# Spin Fluctuations in Normal State $CeCu_2Si_2$ on Approaching the Quantum Critical Point

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We report on the magnetic excitation spectrum in the normal state of the heavy-fermion superconductor $CeCu_2Si_2$ on approaching the quantum critical point (QCP). The magnetic response in the superconducting state is characterized by a transfer of spectral weight to energies above a spin excitation gap. In the normal state, a slowing-down of the quasielastic magnetic response is observed, which conforms to the scaling expected for a QCP of spin-density-wave type. This interpretation is substantiated by an analysis of specific heat data and the momentum dependence of the magnetic excitation spectrum. Our study represents the first direct observation of an almost critical slowing-down of the normal state magnetic response at a QCP when suppressing superconductivity. The results strongly imply that the coupling of Cooper pairs in $CeCu_2Si_2$ is mediated by overdamped spin fluctuations.

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The coupling mechanism of unconventional superconductivity, such as in the high-$T_c$ cuprates and the heavy-fermion superconductors, is still an unsettled matter. While magnetism and conventional, phonon-mediated, coupling of Cooper pairs are mutually exclusive, the occurrence of unconventional superconductivity is often associated with the vicinity to a magnetic instability. A prominent example is the heavy-fermion compound $CePd_2Si_2$, where a superconducting (SC) region appears around an antiferromagnetic (AF) quantum critical point (QCP), when the magnetic order is suppressed to $T = 0$ by hydrostatic pressure [1].

A QCP is characterized by a divergence of the lifetime of critical spin fluctuations for $T \rightarrow 0$, referred to as critical slowing-down. Accordingly, their energy width should approach zero. QCP of the spin-density-wave (SDW) type are described in the conventional Hertz-Millis-Moriya theory, which is a quantum generalization of finite-temperature continuous phase transitions [2–4]. In the case of three-dimensional (3D) AF fluctuations, scaling of the form $\chi''(\mathbf{Q}, \omega) = T^{-3/2} f(\omega/T^{3/2})$ is expected, where $f$ is an appropriate scaling function. The development of a different scenario for quantum critical behavior was fuelled by the observation of scaling of the form $\chi''(\mathbf{Q}, \omega) = T^{-0.75} f(\omega/T)$ in $CeCu_{5.9}Au_{0.1}$ [5]. This "local" scenario is characterized by a breakdown of the Kondo effect at the QCP [6–8].

This Letter comprises an investigation of the magnetic excitation spectrum in the normal state of the heavy-fermion superconductor $CeCu_2Si_2$ by means of inelastic neutron scattering. Our study represents the first direct observation of an almost critical slowing-down of the normal state magnetic response at a QCP when superconductivity is suppressed.

Mediation of Cooper-pair coupling by collective magnetic fluctuations (paramagnons), which emerge in the vicinity to a SDW instability, has already been proposed earlier [9,10], shortly after the discovery of superconductivity in several heavy-fermion metals, first and foremost in $CeCu_2Si_2$ [11]. Strong implications for magnetically mediated superconductivity are given by the observation of a spin resonance in the SC state of the high-$T_c$ cuprate [12,13] and iron superconductors [14,15] as well as the heavy-fermion superconductors $UPd_2Al_3$ [16,17] and $CeCoIn_5$ [18]. While local-moment antiferromagnetism and superconductivity coexist in $UPd_2Al_3$, $CeCoIn_5$ is nonmagnetic, with the substitution of, for example, several percent of Co with Rh or of In with Cd being necessary to induce magnetic order [19,20]. In contrast to this, the system $CeCu_2Si_2$ ($T_K \approx 10$ K [21]) is characterized by a QCP being located within the narrow homogeneity range of the 1:2:2 phase. Accordingly, homogeneous samples with a slight Cu deficit or excess exhibit AF order ($A$-type) or superconductivity without long-range AF order ($S$-type). In perfectly stoichiometric samples, magnetic order and superconductivity compete microscopically with each other ($A/S$-type). The magnetic order was determined to be of incommensurate SDW type with a propagation vector $\tau = (0.22, 0.22, 0.53)$ corresponding to the nesting vector of the renormalized Fermi surface [22]. The ordered magnetic moment is very small; it amounts to $\approx 0.1\mu_B$/Ce at low $T$ in $A$-type $CeCu_2Si_2$.

