Paramagnon-mediated Superconductivity in $CeCu_2Si_2$

Oliver Stockert, Julia Arndt, Enrico Faulhaber[1], Karin Schmalzl[2], Wolfgang Schmidt[2], Astrid Schneidewind[1,3], Hirale S. Jeevan, Christoph Geibel, Michael Loewenhaupt[1], and Frank Steglich

Coexistence or competition of superconductivity and magnetic order remains an important issue in condensed matter physics, especially in strongly correlated electron or heavy-fermion systems. While conventional superconductivity is generally incompatible with magnetism, the magnetic 4f elements play a crucial role in heavy-fermion superconductors and are absolutely indispensable for the appearance of unconventional superconductivity. Instead of phonons the spin excitations are thought to be involved in the mechanism of Cooper pair formation. The situation seems to be similar to the high-temperature cuprate superconductors where a spin excitation resonance in the superconducting state has been observed. However, due to the enhanced superconducting T_c of order one hundred Kelvin in the cuprate superconductors and the associated large energies, experiments are quite often hampered by phonons and other excitations in the energy range of interest. This inhibits an unambiguous detailed observation of the spin excitations below T_c. Here we show that a clear spin excitation resonance is present in the superconducting state of the prototypical heavy-fermion compound $CeCu_2Si_2$, for the first time observed at an incommensurate wave vector in a heavy-fermion superconductor. As a consequence of the low superconducting T_c phonon processes do not significantly contribute to the excitation spectrum. Our results demonstrate that the spin excitations are highly relevant for the superconducting pairing in $CeCu_2Si_2$. In contrast to the cuprates with quasi two-dimensional electronic structure, the observation of the spin excitation resonance in $CeCu_2Si_2$ with a three-dimensional structure suggests that the appearance of a spin resonance is a general feature of unconventional superconductivity.

Many open questions still persist concerning the interplay of superconductivity and antiferromagnetism in the tetragonal heavy-fermion compound $CeCu_2Si_2$. Sensitively depending on the stoichiometry in the homogeneity range and on the growing conditions, $CeCu_2Si_2$ exhibits different ground states: It shows either antiferromagnetism (A-type), superconductivity (S-type), or a ground state in which the two compete with each other without coexistence (A/S-type) [1]. The nature of the antiferromagnetic order was found to be an incommensurate spin-density wave [2]. At T = 50 mK, well below $T_N \approx 800$ mK, A-type $CeCu_2Si_2$ exhibits an ordered magnetic moment $\mu_{ord} \approx 0.1\ \mu_B$ and a propagation vector $\tau \approx (0.215\ 0.215\ 0.53)$, the latter being temperature dependent and determined by the nesting of the Fermi surface. Different neutron scattering and muon-spin rotation experiments on A/S-type crystals indicate that superconductivity and antiferromagnetism do not coexist on a microscopic scale, rather a separation between superconducting and magnetic volumes takes place [3,4].

Therefore, inelastic neutron scattering measurements on S-type $CeCu_2Si_2$ were performed on the cold-neutron triple-axis spectrometers PANDA at the new Munich research reactor FRM-II and IN12 at the high-flux reactor of the Institut Laue-Langevin in Grenoble/France. For all experiments the S-type $CeCu_2Si_2$ single crystal ($m \approx 2$ g) was mounted with the $[1\,\bar{1}\,0]$ axis vertical on a copper pin attached to the mixing chamber of a dilution refrigerator. The setup results in a [110]-[001] scattering plane. Data were taken at temperatures between

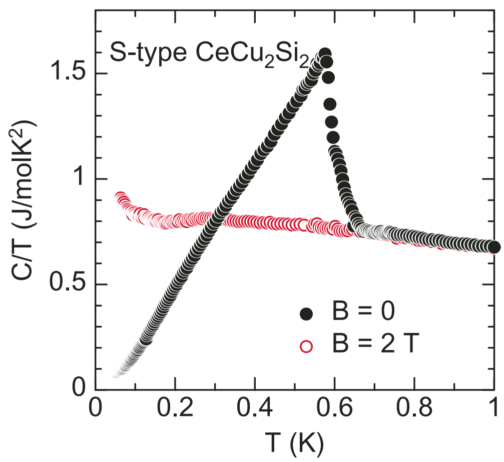

Fig. 1: Specific heat C of S-type $CeCu_2Si_2$ plotted as C/T versus temperature T in zero magnetic field B = 0 and a magnetic field B = 2 T applied along $[1\,\bar{1}\,0]$.

T = 60 mK and 1 K and in magnetic fields up to B = 2.5 T applied along the vertical $[1\,\bar{1}\,0]$ axis.

