Spin Fluctuations in the Normal and Superconducting States of the Prototypical Heavy-Fermion Compound $CeCu_2Si_2$

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The origin of unconventional superconductivity, as observed in the cuprate high-temperature superconductors or in the heavy-fermion superconductors, is still one of the open issues in condensed matter physics. While conventional, phonon-mediated superconductivity is quite incompatible with magnetism, magnetic excitations are thought to be at the heart of unconventional superconductivity. Spin excitations seem to be at the origin of Cooper pair formation in these compounds. The occurrence of unconventional superconductivity is often associated with the vicinity to a $T = 0$ magnetic instability, i.e., a magnetic quantum critical point (QCP). Soon after the discovery of heavy-fermion superconductivity spin fluctuations which become critical at a 3D spin-density-wave QCP, have been proposed for the coupling mechanism in these unconventional superconductors [1,2]. As a signature of a magnetic pairing mechanism spin resonances have been observed in several cuprate superconductors [3,4], in recently discovered iron-based superconductors [5,6], but also a few heavy-fermion superconductors [7–9]. However, so far no direct evidence has been given that these systems indeed are located close to a magnetic instability with strongly enhanced magnetic fluctuations which can enable magnetic pairing. For such a study the prototypical heavy-fermion compound $CeCu_2Si_2$ is ideally suited since this tetragonal compound is located very close to a magnetic QCP already at ambient conditions (cf. Fig. 1) [10]. Incommensurate antiferromagnetic order of spin-density-wave type (with a $T_N \approx 800$ mK) is easily suppressed by a tiny pressure or the appropriate stoichiometry giving rise to a superconducting ground state with $T_c \approx 600$ mK. Here, the magnetic order is characterized by a wave vector $\mathbf{Q}_{AF} \approx (0.215\ 0.215\ 0.53)$, which is determined by the nesting properties of the Fermi surface [11].

Using high-resolution inelastic neutron scattering we studied in detail the magnetic response in $CeCu_2Si_2$. For the first time the spin excitations in the normal and superconducting states of a heavy-fermion superconductor were measured in dependence of momentum and energy transfer throughout the relevant part of the Brillouin zone around the antiferromagnetic wave vector $\mathbf{Q}_{AF}$ [12]. The aim of the measurements were twofold: first, to characterize the magnetic excitation spectrum and to calculate the magnetic exchange energies and compare the results to the condensation energy, and second, to verify the vicinity of the compound to a QCP. For the experiments we used a $CeCu_2Si_2$ single crystal ($m \approx 2$ g) with a superconducting $T_c \approx 600$ mK and an upper critical magnetic field $B_{c2} \approx 1.7$ T to fully suppress superconductivity. The sample exhibits only a superconducting ground state and does not show any long-range antiferromagnetic order (so-called S-type crystal). Neutron scattering measurements were performed on the triple-axis spectrometer IN12 located at the high-flux

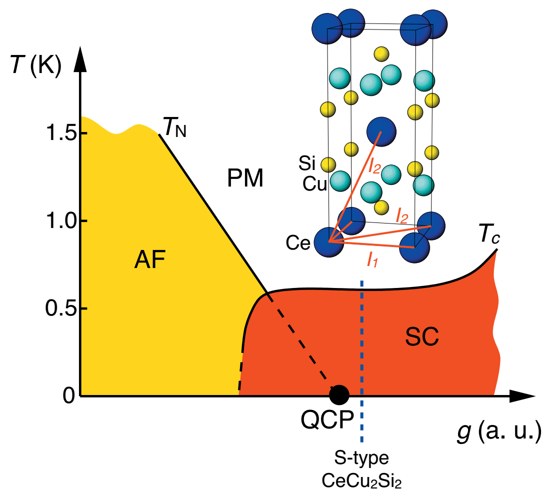

Fig. 1: Schematic T-g phase diagram of $CeCu_2Si_2$ near the magnetic quantum critical point (QCP) where antiferromagnetism (AF) vanishes and superconductivity (SC) appears as function of the effective coupling constant g. Here, g can be changed by hydrostatic pressure or composition (stoichiometry). Superconducting S-type $CeCu_2Si_2$ is loacted on the paramagnetic side of the QCP. The inset displays the tetragonal crystal structure (space group: $I4/mmm$) of $CeCu_2Si_2$ indicating the relevant nearest and next-nearest neighbor cerium interactions I_1 and I_2.

