work has been devoted to understand the unusual properties of this prototypical heavy-fermion superconductor. For $CeCu_2Si_2$ an unusual type of magnetic order, the so-called "*A* phase" [2], discovered about ten years after the superconductivity by NMR [3] and muon spin rotation (μSR) [4], has attracted much interest. Subsequent thermodynamic and transport measurements revealed a complex magnetic (B, T) phase diagram with different superconducting and magnetically ordered phases and a ground state depending very delicately on the actual stoichiometry [2]. The ground state can be either *A* phase, sometimes coexisting with superconductivity, A/S where superconductivity expels the *A* phase, or only *S*(uperconducting) [5]. Doping experiments with Ge substituting the Si as well as experiments under hydrostatic pressure strongly suggest that $CeCu_2Si_2$ is located very close to a quantum critical point connected with the disappearance of the *A* phase. Non-Fermi-liquid behavior in the vicinity of the quantum critical point is observed, e.g., in the specific heat and the electrical resistivity [6]. For an understanding of this non-Fermi-liquid behavior at the quantum critical point in $CeCu_2Si_2$, it is of utmost importance to unravel the nature of the *A* phase. Although several attempts were made to detect magnetic order by neutron diffraction, all previous experiments failed in observing magnetic scattering intensity. However, clear signatures pointing to a spin-density wave (SDW) as origin of the *A* phase in $CeCu_2Si_2$ were found in measurements of the electrical resistivity, showing the opening of a gap below the ordering temperature in certain directions [6], or in μSR experiments, already giving a rough estimation of the ordered moment of $\sim 0.1\mu_B$ [4].

Another possibility to gain more insight into the nature of the *A* phase in $CeCu_2Si_2$ is to investigate the alloyed system $CeCu_2(Si_{1-x}Ge_x)_2$ [7,8]. In particular, one can follow the magnetic order as a function of Ge doping. The advantage of the doped system lies in the fact that the ordering temperature is higher and the ordered moment enhanced. First neutron diffraction experiments were performed on $CeCu_2Ge_2$ [9] and $CeCu_2(Si_{1-x}Ge_x)_2$ [7,10] powder samples and a $CeCu_2Ge_2$ single crystal [11]. They revealed an incommensurate antiferromagnetic order with a propagation vector $\tau \approx (0.28\ 0.28\ 0.53)$ for $CeCu_2Ge_2$ which is only weakly dependent on the Ge concentration. The magnetic moments of the sine-modulated structure are confined to the [110] plane [11]. However, no magnetic intensity was detected for Ge concentrations $x < 0.6$. Later, single crystal neutron diffraction on $CeCu_2(Si_{1-x}Ge_x)_2$ with $0.05 \leq x \leq 0.45$ succeeded in observing the antiferromagnetic order [12]. The propagation vector remains roughly constant, i.e., in the vicinity of $\tau = (0.25\ 0.25\ 0.5)$, but the ordered moment is strongly reduced compared to the alloys with higher Ge content.

Recently, we were able to grow large single crystals of $CeCu_2Si_2$ with well-defined properties [13]. Taking advantage of the new knowledge on the magnetic structure in the Ge-doped system, we started a new attempt to solve the difficult, but fundamental question about the magnetic origin of the *A* phase by performing magnetic neutron diffraction measurements on these large single crystals.

The experiments were performed on an *A*-phase $CeCu_2Si_2$ single crystal. The single crystal was grown in an Al_2O_3 crucible by a modified Bridgman technique using Cu excess as flux medium. The largest crystal, used for the present study, has the dimensions $3 \times 4 \times 4$ mm^3 ($m = 350$ mg) and was oriented by x-ray

Laue backscattering. X-ray powder diffraction confirmed the tetragonal $ThCr_2Si_2$ structure with the lattice parameters $a = 4.102$ Å and $c = 9.930$ Å at room temperature. However, additional energy dispersive x-ray spectroscopy on the $CeCu_2Si_2$ single crystal yielded a small amount of aluminium impurities ($\approx 0.5\%$) in the sample. This might be caused by a diffusion of aluminum from the crucible into the melt during the growth process. In order to verify that the main findings of our study are not influenced by Al impurities, a second single crystal was investigated. This second crystal, grown under different conditions, did not show any impurities.