Previous measurements of electrical resistivity $\rho(T)$ and heat capacity $C(T)$ on $S$-type $CeCu_2Si_2$ polycrystals give indications of non-Fermi-liquid (NFL) behavior consistent with predictions made in the frame of the Hertz-Millis-Moriya theory in the vicinity of a QCP of SDW type with 3D AF fluctuations, namely, $\Delta\rho = \beta T^{3/2}$

 

and $C/T = \gamma^* - a\sqrt{T}$ [23]. As inferred from susceptibility measurements [Fig. 1(a)], the upper critical magnetic field is determined to $B_{c2}(T \to 0) = 1.7$ T [see also Fig. 1(c)].

In the SC state, $S$-type $CeCu_2Si_2$ exhibits an inelastic magnetic response at the position $\mathbf{Q}_{AF}$, where magnetic Bragg peaks appear in $A$- and $A/S$-type crystals, whereas the excitation spectrum is quasielastic in the normal state at low $T$ and $B > B_{c2}$ as well as at $B = 0$ and $T > T_c$ [24]. The inelastic signal results from a transfer of spectral weight from energies below to above a spin excitation gap $\hbar\omega_{gap}$. In contrast to $CeCoIn_5$, the signal in the SC state is broad in energy with a tail extending to more than 10 times $\hbar\omega_{gap}$. With $\hbar\omega_{gap} = 0.2$ meV at $T = 0.07$ K the magnitude of the spin gap is consistent with estimations for the quasiparticle excitation gap of a superconductor within the frame of BCS theory ($\hbar\omega_{gap}/k_B T_c = 3.9$), as is its $T$ dependence [24]. The $\mathbf{Q}$ dependence of the magnetic response at different energy transfers in the SC state shows an overdamped mode with linear dispersion [24]. These results imply magnetically mediated superconductivity. Yet, in order to verify the concept of AF fluctuations acting as a driving force for Cooper-pair formation in $CeCu_2Si_2$, the magnetic response upon approaching the QCP is to be studied. The QCP occurs at ambient pressure, which facilitates investigations of the critical magnetic excitation spectrum by neutron scattering in $CeCu_2Si_2$ drastically, as compared to $CePd_2Si_2$. The presented study of the AF fluctuations in the vicinity of the QCP is important towards an understanding of superconductivity in $CeCu_2Si_2$, particularly in light of the recent findings concerning the spin excitation gap in the SC state of the $S$-type variant.

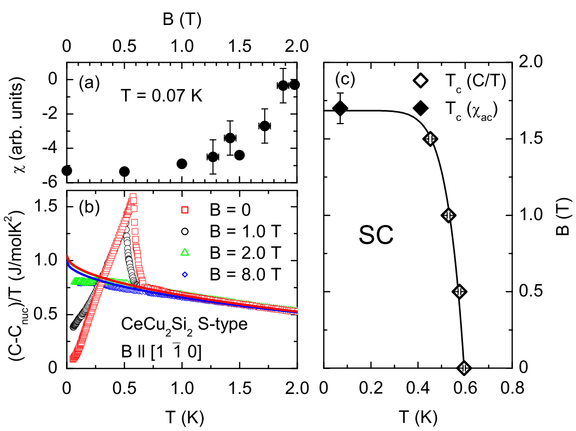


FIG. 1 (color online). (a) Magnetic field dependence of $\chi_{ac}$ at $T = 0.07$ K. Data with large error bars were taken while sweeping the field, whereas all other data were measured at constant fields. (b) Temperature dependence of the specific heat as $C(T)/T$ in different magnetic fields after subtraction of the nuclear Schottky contribution. Solid lines indicate fits to $C/T = \gamma^* - a\sqrt{T}$. (c) $B$-$T$ phase diagram, compiled from the $C(T)$ and $\chi_{ac}$ measurements.