The S-type single crystal was characterized by bulk measurements, namely heat capacity and ac susceptibility. The heat capacity was measured using a compensated heat-pulse technique with background heating [6]. The ac susceptibility was obtained by means of a homemade susceptibility setup which allowed data recording simultaneous to the neutron scattering experiment, i.e., while taking neutron data. As shown in Fig. 1 the specific heat, plotted as C/T vs. T, exhibits a pronounced maximum indicating the onset of superconductivity at T_c = 0.6 K. The shape of the anomaly at T_c in comparison to heat capacity measurements on other $CeCu_2Si_2$ samples and its complete suppression in a magnetic field of B = 2 T clearly show that this is a transition into the superconducting state. The superconducting nature of this phase transition is further confirmed by susceptibility measurements (not shown).

Elastic neutron scattering measurements featured no resolution-limited magnetic Bragg peaks in S-type $CeCu_2Si_2$ [5]. This is in accordance with thermodynamic measurements where no magnetic transition was detected. However, at positions where magnetic satellite peaks are observed in A-type $CeCu_2Si_2$ [2], e.g. at $\mathbf{Q} \approx (0.22\ 0.22\ 1.46)$, the S-type crystal exhibits quite weak and broad magnetic correlation peaks at low temperatures as can be seen in Fig. 2 [5]. These peaks were found to be purely elastic within the energy resolution of 55-60 μeV. However, their line width in $\mathbf{q}$ space is considerably broadened corresponding to a correlation length of 50-60 Å. It is worth noting that this correlation length is comparable to the superconducting coherence length of about 100 Å [7]. These short-range magnetic correlations do not vanish at T_c = 0.6 K, but are still present above T_c and disappear at $T \approx 0.8$ K.

Inelastic neutron scattering to study the magnetic response focused around the nesting wave vector in the system, $\mathbf{Q} = \mathbf{Q}_{AF} = (0.215\ 0.215\ 1.458)$. Fig. 3(a) displays energy scans at this $\mathbf{Q}_{AF}$ position and at a general position $\mathbf{Q} = \mathbf{Q}_{arb} = (0.1\ 0.1\ 1.6)$, where no correlation peaks emerge, but which has the same $|\mathbf{Q}|$. Both data sets were recorded at T = 0.07 K, i.e., in the superconducting state. At the general $\mathbf{Q}_{arb}$ position only the incoherent elastic background contribution with instrument resolution is seen, while no magnetic intensity could be

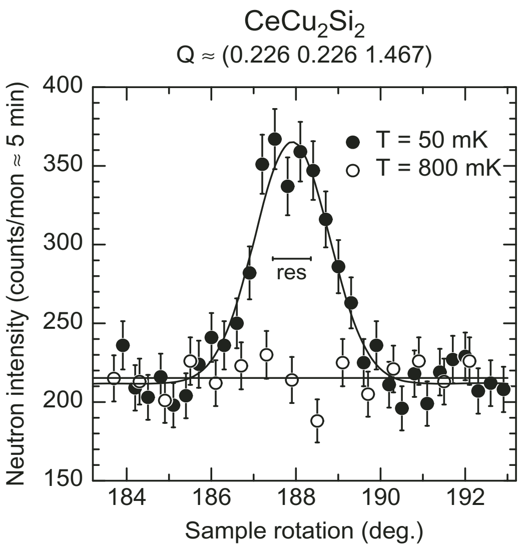

Fig. 2: Short-range magnetic correlations in S-type $CeCu_2Si_2$ at B = 0 measured at $\hbar\omega$ = 0. Rocking scan (neutron counts versus sample rotation ω) across $\mathbf{Q} \approx (0.22\ 0.22\ 1.46)$ at T = 0.05 K and 0.8 K [5].