The first single crystal was characterized by specific heat and thermal expansion measurements in the temperature range $T = 0.4$ K to 3 K and 80 mK to 3 K, respectively. A quasiadiabatic heat-pulse method was utilized for the specific heat measurements and a magnetic field could be applied along the crystallographic c axis. The thermal expansion was measured with a high-resolution capacitive dilatometer. The neutron diffraction experiment was performed on the two-axis diffractometer E6 at the HMI Berlin with a neutron wavelength $\lambda = 2.45$ Å in the temperature range between $T = 50$ mK and 1 K using a dilution refrigerator. The E6 diffractometer, equipped with a position sensitive detector covering a range of 20° in scattering angle 2Θ, allows one to record intensity maps of the reciprocal scattering plane around interesting reciprocal lattice vectors. As a consequence of the previous neutron diffraction experiments on $CeCu_2(Si_{1-x}Ge_x)_2$, the reciprocal $(h\,h\,l)$ plane was chosen as scattering plane. Scans were always performed in angle space (sample rotation ω and scattering angle 2Θ) and data were then transformed to the reciprocal space $(h\,h\,l)$ during analysis. The second crystal was only measured on the diffractometer E4 at the HMI Berlin in the same scattering plane with a neutron wavelength $\lambda = 2.436$ Å at temperatures $T = 0.5$ K and 1 K using a ^{3}He cryostat.

The thermodynamic properties of the $CeCu_2Si_2$ single crystal will be presented first. The specific heat plotted as C/T versus T in Fig. 1(a) exhibits a transition into the ordered state at $T_N = 0.86$ K which is lowered to slightly above 0.6 K in a magnetic field of $B = 2$ T applied along the c axis. The shape and size of the anomaly as well as the magnetic field dependence point to a transition into the magnetically ordered A phase [5]. In contrast, a superconducting transition would have been completely suppressed already in a magnetic field of $B = 2$ T [5]. A large jumplike anomaly in the coefficient α of the thermal expansion at T_N marks the onset of the A phase via a second order transition confirming the specific heat results [cf. Figure 1(b)]. In addition, a further transition of first order as indicated by the pronounced hysteresis is observed at a lower temperature $T \approx 0.3$ K as displayed in the inset of Fig. 1(b).

In our neutron diffraction experiment on $CeCu_2Si_2$ on E6 intensity maps of the reciprocal $(h\,h\,l)$ plane around

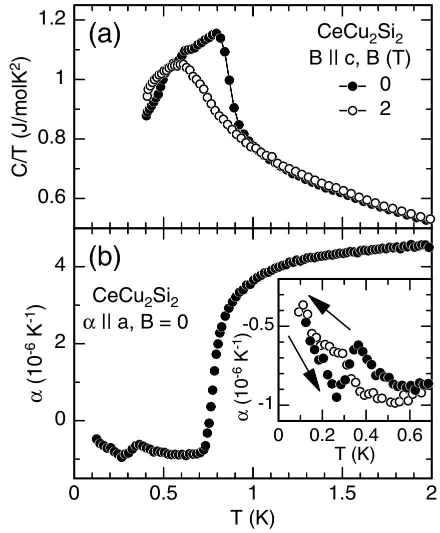

FIG. 1. (a) Specific heat plotted as C/T versus temperature T of the $CeCu_2Si_2$ single crystal in zero magnetic field and $B = 2$ T applied along the c axis. (b) Temperature dependence of the thermal expansion coefficient α along the a axis in $CeCu_2Si_2$. The inset shows the hysteretic behavior of α at low temperatures.

$q = (0.21\ 0.21\ 1.45)$ were recorded at the lowest temperature $T = 50$ mK and above the ordering temperature at $T = 1$ K as shown in Fig. 2(a). While at 1 K only a q-dependent background is detected, increasing towards lower momentum transfer q, a well resolved magnetic superstructure peak is visible at 50 mK. This becomes even more apparent after integration of the data over scattering angle 2Θ and then displaying the data as a function of sample rotation ω as in Fig. 2(b). Magnetic peaks were also found at symmetry equivalent positions. However, due to their low intensity on a high background and the resulting long counting times of ≈ 11 min per point, only a few have been measured. At the lowest temperature, the propagation vector has been determined from the positions of the magnetic peaks with respect to nuclear peaks yielding $\tau = (0.215\ 0.215\ 0.530)$. Because of the experimental setup on E6, the instrumental resolution in ω scans is quite relaxed and no information about the correlation length can be deduced. However, cuts along 2Θ through the data, where the resolution is better, do not show any broadening of the magnetic peak compared with the experimental resolution within the errors. To further clarify this point and to check the propagation vector, the second crystal was measured on E4 with higher resolution in ω. As displayed in Fig. 2(c), the magnetic peak, observed at $q = (0.21\ 0.21\ 1.46)$ at $T = 0.5$ K while absent at $T = 1$ K, confirms the propagation vector τ and yields a width in ω of the magnetic peak given by the instrumental resolution. Therefore it is concluded that the antiferromagnetic order is long range in nature. The ordered moment is estimated from a comparison to the Ge-doped system which has a magnetic structure with a sinusoidal modulation of the magnetic