Neutron scattering experiments were performed at the cold-neutron three-axis spectrometer IN12 at the high-flux reactor of Institut Laue-Langevin, Grenoble (France). A vertically focusing graphite (002) monochromator and a doubly focusing graphite (002) analyzer were used. The horizontal collimation was 60′ before the sample, while no collimation was inserted before the analyzer. A liquid nitrogen-cooled Be filter was placed in the incident neutron beam. The final neutron wave vector was fixed to $k_f = 1.15$ Å$^{-1}$, which corresponds to a final neutron energy of $E_f = 2.74$ meV. The energy resolution, i.e., full width at half maximum (FWHM), therefore amounts to 0.057 meV. Complementary heat capacity measurements were performed by applying a quasiadiabatic heat-pulse method with additional background heating [25]. Figure 1(b) is a plot of $C/T$ vs $T$ in different representative magnetic fields, measured on the same large single crystal ($m \approx 2$ g) as used for neutron scattering studies. The crystal exhibits superconductivity below $T_c \approx 0.6$ K at $B = 0$. For $B = 0$ and 1.0 T, the expected NFL behavior $C/T = \gamma^* - a\sqrt{T}$ is observed in the normal state for $T > T_c$ up to at least $T = 2.0$ K. For $B = 2.0$ and 8.0 T, i.e., $B > B_{c2}$, a $\sqrt{T}$ fit is appropriate down to $T \approx 0.8$ K, before a crossover to Fermi-liquid behavior sets in, and $C/T$ becomes constant for $T < 0.3$ K.

In order to be able to investigate the magnetic excitation spectrum in the normal state on approach to the QCP, superconductivity was suppressed by applying a magnetic field close to $B_{c2}(T \to 0) = 1.7$ T. The crystal was mounted with the $[1\bar{1}0]$ axis vertical on a copper pin attached to the mixing chamber of a dilution refrigerator, yielding an ($hhl$) scattering plane. The magnetic field was applied parallel to the $[1\bar{1}0]$ direction.

Energy scans were performed at a wave vector transfer corresponding to $\mathbf{Q}_{AF} = (0.22, 0.22, 1.46)$ at different temperatures from 0.05 to 10 K. Three representative spectra are shown in Fig. 2. The measured scattering intensity is proportional to the scattering function $S(\mathbf{Q}, \omega) = [1 - \exp(-\hbar\omega/k_B T)]^{-1}\chi''(\mathbf{Q}, \omega)$, where $\chi''(\mathbf{Q}, \omega)$ denotes the imaginary part of the dynamical susceptibility. In $S$-type $CeCu_2Si_2$, the scattering function at $\mathbf{Q} = \mathbf{Q}_{AF}$ and energy transfer $\hbar\omega$ is composed of an incoherent elastic and a coherent quasielastic contribution in the whole temperature range considered. The quasielastic contribution has a Lorentzian line shape:

$$S_{qel}(\mathbf{Q}_{AF}, \omega) = \frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} \frac{\chi(\mathbf{Q}_{AF})}{\pi[\Gamma(\mathbf{Q}_{AF})/2]} \times \frac{1}{1 + \{\hbar\omega/[\Gamma(\mathbf{Q}_{AF})/2]\}^2}, \qquad (1)$$

where $\chi(\mathbf{Q}_{AF})$ denotes the spin susceptibility and $\Gamma(\mathbf{Q}_{AF})$ the energy width (FWHM) of the spin fluctuations, which is a measure for the fluctuation rate and therefore inversely