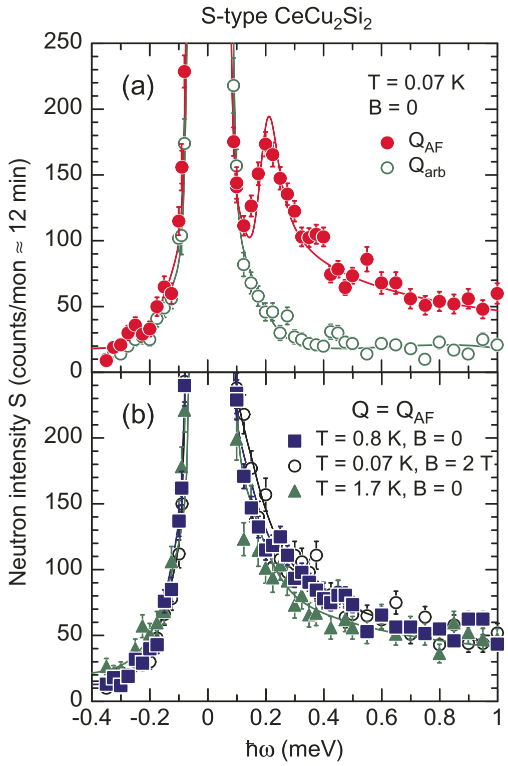

Fig. 3: Energy scans (neutron intensity $S = S_{ela} + S_{qe/ine,\ mag}$ versus energy transfer $\hbar\omega$) in S-type $CeCu_2Si_2$ at $\mathbf{Q} = \mathbf{Q}_{AF}$ = (0.215 0.215 1.458) in (a) the superconducting state at T = 0.07 K, B = 0 and in (b) the normal state at T = 0.8 and 1.7 K, B = 0 and T = 0.07 K, B = 2 T. For comparison the magnetic response at an arbitrary, general $\mathbf{Q}$ position $\mathbf{Q} = \mathbf{Q}_{arb}$ = (0.1 0.1 1.6) at T = 0.07 K, B = 0 is also plotted in (a). Solid lines represent fits to the data (see text).

detected. In contrast, at $\mathbf{Q}_{AF}$ the response shows a strong inelastic signal ("spin resonance" peak). The missing spectral weight at lower energies is a first indication for a magnetic excitation gap in the superconducting state. The data can be described by a quasielastic Lorentzian line with a spin excitation gap $\hbar\omega_{gap} \approx$ 0.2 meV and with a density of states in accord with the electronic gap of a superconductor (solid line in Fig. 3(a)). Setting $\hbar\omega_{gap}$ equal to the superconducting gap $2\Delta_0$, one obtains $2\Delta_0/k_B T_c \approx 3.9$, close to the value that was predicted for a weak-coupling d-wave superconductor [8]. To unambiguously relate the inelastic magnetic excitation to the superconducting state it was necessary to perform additional measurements in the normal state, i.e., above T_c at zero magnetic field and above B_{c2} at lowest temperatures.

Energy scans recorded at $\mathbf{Q}_{AF}$ in the normal state are shown in Fig. 3(b). Surprisingly, independent of how the normal state is reached, i.e., above T_c at T = 0.8 K and B = 0 or above B_{c2} at T = 0.07 K and B = 2 T, the magnetic response is quite identical and appears to be quasielastic. The fits to the quasielastic magnetic response with a Lorentzian line shape give a good description of the data as seen in Fig. 3(b). At higher temperature in zero magnetic field the magnetic response weakens in intensity and broadens considerably. Starting from Γ = 0.32 meV at T = 0.8 K the line width of the quasielastic response at $\mathbf{Q}_{AF}$ increases to Γ = 0.47 meV at T = 1.7 K. This considerable slowing down of the response when lowering the temperature indicates the vicinity of S-type $CeCu_2Si_2$ to a quantum phase transition with an adjacent magnetically ordered phase. However, a critical slowing down has only been observed in magnetically ordered A-type $CeCu_2Si_2$ [9].

The fact that the magnetic excitation gap disappears not only in the paramagnetic state above T_c where also the magnetic short-range correlations vanish, but also above B_{c2} at low temperatures where these magnetic correlations still persist, gives clear evidence that the spin resonance is directly related to the superconducting state. The fact that a spin resonance occurs at $\mathbf{Q}_{AF}$ points directly to a $d_{x^2-y^2}$-wave symmetry of the superconducting gap function in $CeCu_2Si_2$ as recent theoretical calculations indicate [10]. Several superconductors with strong electronic correlations show a spin resonance in their magnetic excitation spectrum in the superconducting state, namely the high-temperature cuprate superconductors [11] and the heavy-fermion superconductors UPd_2Al_3 [12,13] and $CeCoIn_5$ [14]. In all of them a d-wave symmetry of the superconducting gap is discussed.

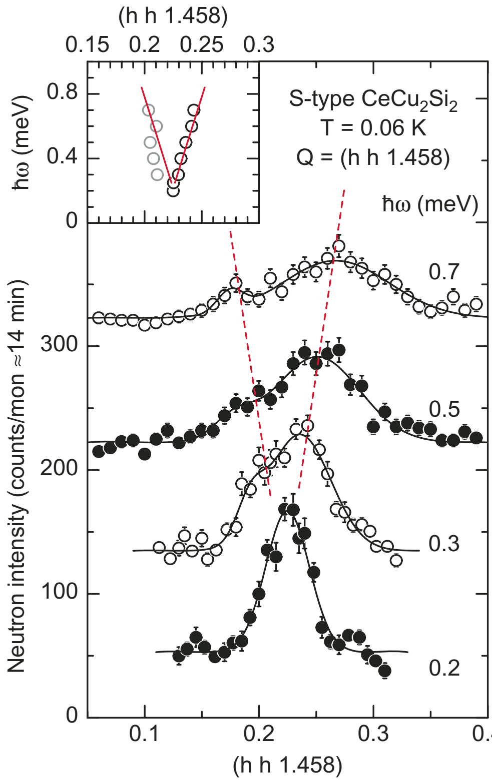

Fig. 4: Wave vector $\mathbf{Q}$ *dependence of the magnetic response around* $\mathbf{Q}_{AF}$ *in S-type* $CeCu_2Si_2$ *in the superconducting state at T = 0.06 K for different energy transfers ħω. The scans are shifted by 100 counts with respect to each other. Inset: Dispersion of the magnetic excitation around* $\mathbf{Q}_{AF}$ *at T = 0.06 K as a result of the fits to the* $\mathbf{Q}$ *scans.*