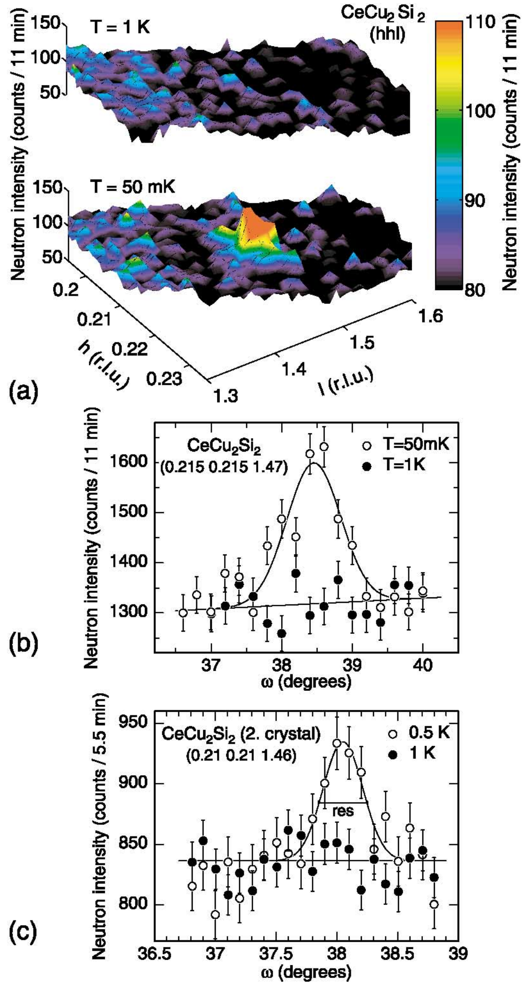

FIG. 2 (color). (a) Intensity map of the reciprocal $(h\,h\,l)$ plane around $q = (0.21\,0.21\,1.45)$ in $CeCu_2Si_2$ at $T = 50$ mK and 1 K. (b) ω scan across the magnetic peak at $q = (0.215\ 0.215\ 1.47)$ at 50 mK and 1 K [data shown in (a) and (b) taken on E6]. (c) ω scan across the magnetic peak at roughly the same wave vector q as in (b) at 0.5 K and 1 K for the second crystal measured on E4 ("res" denotes the instrumental resolution).

moments lying in the basal plane, roughly perpendicular to the propagation vector. Assuming the same structure yields an effective ordered moment $\sim 0.1\mu_B$ for $CeCu_2Si_2$ [14]. It is worth noting that in the non-Fermi-liquid compound $CeNi_2Ge_2$, having the same crystal structure as $CeCu_2Si_2$, high-energy spin fluctuations with a characteristic energy of 4 meV have been found at the incommensurate wave vector $q = (0.23\ 0.23\ 0.5)$ [15], i.e., at roughly the same wave vector the long-range order in $CeCu_2Si_2$ is observed. However, these spin fluctuations in $CeNi_2Ge_2$ do not show any critical slowing down as the temperature is lowered $T \to 0$.

The antiferromagnetic order in $CeCu_2Si_2$ results from an instability of the Fermi surface. This is inferred from Fig. 3(a) which displays the calculated static magnetic susceptibility $\chi_0(q)$ of noninteracting quasiparticles as a function of the wave vector q. $\chi_0(q)$ exhibits a pronounced maximum at the position of the observed propagation vector τ, which suggests that the normal Fermi liquid may become unstable with respect to the formation of a spin-density wave.