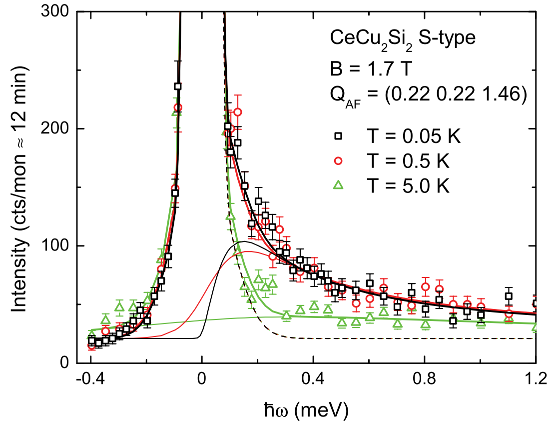


FIG. 2 (color online). Energy scans in $S$-type $CeCu_2Si_2$ at three different temperatures in a field $B = 1.7\ \mathrm{T} \approx B_{c2}(T \rightarrow 0)$, performed at the wave vector $\mathbf{Q}_{\mathrm{AF}}$. Dashed lines indicate fits to the elastic part of the scattering intensity with two Gaussians to model the instrumental resolution as determined by measurements of a vanadium sample [28]; thin full lines show fits of the quasielastic part; thick full lines are the sum of both contributions.

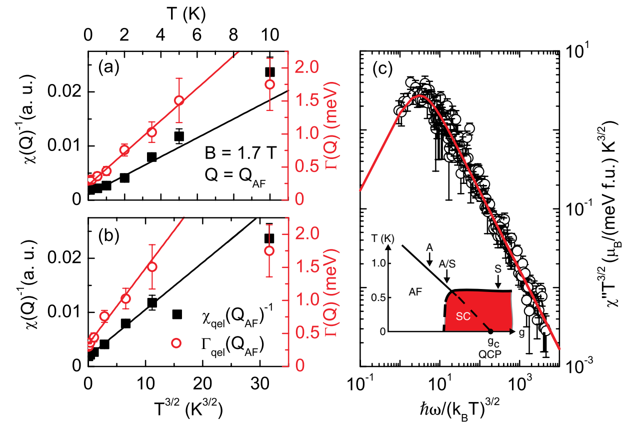


FIG. 3 (color online). Inverse spin susceptibility and energy width of the quasielastic contribution to the neutron scattering intensity in $S$-type $CeCu_2Si_2$ at $\mathbf{Q}_{\mathrm{AF}}$ and $B = 1.7$ T, (a) plotted vs $T$ and (b) plotted vs $T^{3/2}$. Lines indicate linear fits as described in the text. (c) Scaling plot of the imaginary part of the dynamical susceptibility $\chi''(\mathbf{Q}, \omega)$ as $\chi'' T^{3/2} = f[\hbar\omega/(k_B T)^{3/2}]$. The solid line represents a fit to the data as described in the text. The inset shows the $T$-$g$ phase diagram of $CeCu_2Si_2$, where $g$ is the effective coupling constant, which is proportional to the square of the hybridization between $4f$ and conduction electrons. $A$, $A/S$, and $S$ refer to the respective types of $CeCu_2Si_2$ crystals. $g$ can be varied by chemical composition as well as hydrostatic pressure.

proportional to the lifetime. The applicability of the scaling law for $\chi''(\mathbf{Q}_{\mathrm{AF}}, \omega)$ expected for QCP of the SDW type can be checked by showing that $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1} \propto T^{3/2}$ as well as $\Gamma(\mathbf{Q}_{\mathrm{AF}}) \propto T^{3/2}$ [2–4]. This holds in the NFL regime, i.e., in the temperature range $T_{\mathrm{FL}} < T \ll T_K$, where $T_{\mathrm{FL}}$ is a crossover temperature below which Fermi-liquid behavior is observed.

