Anti-ferromagnetic (AF) heavy-fermion (HF) metals frequently exhibit a quantum critical point (QCP), i.e. a continuous disappearance of AF order at zero temperature as a function of a non-thermal control parameter, like pressure. Commonly, unconventional ($d$-wave) superconductivity (SC) is observed in the vicinity of such a QCP. In this article, the interplay between quantum criticality and SC in HF metals is discussed. The latter can be well described in the framework of the Kondo lattice (KL) model. After introducing HFs and giving an overview on HF superconductivity, the characteristic energy scales in KL systems are addressed. Subsequently, we concentrate on the isostructural compounds CeCu$_2$Si$_2$ and YbRh$_2$Si$_2$. CeCu$_2$Si$_2$ is a prototypical HF superconductor which shows a QCP of itinerant, i.e. three-dimensional (3D) spin-density-wave (SDW) type. Overdamped, nearly quantum-critical SDW fluctuations are concluded from inelastic-neutron-scattering results to be the driving force for SC ($T_c \approx 0.6$ K) in this compound. While the QCP in several other Ce-based HF superconductors is presumably of the SDW variety too, it is likely to be of the local, i.e. Kondo destroying type in the pressure-induced superconductor CeRhIn$_5$. YbRh$_2$Si$_2$ has been identified as a prototype HF metal exhibiting such a Kondo breakdown QCP, often named a ($T = 0$) 4f-orbital selective Mott transition. This compound is not a superconductor at $T > 10$ mK. Systematically searching for and studying SC near Kondo destroying QCPs may offer a link to unconventional SC in other families of correlated materials, such as the doped Mott insulators in the high-$T_c$ cuprates and some of the organic charge-transfer salts.

**Keywords:** heavy fermions; quantum criticality; superconductivity

1. Kondo lattice and heavy fermions

Certain Ce- and Yb- as well as U-, Pu- and Np-based intermetallics form the class of heavy-fermion (HF) metals. The rare-earth-based members are commonly considered Kondo lattice (KL) systems [1]. They exhibit a characteristic crossover scale (determined by the Kondo temperature, $T_K$, typically a few 10s of Kelvin), which separates a high-temperature ($T$) regime of local 4f-derived magnetic moments from a low-$T$ regime which gives rise to the emergence of a number of different ground-state properties. While the localized 4f states at $T > T_K$ are not part of the “small” Fermi surface, they become entangled with the delocalized conduction-band states below $T_K$. Here,
so-called Kondo singlets are formed locally. They are inferred from, upon cooling, a continuous reduction of the 4f-derived magnetic moment and may even lead to a complete quenching at $T << T_K$ [1].

In a KL system, the Kondo singlets develop dispersion owing to the periodic arrangement of the rare-earth ions (Bloch’s theorem). Therefore, they take the role of (composite) charge carriers, with the same quantum numbers as the conduction electrons. Because of the strong Coulomb correlations on the localized 4f shell, their mobility is very poor, i.e. their Fermi velocity is about 1000 times smaller than that of uncorrelated conduction electrons. As a consequence the composite charge carriers have a correspondingly large effective mass $m^*$ and are frequently called “heavy fermions” (HFs). HF phenomena were discovered with CeAl₃, whose specific heat ($C = \gamma(T)$) and electrical resistivity ($\rho = \rho_0 + AT^2$, $\rho_0$: residual resistivity) indicate a non-magnetic “heavy Fermi liquid” low-$T$ state [2]. The Sommerfeld coefficient of the huge electronic specific heat of CeAl₃, which is practically identical with the measured one, amounts to $\gamma \approx 1.6 \text{ J/K}^2\text{mole}$ – as compared to $\gamma \approx 0.0007 \text{ J/K}^2\text{ mole}$ for Cu.

In other HF metals, like CeAl₂ [3] and anti-ferromagnetic (AF) order forms at low temperature. Usually, the magnetic-ordering temperature can be suppressed by varying a non-thermal control parameter, e.g. chemical doping, pressure and magnetic field. This gives rise to a quantum phase transition between the ordered and the paramagnetic phases at absolute zero temperature. Once this transition is continuous, it denotes a quantum critical point (QCP). The violent critical fluctuations near the QCP are the origin of pronounced non-Fermi liquid (NFL) phenomena, i.e. unusual temperature dependences of physical quantities which are often observed up to surprisingly high temperatures [4]. An increasing number of HF metals, like Au-doped CeCu₆ [5] and YbRh₂Si₂ [6], are known to exhibit a NFL low-$T$ state.