The magnetic susceptibility $\chi_0(q)$ was evaluated adopting the Lindhard formula. The quasiparticle energies were determined from the renormalized band method [16]. This method which is essentially a one-parameter theory reproduces the Fermi surfaces and the highly anisotropic effective masses of heavy-fermion compounds quite well [17]. The ansatz starts from a standard *ab initio* band-structure calculation for the weakly correlated conduction states. The strong local correlations are included by choosing the f-phase shifts at the Ce sites according to

$$\tilde{\eta}_{fm}(E) = \arctan\frac{\tilde{\Gamma}_f}{E - \tilde{\epsilon}_{fm}},$$

where the index m refers to the eigenstate $|m\rangle$ of the crystalline electric field (CEF) split $J = 5/2$ spin-orbit multiplet. The corresponding energies $\tilde{\epsilon}_{fm} = \tilde{\epsilon}_f + \Delta_m$, the band centers, are separated by an excitation energy Δ_m from the centers corresponding to the CEF ground state. The states $|m\rangle$ as well as the energies Δ_m are taken from experiment. The renormalized width $\tilde{\Gamma}_f = k_B T^*$ of the quasiparticle band is adjusted to reproduce a C/T value of $\approx 0.7\ \mathrm{J/(mol\,K^2)}$ for $T \to 0$ [18]. The structure in $\chi_0(q)$

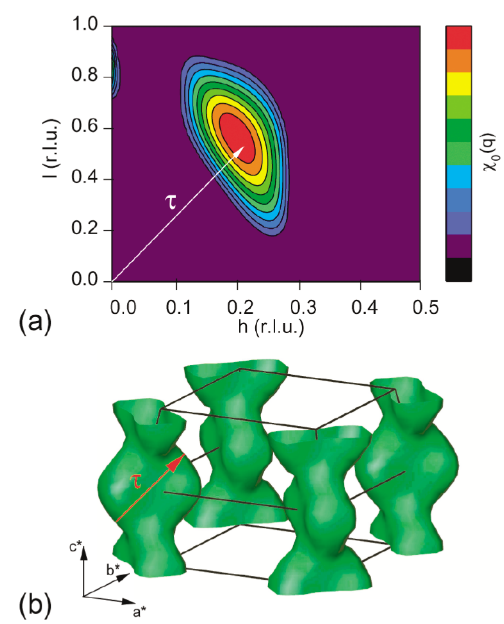

FIG. 3 (color). (a) Comparison of the measured propagation vector τ with theoretical intensity map for the wave vector dependent magnetic susceptibility $\chi_0(q)$ of noninteracting quasiparticles in the reciprocal $(h\,h\,l)$ plane of $CeCu_2Si_2$. The calculations are performed at $T = 100$ mK using the following parameters to characterize the low-energy excitations: $T^* \sim 10$ K, $\Delta_{CEF} = 330$ K. The contribution of the incoherent background has been subtracted. (b) Fermi surface of the heavy quasiparticles in $CeCu_2Si_2$ as calculated with the renormalized band method. The vector τ connects parallel flat parts ("nesting") of the Fermi surface.

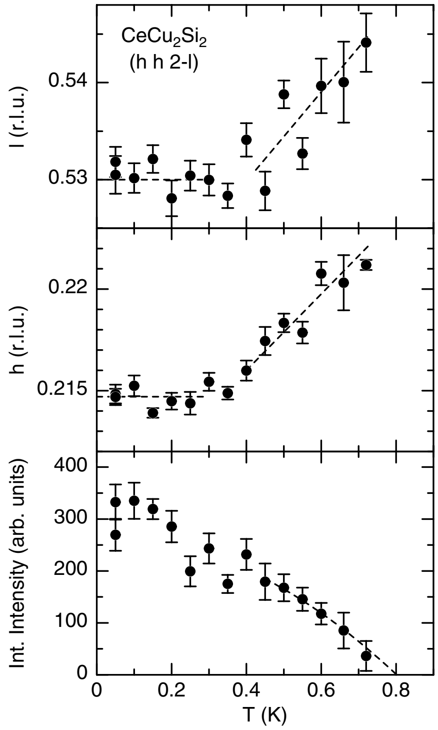

FIG. 4. Temperature dependence of the components of the propagation vector $\tau = (h\,h\,l)$ and of the intensity of the magnetic $(h\,h\,2\text{-}l)$ Bragg peak in $CeCu_2Si_2$ (measured on E6). Dashed lines are only guides to the eye.

does not depend sensitively upon the specific value. Moreover, the position $\tilde{\epsilon}_f$ is eliminated by imposing the condition that the charge distribution is not significantly altered by introducing the renormalization as compared to the local-density approximation (LDA) result.