Figure 3 displays $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1}$ and $\Gamma(\mathbf{Q}_{\mathrm{AF}})$ as extracted from fits to Eq. (1), on the one hand plotted vs $T$ [Fig. 3(a)] and on the other hand plotted vs $T^{3/2}$ [Fig. 3(b)]. The data sets are fitted linearly in both plots for $T \leq 5.0$ K , as the condition $T \ll T_K$ is not fulfilled for $T = 10.0$ K. Especially at low $T$, $\Gamma(\mathbf{Q}_{\mathrm{AF}})$ and $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1}$ are better described by a $T^{3/2}$ [goodness of fit: $\chi^2(\chi^{-1}) = 0.92$; $\chi^2(\Gamma) = 1.29$] than by a linear $T$ dependence [$\chi^2(\chi^{-1}) = 7.51$; $\chi^2(\Gamma) = 1.49$]. Strikingly, extrapolating both quantities to $T \rightarrow 0$ does not lead to zero but to finite values. This is due to the fact that $S$-type $CeCu_2Si_2$ is positioned slightly on the paramagnetic side of the QCP, with the $y$-axis intercept being a measure of the distance to the QCP, as presented in the schematic $T$-$g$ phase diagram of $CeCu_2Si_2$ [inset in Fig. 3(c)] (see also [26]). By fitting $\Gamma(\mathbf{Q}_{\mathrm{AF}})$ and $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1}$ to an expression of the form $c_1 + c_2 T^{\alpha}$, one obtains $\Gamma(\mathbf{Q}_{\mathrm{AF}}) = (0.30 \pm 0.02) + (0.14 \pm 0.03) T^{1.38 \pm 0.16}$ (in units of meV) (goodness of fit: $\chi^2 = 0.101$) and $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1} = (1.88 \pm 0.07) \times 10^{-3} + (0.74 \pm 0.08) \times 10^{-3} T^{1.57 \pm 0.08}$ (in arbitrary units) ($\chi^2 = 0.107$). Thus, it is justified to conclude that the critical exponent of the inverse susceptibility and the energy width as determined by neutron scattering experiments, $\alpha = (1.5 \pm 0.15)$, conforms to the theoretical value 1.5 expected for a QCP of the itinerant 3D SDW type [2–4]. The product $\chi(\mathbf{Q}_{\mathrm{AF}})\Gamma(\mathbf{Q}_{\mathrm{AF}})$ has a constant $T$ dependence to within 10% from lowest $T$ to $T = 5.0$ K, as expected for a paramagnetic heavy-fermion system [27].

A further test of the scaling behavior can be performed by directly plotting the imaginary part of the dynamical susceptibility $\chi''(\mathbf{Q}_{\mathrm{AF}}, \omega)$ multiplied by $T^{3/2}$ as a function of $\hbar\omega/(k_B T)^{3/2}$. If scaling as expected for QCP of the itinerant SDW type occurs, all data should conform to a single function $f(x) = a \times b \times x/[1 + (b \times x)^2]$ [28]. $\chi''(\mathbf{Q}_{\mathrm{AF}}, \omega)$ is determined by subtracting the incoherent elastic contribution from the measured neutron scattering intensity, under consideration of the Bose factor. As shown in Fig. 3(c), the data collapse on a single scaling curve of the expected form for $\hbar\omega \geq 0.2$ meV and $T < 5.0$ K, despite the small finite intercept in $\Gamma(\mathbf{Q})$.

The momentum dependence of the magnetic excitation spectrum around $\mathbf{Q}_{\mathrm{AF}}$ was measured, reaching the normal state in two different ways: either at $B = 0$ and $T = 1.0$ K, that is, $T > T_c$, or at $T = 0.06$ K and $B = 1.7$ T, that is, $B \approx B_{c2}$. Figure 4(a) displays measurements under the latter conditions at different energy transfers. Results under the former conditions are analogous. As in the SC state [24], with increasing energy transfer the signal is split into two peaks of Gaussian line shape, which broaden and move further apart in $\mathbf{Q}$, accompanied by a decrease in intensity. Figure 4(b) shows the linear dispersion of the magnetic excitation starting at $\hbar\omega = 0$ in the normal state in comparison to the dispersion in the SC state, where an

