Nature of the Magnetic Order in $CeCu_2Si_2$ and Interplay with Superconductivity

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The interplay of superconductivity and antiferromagnetic order continues to be of general interest in condensed matter physics. While the coexistence of antiferromagnetism and superconductivity can easily be understood if different electronic subsystems are involved in the two phenomena, the situation might be different if the same electron system is responsible for both. In the case of the heavy-fermion compound $CeCu_2Si_2$ the Ce-4*f* electrons do not only order magnetically, but are also involved in the formation of superconducting Cooper pairs [1,2].

The heavy-fermion compound $CeCu_2Si_2$ forms only in a very narrow range of the ternary phase diagram Ce:Cu:Si around the 1:2:2 composition. However, the ground state in $CeCu_2Si_2$ depends very delicately on the actual composition. Thus, Cu-rich samples exhibit only superconductivity (S-type) and Si-rich only show a magnetically ordered phase, called A-phase, while in stoichiometric samples (A/S-type) a complex interaction between superconductivity and magnetic order occurs [2]. Hydrostatic pressure experiments as well as experiments substituting Si by Ge strongly suggest that $CeCu_2Si_2$ is located very close to a quantum phase transition at the disappearance of the A phase [3,4]. During the last few years the crystal growth technique improved tremendously allowing to grow large $CeCu_2Si_2$ single crystals with well defined properties. This enabled us not only to measure thermodynamic and transport properties, but also to perform neutron diffraction to investigate magnetism on a microscopic level.

First experiments were carried out on an A-phase crystal [5] which orders magnetically below $T_N \approx$ 850 mK as evidenced by anomalies in the specific heat and the thermal expansion. The neutron diffraction experiments were performed on the diffractometer E6 at the HMI/Berlin in the temperature range between T = 50 mK and 1 K using a dilution refrigerator. The advantage of the E6 diffractometer is the existence of a position-sensitive detector covering a range of 20° in scattering angle 2Θ. This allows to record intensity maps of the reciprocal scattering plane around interesting reciprocal lattice vectors. Fig. 1(a) displays intensity maps of the reciprocal (*hhl*) plane around q = (0.21 0.21 1.45) at the lowest temperature T = 50 mK and above the ordering temperature, at T = 1 K. While at 1 K only a q-dependent background is detected, which increases slightly towards lower momentum trans-

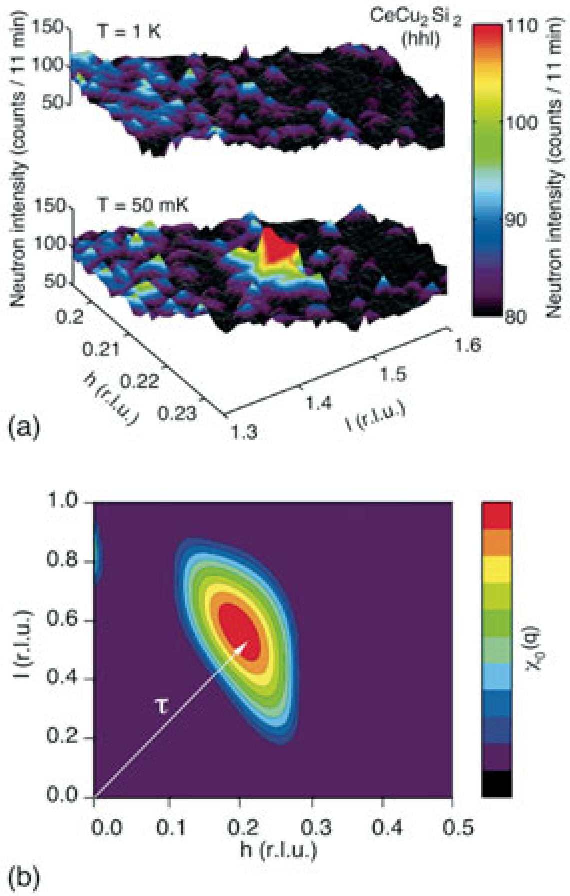
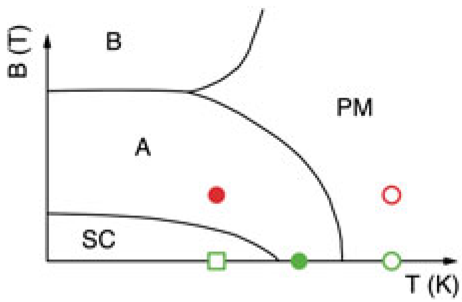