In the calculations leading to Fig. 3, the CEF scheme given in [19] was adopted, yielding a value $T^* \simeq 10$ K for the characteristic temperature. The results for the Fermi surface are in qualitative agreement to [16]. There are two bands intersecting the Fermi energy. For the thermodynamically most relevant sheet, displayed in Fig. 3(b), heavy quasiparticles with effective masses $m^* \sim 500\,m_e$, m_e being the bare electron mass, are found. This heavy Fermi surface consists mainly of columns parallel to the tetragonal c^* axis. The quasiparticles on this surface have predominantly $4f$ character as reflected in the large masses. It should be mentioned that the topology of this surface depends sensitively on the detailed symmetry of the CEF states. It is not surprising that it differs fundamentally from the predictions of standard LDA calculations which treat the $4f$ electrons as normal band states [20].

As a final point, the temperature dependence of the magnetic intensity and of the propagation vector τ, shown in Fig. 4, will be discussed. The results are obtained by analyzing the intensity and position of the magnetic Bragg peak shown in Figs. 2(a) and 2(b) being a satellite of the (002) nuclear peak. The magnetic intensity vanishes at $T_N \approx 0.8$ K in line with the results of the thermodynamic measurements. No magnetic intensity has been observed for $T \geq 0.8$ K in the reciprocal $(h\,h\,l)$ plane. The propagation vector τ shows an unusual behavior. With lowering the temperature below the Néel temperature T_N, the propagation vector $\tau = (h\,h\,l)$ is temperature dependent and becomes smaller, with the components h and l decreasing roughly linearly with T. Furthermore, τ exhibits a kink at $T \approx 0.3$–0.35 K and remains constant at lower temperatures. This lock-in transition into a probably commensurate low temperature phase is identified as a first order transition in the thermal expansion measurement [cf. inset of Fig. 1(b)].

In conclusion, long-range antiferromagnetic order has been identified as the nature of the A phase in $CeCu_2Si_2$. The observed instability of the Fermi liquid is related to the fact that the Fermi surface as calculated exhibits parallel flat parts separated by the measured propagation vector. These results suggest that a spin-density-wave instability is the origin of the quantum critical point observed in $CeCu_2Si_2$. However, the discrepancy between the small ordered moment and the observed large anomalies in the thermal expansion as well as the elastic constants [2] calls for further examination.

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[1] F. Steglich *et al.*, Phys. Rev. Lett. **43**, 1892 (1979).
[2] G. Bruls *et al.*, Phys. Rev. Lett. **72**, 1754 (1994).
[3] H. Nakamura *et al.*, J. Magn. Magn. Mater. **76&77**, 517 (1988).
[4] Y. J. Uemura *et al.*, Phys. Rev. B **39**, 4726 (1989).
[5] F. Steglich *et al.*, in *More is Different–Fifty Years of Condensed Matter Physics* (Princeton University Press, Princeton, 2001), p. 191.
[6] P. Gegenwart *et al.*, Phys. Rev. Lett. **81**, 1501 (1998).
[7] G. Knebel *et al.*, Phys. Rev. B **53**, 11 586 (1996).
[8] O. Trovarelli *et al.*, Phys. Rev. B **56**, 678 (1997).
[9] G. Knopp *et al.*, Z. Phys. B **77**, 95 (1989).
[10] A. Krimmel and A. Loidl, Physica (Amsterdam) **234B–236B**, 877 (1997).
[11] A. Krimmel *et al.*, Phys. Rev. B **55**, 6416 (1997).
[12] O. Stockert *et al.*, Acta Phys. Pol. B **34**, 963 (2003).
[13] H. S. Jeevan (unpublished).
[14] A helical structure, which cannot be completely ruled out, would result in an ordered moment larger by a factor of $\sqrt{2}$.
[15] B. Fåk *et al.*, J. Phys. Condens. Matter **12**, 5423 (2000).
[16] G. Zwicknagl and U. Pulst, Physica (Amsterdam) **186B–188B**, 895 (1993).
[17] G. Zwicknagl, Adv. Phys. **41**, 203 (1992).
[18] F. Steglich, Phys. Scr. T **29**, 15 (1989).
[19] E. A. Goremychkin and R. Osborn, Phys. Rev. B **47**, 14 280 (1993).
[20] A. Yaresko (unpublished).