The interrelation between the spin excitation and the charge properties being responsible for superconductivity, can be understood since the cerium 4f electrons carrying the magnetism, are located at the Fermi surface and therefore, are also involved in the formation of the superconducting Cooper pairs. Hence, inelastic neutron scattering can give direct information about the superconducting gap properties. A similar temperature evolution of the spin resonance energy as in $CeCu_2Si_2$ has been reported in UPd_2Al_3 [15], while in contrast the variation of the resonance peak with temperature in $CeCoIn_5$ [14] is much weaker than expected from BCS theory.

We now turn to the momentum dependence of the magnetic response around $\mathbf{Q}_{AF}$ in the superconducting state. Fig. 4 displays $\mathbf{Q}$ scans along (h h 1.458) across $\mathbf{Q}_{AF}$ recorded at different energy transfers $\hbar\omega$ and at T = 0.06 K. The single peak seen at $\hbar\omega \approx 0.2$ meV splits upon increasing energy transfer into two peaks which move further apart from each other and broaden in **Q** along with an overall decrease in intensity. Hence, the spin resonance observed in $CeCu_2Si_2$ at $\mathbf{Q}_{AF}$ and $\hbar\omega \approx$ 0.2 meV is not a single point in **Q**-ω space, but the origin of a damped propagating mode. It is unknown why the corresponding peak intensities differ considerably. Effects of the instrumental resolution can be ruled out as possible origin. Fits with two peaks of Gaussian line shape (solid lines) yield a good description of the data. As a result the peak positions for different $\hbar\omega$ have been drawn in the inset of Fig. 4. A strong upward dispersion of the spin resonance mode is visible which appears to be linear in **Q**. From a combined linear fit to both branches the velocity of the mode has been determined to v = (4.44 ± 0.86) meVÅ. Our findings of a strong in-plane dispersion of the resonance mode is in line with calculations yielding a considerable dispersion in-plane while the dispersion along c^* is very flat [10]. In comparison, UPd_2Al_3 also exhibits a dispersive spin excitation starting at the spin resonance with an upward dispersion with a slightly higher in-plane mode velocity [16]. However, the situation in the cuprate superconductors is more complex with an hour-glass like dispersion of the resonance mode [17-20].

Although $CeCu_2Si_2$ shares a lot of similarities with other heavy-fermion superconductors, some striking differences between the systems are apparent. The most prominent one is the **Q** position of the spin resonance. In UPd_2Al_3 and $CeCoIn_5$ it occurs at commensurate positions, **Q** = (0 0 ½) and (½ ½ ½), respectively [14,16]. In contrast, in S-type $CeCu_2Si_2$ the spin resonance is seen at the nesting wave vector of the system which is incommensurate. Furthermore, unlike $CeCoIn_5$ the spin excitation gap in $CeCu_2Si_2$ and UPd_2Al_3 do follow the expected BCS form. While UPd_2Al_3 becomes superconducting within an antiferromagnetically ordered state with coexistence of both phenomena, superconductivity and long-range antiferromagnetism exclude each other in $CeCu_2Si_2$. The slowing down of the magnetic response at $\mathbf{Q}_{AF}$ in the normal state of $CeCu_2Si_2$ together with the spin excitation mode with upward dispersion strongly suggest that antiferromagnetic paramagnons are directly involved in the superconducting pairing mechanism. These paramagnons, propagating from $\mathbf{Q}_{AF}$, slow down in the normal state when lowering the temperature. However, they do not become critical which would lead to an antiferromagnetically ordered state as in A-type $CeCu_2Si_2$ [2], but become gapped upon entering the superconducting state and result in the observed spin resonance mode. We conclude by stating that our inelastic neutron scattering data provide convincing experimental evidence for paramagnon-mediated superconductivity as first proposed for heavy-fermion superconductors by Scalapino et al. [21] and subsequently by Mathur et al. [22] for the pressure-induced superconductor $CePd_2Si_2$.

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[1] Institut für Festkörperphysik, Technische Universität Dresden, 01062 Dresden
[2] Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, Jülich Centre for Neutron Science at Institut Laue-Langevin, 38042 Grenoble, France
[3] Neutronenforschungsquelle Heinz-Maier-Leibnitz (FRM-II), TU München, 85747 Garching