In the vicinity of the QCP, there exists a large excess of entropy, which the system may succeed to remove by creating some novel, symmetry-broken phase. This way, unconventional SC is often found in HF metals near an AF QCP. A prototype system is CePd₂Si₂ whose pressure ($p$)-induced QCP at $p_c \approx 2.8 \text{ GPa}$ is masked by a narrow superconducting dome below $T_{c_{\text{max}}} \approx 0.4 \text{ K}$ – and in very clean samples only [7].

This paper is organized as follows. HFSC, the energy scales in HFs and HF quantum criticality are addressed in chapters 2–6, while the interplay of SC and quantum criticality is briefly alluded to in the final chapter.

2. HF superconductivity

The 1979 discovery of HFSC in CeCu₂Si₂ [8,9], see Figure 1, followed the discoveries of superfluidity in $^3$He [10] and HF phenomena in CeAl₃ [2]. It came as a big surprise because, at this time, SC and magnetism were considered antagonistic phenomena, as is best illustrated by the disastrous pairbreaking effect which isolated paramagnetic impurities have on SC. In any superconductor known before 1979, a tiny amount (not more than a few at %) of magnetic impurities is enough to suppress SC [11]. This originates in the spin-exchange scattering from the magnetic impurities, which break up the spin-singlet pairing of the Cooper pairs [12]. Obviously 100 at% of periodically arranged magnetic Ce$^{3+}$ ions in CeCu₂Si₂ are not pairbreaking. Rather, they appear to be prerequisite for SC to form: The non-magnetic reference compound LaCu₂Si₂ is not a
superconductor [8]. Furthermore, a small concentration of non-magnetic impurities is known to suppress the superconducting state [13].

From Figure 1b, it is inferred that the Sommerfeld coefficient of CeCu$_2$Si$_2$ as extrapolated from the normal-state-specific heat $C_n(T > T_c)$ to $T = 0$ is $C_n(T \to 0)/T \approx 1$ J/K$^2$mole, similar to $\gamma = 1.6$ J/K$^2$mole of CeAl$_3$ [2]. The jump $\Delta C/T_c$ at $T_c (\approx 0.6 \text{ K})$ is of the same gigantic size, which indicates that the Cooper pairs in CeCu$_2$Si$_2$ are formed by the heavy quasi-particles [8]. As their Fermi velocity is extremely small, i.e. only of the same order as the velocity of sound [14], electron-phonon coupling turns out to be unretarded and, thus, unapt to mediate the Cooper pair formation. Therefore, CeCu$_2$Si$_2$ was the first so-called unconventional (non-phonon mediated) superconductor. By the mid of the 1980s magnetic pairings were proposed [15,16] for this compound as well as its subsequently discovered U-based counterparts UBe$_{13}$ [17], UPt$_3$ [18], U$_2$PtC$_2$ [19] and URu$_2$Si$_2$ [20].

The early investigations on CeCu$_2$Si$_2$ polycrystals were severely plagued by the fact that physical properties varied considerably from sample to sample, see Figure 1b. Those sample dependences added to the reservations of the community about the discovery of bulk SC in CeCu$_2$Si$_2$. However, this skepticism was overcome in 1984 when it was demonstrated that high-quality single crystals of CeCu$_2$Si$_2$, when compared with the polycrystals [8], showed even more pronounced superconducting signatures [21]. The origin of the afore-mentioned sample dependences was unravelled several years later by thorough studies of the chemical Ce-Cu-Si phase diagram [22]. Different, closely spaced ground-state properties were found to exist within the very narrow homogeneity of the tetragonal 122 phase (ThCr$_2$Si$_2$ structure): AF order which – at very low temperatures – coexists with SC (“A-type” samples), SC which competes with and expels AF order (“A/S-type”), and SC only (“S-type”) [23].
The number of known HF superconductors has increased from 5 in 1984, to 11 in 1998 and about 40 at present [24]. A variety of unconventional pairing mechanisms, including AF [7] and ferromagnetic (FM) [25] spin fluctuations, FM magnons [26] and even electric quadrupole fluctuations [27] have been proposed for these materials. Both CeCu$_2$Si$_2$ [28] and its Ge-homologue [29] present a unique case, as they exhibit SC up to very high pressures. By substituting 10 at% Ge for Si in the former material, $T_c$ could be substantially depressed. This way, two separate superconducting “domes” were resolved in different pressure ranges (Figure 2). The low-$p$ dome centred around $p \approx 0$ masks an AF QCP, similar to what was discovered for CePd$_2$Si$_2$ [7]. The second dome at high pressure is intersected by a weak first-order valence transition line, with a surprisingly low-lying critical end point of 10–20 K [30]. As will be discussed in chapter 4, SC under the low-$p$ dome is due to nearly quantum critical AF spin fluctuations, while it was proposed [31] that another unconventional pairing mechanism implying nearly quantum critical valence (or charge) fluctuations, is operating at high pressures.