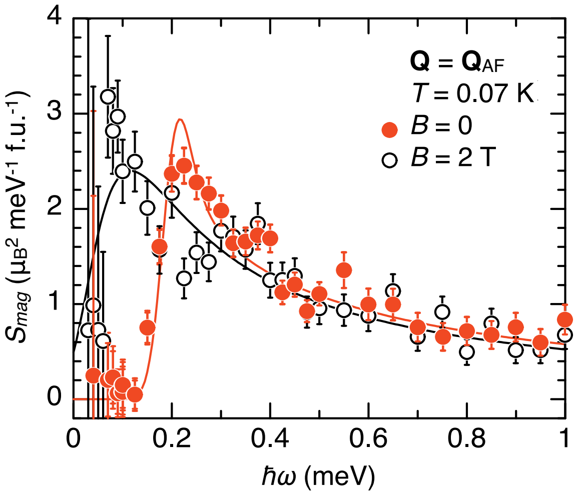

Fig. 2: Magnetic response S_{mag} of S-type $CeCu_2Si_2$ at the antiferromagnetic wave vector $\mathbf{Q}_{AF}$ and T = 0.07 K in the superconducting (B = 0) and normal (B = 2 T) state. While the response in the normal state is quasielastic, it clearly displays a spin excitation gap $\hbar\omega_{gap} \approx$ 0.2 meV in the superconducting state. Solid lines indicate fits to the data.

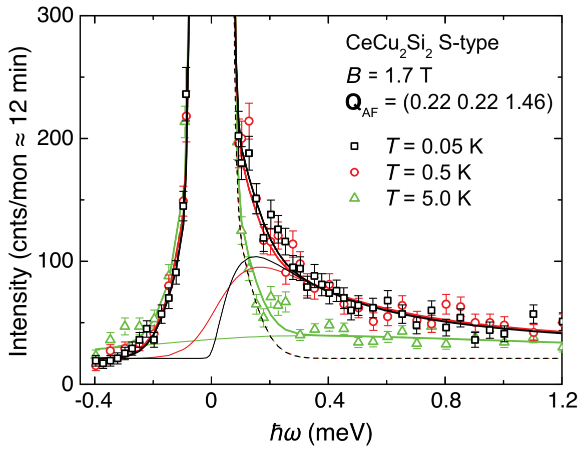

Fig. 3: Energy scans in the normal state of S-type $CeCu_2Si_2$ at several temperatures and at a magnetic field of B = 1.7 T performed at the antiferromagnetic wave vector $\mathbf{Q}_{AF}$. Dashed lines indicate the elastic incoherent scattering and solid lines denote the quasielastic magnetic response.

neutron reactor of the Institut Laue-Langevin in Grenoble. Data were taken with the $CeCu_2Si_2$ crystal aligned in a [110][001] horizontal scattering plane at temperatures between T = 50 mK and 10 K and in magnetic fields up to B = 2 T applied along $[1\bar{1}0]$.

Energy scans in S-type $CeCu_2Si_2$ have been recorded at the antiferromagnetic wave vector $\mathbf{Q}_{AF}$ in the normal and superconducting states as displayed in Figures 2 and 3. Here, measurements in the superconducting state were performed in zero magnetic field at T = 0.07 K, i.e. well below T_c, while the normal state has been generated by applying an overcritical magnetic field at lowest temperatures. In Figure 2 the elastic incoherent part has already been subtracted from the raw data leaving only the magnetic response. The magnetic fluctuations in the normal state are quasielastic and can be well described by a simple Lorentzian line multiplied by the Bose factor (solid lines). It should be noted that irrespective on how the normal state was approached, i.e., either by application of magnetic field or by increasing the temperature above T_c, the magnetic response appears quasielastic. In contrast, the response in the superconducting state appears to be gapped, and spectral weight is transferred from low energies to energies above the spin excitation gap [12]. This gap attains a value of $\hbar\omega_{gap} \approx$ 0.2 meV at T = 0.07 K which results in $\hbar\omega_{gap} \approx 3.9\, k_B T_c$ being roughly 10% smaller than the value predicted for a weak-coupling *d*-wave superconductor [12]. With increasing temperature the spin gap becomes smaller and closes at T_c.