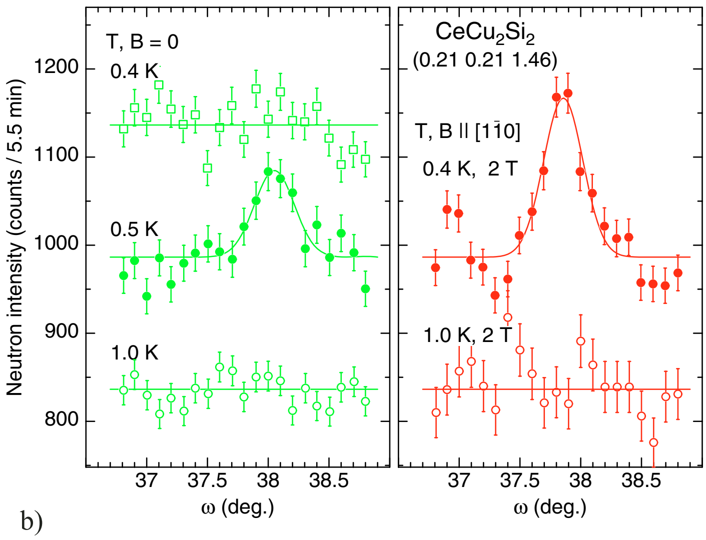
Fig. 1: (a) Intensity map of the reciprocal (hhl) plane around q = (0.21 0.21 1.45) in $CeCu_2Si_2$ at T = 50 mK and 1 K (data taken on diffractometer E6 at HMI/Berlin). (b) Comparison of the measured propagation vector τ with theoretical intensity map for the wave-vector dependent magnetic susceptibility $\chi_0(q)$ of noninteracting quasiparticles in the reciprocal (hhl) plane of $CeCu_2Si_2$ [5].

fer q, a well resolved magnetic superstructure peak is visible at 50 mK. Magnetic peaks were also found at symmetry equivalent positions. Therefore, for the first time incommensurate magnetic peaks were detected in $CeCu_2Si_2$ being direct microscopic evidence for the antiferromagnetic nature of the magnetic order. At the lowest temperature the propagation vector τ was determined from the positions of the magnetic peaks with respect to nuclear peaks yielding $\tau = (0.215\ 0.215\ 0.530)$. The antiferromagnetic superstructure peaks vanish at $T_N \approx 800$ mK in agreement with thermodynamic results. At the lowest temperature the ordered moment is estimated to ~ 0.1 μ_B assuming a sinusoidal modulation of the magnetic moments lying in the basal plane. To understand the value of the propagation vector the band structure of the heavy quasiparticles has been calculated using a renormalized band method [6]. The wave-vector dependent magnetic susceptibility $\chi(q)$ obtained by these calculations is displayed in Fig. 1(b) and exhibits a pronounced maximum at $q \approx (0.2\ 0.2\ 0.5)$ indicating the nesting properties of the Fermi surface. The results for the Fermi surface are in qualitative agreement to [7]. Due to the fact that the position of the maximum of $\chi(q)$ coincides quite well with the observed propagation vector $\tau = (0.215\ 0.215\ 0.53)$, it can be concluded that the antiferromagnetic order in $CeCu_2Si_2$ results from an instability of the renormalized Fermi surface with respect to the formation of a spin-density wave.

With the knowledge of the A-phase magnetism, we started our investigation of the interplay between antiferromagnetism and superconductivity on an A/S single crystal with $T_N \approx 700$ mK and a superconducting transition temperature $T_c \approx 550$ mK [8]. We performed neutron-diffraction experiments on the diffractometers E4 and E6 at HMI/Berlin at temperatures below $T = 1$ K and in magnetic fields up to $B = 2$ T. Rocking scans across the positions of magnetic superstructure peaks were performed at different points in the magnetic (B,T) phase diagram as shown in Fig. 2(a). Here, the (B,T) phase diagram had already been known from thermodynamic and transport measurements. We focused our study to low temperatures and low magnetic fields in order to investigate the interplay between magnetic order and superconductivity. Our main results are displayed in Fig. 2 (b). In zero magnetic field just below the Néel temperature, the magnetic Bragg peak is observed, but has disappeared at $T = 400$ mK, i.e., well inside the superconducting phase. Applying an overcritical magnetic field of $B = 2$ T at $T = 400$ mK to kill superconductivity, leads to a recovery of the antiferromagnetic order. The absence of magnetic Bragg peaks in the superconducting phase gives evidence that in this crystal antiferromagnetism and superconductivity exclude each other on a microscopic scale. These findings are in line with results obtained by μSR and NMR measurements [9,10].