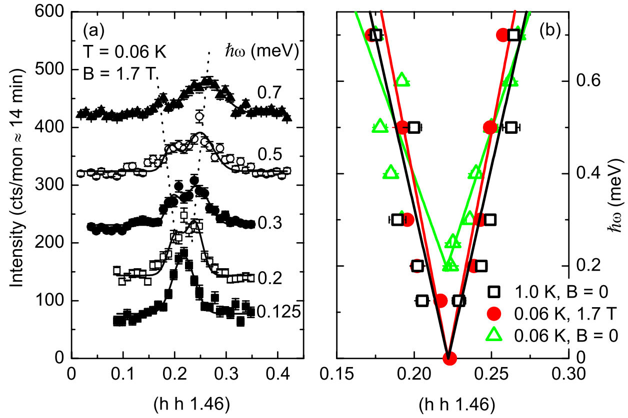


FIG. 4 (color online). (a) Momentum dependence of the magnetic response around $\mathbf{Q}_{\mathrm{AF}}$ in $S$-type $CeCu_2Si_2$ for different energy transfers $\hbar\omega$ at $T = 0.06$ K, $B = 1.7$ T. The scans are shifted by 100 counts with respect to each other. Solid lines indicate fits with two Gaussian peaks; dashed lines are guides to the eye. (b) Spin fluctuation dispersion as derived from fits to $\mathbf{Q}$ scans in the normal state at $B = 0$, $T = 1.0$ K or at $T = 0.06$ K, $B = 1.7$ T as well as in the SC state at $T = 0.06$ K, $B = 0$. Solid lines indicate linear fits.

energy gap $\hbar\omega_{\mathrm{gap}} \approx 0.2$ meV is observed. The resulting mode velocities are $v = (7.08 \pm 1.86)$ meV Å ($T = 0.06$ K, $B = 1.7$ T) and $v = (6.75 \pm 1.69)$ meV Å ($T = 1.0$ K, $B = 0$). This is about 50% larger than in the SC state [$v_{\mathrm{sc}} = (4.44 \pm 0.86)$ meV Å] but still significantly smaller than the Fermi velocity $v_F \approx 57$ meV Å [29]. This indicates a retardation of the interaction between the heavy quasiparticles and the critical fluctuations, which is of great importance for Cooper-pair formation.

Several important conclusions can be drawn from the critical spin dynamics of $S$-type $CeCu_2Si_2$. First of all, the slowing-down and the scaling behavior of the critical spin fluctuations are in line with predictions of the Hertz-Millis-Moriya theory, which shows that the QCP in $CeCu_2Si_2$ is of SDW type with 3D AF fluctuations. The finite values of $\chi(\mathbf{Q}_{\mathrm{AF}})^{-1}$ and $\Gamma(\mathbf{Q}_{\mathrm{AF}})$ for $T \rightarrow 0$ indicate that the $S$-type crystal is positioned slightly on the right side of the QCP in the $T$-$g$ phase diagram of $CeCu_2Si_2$ [cf. Figure 3(c)]. This interpretation is corroborated by an analysis of specific heat data, where $C(T)/T$ shows the expected NFL behavior for $T \leq 2.0$ K, before a crossover to Fermi-liquid behavior occurs, and $C(T)/T$ becomes constant for $T < 0.3$ K.

Furthermore, we have shown that, in $S$-type $CeCu_2Si_2$, $\mathbf{Q}_{\mathrm{AF}}$ is the origin of a damped dispersive mode, which exhibits an energy gap in the SC state, whereas the mode is gapless in the normal state. The magnetic response is independent of how the normal state has been reached, either by applying a magnetic field $B \approx B_{c2}$ or by elevating the temperature to $T > T_c$. The mode velocity is the same in both cases and somewhat larger than the initial velocity in the SC state.

In summary, inelastic neutron scattering data complemented by heat capacity measurements on $S$-type $CeCu_2Si_2$ show that 3D AF fluctuations occurring in the vicinity of a conventional QCP of SDW type can be traced from the normal to the SC state, where they persist above an energy gap $\hbar\omega_{\mathrm{gap}} = 3.9 k_B T_c$. As for the coupling mechanism of the superconductivity in $CeCu_2Si_2$, these results speak strongly in favor of the paramagnon scenario as proposed by Scalapino, Loh, and Hirsch [10] instead of the exciton scenario suggested for $UPd_2Al_3$ or $CeCoIn_5$.

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