Our measurements were not restricted to $\mathbf{Q}_{AF}$, we carefully investigated the momentum dependence of the magnetic response. The main finding is that the spin excitation at $\mathbf{Q}_{AF}$ is part of an overdamped dispersive mode with a mode velocity being more than an order of magnitude smaller than the renormalized Fermi velocity. This indicates a clear retardation in the interactions between the spin excitations and the heavy quasiparticles. With this detailed knowledge of the energy and momentum dependence of the spin excitations in S-type $CeCu_2Si_2$, we were able to calculate the difference in magnetic exchange energy between the superconducting and the normal state [12]. For the calculation the normal and superconducting states magnetic susceptibilities (as derived from the neutron scattering) were used and the nearest and next-nearest neighbor exchange interactions (cf. Fig. 1) taken into account. In addition, the superconducting condensation energy was obtained from heat capacity measurements performed on the same sample. As a result we find that the gain in exchange energy is larger, roughly by a factor of 20, than the condensation energy [12]. This implies that the spin excitations are the driving force for superconductivity in $CeCu_2Si_2$.

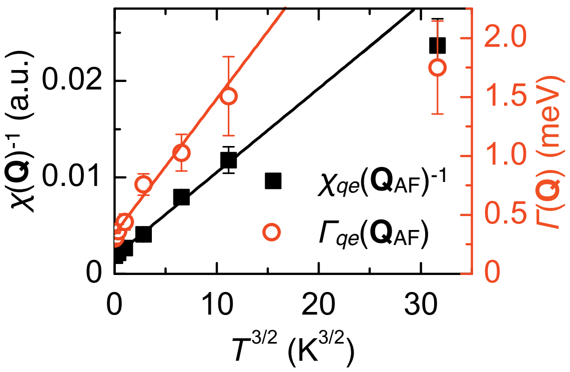

Fig. 4: Inverse spin susceptibility $\chi(\mathbf{Q}_{AF})^{-1}$ and energy width $\Gamma(\mathbf{Q}_{AF})$ of the quasielastic (qe) normal-state magnetic response in S-type $CeCu_2Si_2$ at $B = 1.7$ T plotted against $T^{3/2}$. Solid lines indicate fits to the data.

To be able to study the spin fluctuations in the normal state of $CeCu_2Si_2$ when tuning the system towards the QCP, superconductivity was suppressed in a magnetic field $B = 1.7$ T ($\approx B_{c2}$). As seen in Figure 3 the quasielastic magnetic response at the ordering wave vector $\mathbf{Q}_{AF}$ broadens notably and decreases in intensity at higher temperatures. Fitting the data by quasielastic Lorentzian lines yields the spin susceptibility $\chi(\mathbf{Q}_{AF})$ and the line width in energy (FWHM), $\Gamma(\mathbf{Q}_{AF})$, of the magnetic fluctuations, $\Gamma(\mathbf{Q}_{AF})$ being a measure of the fluctuation rate and hence inversely proportional to the lifetime of the spin fluctuations. In Fig. 4 $\chi(\mathbf{Q}_{AF})^{-1}$ and $\Gamma(\mathbf{Q}_{AF})$ are plotted against $T^{3/2}$. The line width Γ displays a strong temperature dependence and decreases substantially towards lower temperature with only a small residual Γ for $T \rightarrow 0$. Concomitantly, the lifetime of the magnetic fluctuations almost diverges indicating an almost critical slowing down of the normal state magnetic response. The finite intercept in Γ for $T \rightarrow 0$ puts S-type $CeCu_2Si_2$ slightly on the paramagnetic side of the QCP. As expected, the spin susceptibility $\chi(\mathbf{Q}_{AF})$ increases strongly for decreasing temperatures. $\chi(\mathbf{Q}_{AF})^{-1}$ and $\Gamma(\mathbf{Q}_{AF})$ vary as $T^{3/2}$ at temperatures well below the Kondo temperature $T_K \approx 15$ K (solid lines in Figure 4). Such a $T^{3/2}$ dependence of the spin susceptibility and of the fluctuation rate is indeed predicted for a 3D spin-density-wave QCP [13–15].

In summary, we observed for the first time an almost critical slowing down of the normal-state quasielastic magnetic response in the heavy-fermion superconductor $CeCu_2Si_2$, with temperature dependences expected for a 3D spin-density-wave QCP. In contrast, the spin excitations in the superconducting state are gapped with a gap value close to the one predicted for a d-wave BCS superconductor. Based on the measured energy and momentum dependence of the spin excitations the magnetic exchange energies have been calculated. As a result, the gain in magnetic exchange energy from the normal into the superconducting state of $CeCu_2Si_2$ is substantially larger than the superconducting condensation energy. Hence, these spin fluctuations can be regarded as the driving force for the superconducting pairing in this prototypical heavy-fermion system located close to an antiferromagnetic QCP.

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