a)



b)

Fig. 2: (a) Schematic magnetic (B,T) phase diagram of $CeCu_2Si_2$ with different magnetically ordered phases (A-, B-phase) and the superconducting phase. The (B,T) values at which neutron diffraction measurements were performed, are marked. (b) Rocking scans across the position of a magnetic superstructure peak in an A/S-$CeCu_2Si_2$ single crystal at different temperatures and magnetic fields [8].

However, the situation in the vicinity of the phase boundaries, i.e., close to T_c, was not known. A possible coexistence of magnetic order and superconductivity even in a small temperature region would have important implications for the order parameters, because in the case of coexistence they would be coupled. In order to address this question, an *in situ* ac susceptibility setup for neutron scattering was designed and successfully used (see also *"New Setup for Simultaneous Measurements of Neutron Scattering and AC Susceptibility"*). The experiment was performed on the triple-axis spectrometer IN12 at the ILL/Grenoble on the same A/S crystal previously used at the HMI. Fig. 3 shows the temperature dependence of the ac susceptibility and the intensity of a magnetic superstructure peak in zero magnetic field and in B = 2 T (applied along $[1\bar{1}0]$). In zero magnetic field, magnetic intensity appears below $T_N \approx$ 700 mK and increases to lower temperatures. It reaches a maximum at the onset of superconductivity at ≈ 550 mK (sharp drop of the ac susceptibility), decreases and finally vanishes at $T \approx$ 400 mK. Applying B = 2 T kills superconductivity and magnetic order is fully recovered with a slightly reduced Néel temperature. The results for $B = 0$ indicate that a small temperature range between 400 and 550 mK exists where both, superconductivity and antiferromagnetic order, are observed. Additional μSR measurements on the same sample [11] indicate that the superconducting transition is first order in nature and point to phase separation, i.e., superconductivity and magnetic order exist in distinctly different volumes in this crystal.

In conclusion, antiferromagnetic order has been identified as the nature of the A-phase in $CeCu_2Si_2$. The observed instability of the Fermi surface is related to the fact that the renormalized 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 $CeCu_2Si_2$. Our measurements give evidence that superconductivity expels antiferromagnetism in the A/S single crystal at low temperatures and that both phenomena do not coexist in this crystal on a microscopic scale.

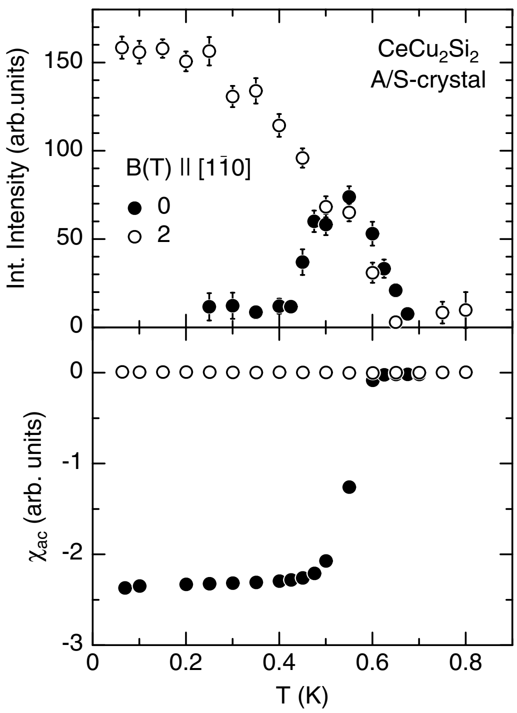


Fig. 3: Temperature dependence of the intensity of a magnetic superstructure peak and the ac susceptibility in an A/S-type $CeCu_2Si_2$ single crystal for B = 0 and 2 T (B || $[1\bar{1}0]$).

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