Typically, HF superconductors exhibit non-exponential temperature dependences, often simple power laws, of the specific heat and related quantities [1]. From the bulk of available experimental data, it was concluded [9] that most of these systems show a spin-singlet ($S = 0$) or even-parity Cooper pair state. Its shape, however, is not of

![Figure 2. (colour online) Phase diagram of CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ showing transition temperatures into the anti-ferromagnetically ordered ($T_N$, open symbols) and the superconducting state ($T_c$, closed symbols) vs. relative pressure $\Delta p = p - p_c(x)$, which reflects the inverse volume. The $p_c(x)$ values are chosen so that the magnetic transition lines for $x = 0.1$ ($p_c = 1.5$ GPa, circles) and $x = 0.25$ ($p_c = 2.5$ GPa, squares) coincide. Pure CeCu$_2$Si$_2$ is assumed here to have $p_c \approx 0.4$ GPa (after [28]).](image-url)
s-wave ($L = 0$) type, but highly anisotropic. Most likely, these are d-wave ($L = 2$) superconductors.

A small number of HFSCs, i.e. UPt$_3$ [32], UNi$_2$Al$_3$ [33] and the ferromagnets UGe$_2$ [34], URhGe [35] and UCoGe [36] are prime candidates for spin-triplet ($S = 1$) or odd-parity pairing.

During the last decade, superconductors lacking inversion symmetry have attracted considerable interest [37]. Here, the anti-symmetric spin-orbit coupling may lead to an admixture of odd-parity to even-parity pair states [38,39]. This novel type of unconventional SC was even observed in “classical”, i.e. phonon-mediated, superconductors [40]. For some of the HF superconductors, like UPt$_3$ [41], U$_{1-x}$Th$_x$Be$_{13}$ [42] and CeCoIn$_5$ [43], multiphase diagrams containing different superconducting phases were discovered, resembling the different superfluid phases of $^3$He [44].

3. Energy scales in heavy fermions

In Ce-based HF systems, the Kondo exchange interaction is increased by the application of (external or chemical) pressure. Simultaneously, the magnetic order becomes weakened. This “demagnetization of the 4f shell” originates in the different ionic radii of the competing Ce$^{3+}$ (4f$^1$) and Ce$^{4+}$ (4f$^0$) configurations. In the former case, the positive nuclear charge is partially screened by the inner 4f shell; in the latter one, it is not leading to a strengthening of the attraction on the outer core (valence) electrons. Due to the 4f electron-hole analogy between Ce and Yb, application of pressure to Yb-based HF systems stabilizes magnetic order and simultaneously weakens the Kondo effect. When investigating quasi-binary alloys of Ce and La or Yb and Lu, the increase in the Ce (or Yb) concentration has a twofold effect: (i) an increase of magnetic inter-site correlations and (ii) a compression (or expansion) of the average unit-cell volume. The effect of chemical pressure has been studied on (moderately) dilute [(La$_{1-x}$Y$_x$)$_{1-y}$Ce$_y$]Al$_2$ alloys with fixed Ce concentrations [45]. Here, the low-temperature resistivity is well described by $\rho(T) = \rho_0 - A T^2$, with $A = \pi^2/4 T_K^2$ [45], indicating local Fermi liquid behaviour. Upon increasing Y-concentration, the coefficient $A$ becomes gradually smaller and the local Fermi liquid regime expanded. The corresponding rise of $T_K$ as a function of increasing volume compression is displayed in Figure 3: $T_K$ increases from $\approx 0.35$ K for dilute (La, Ce)Al$_2$ to $\approx 100$ K for dilute (Y, Ce)Al$_2$, i.e. by more than a factor of 250 [45]. Although, as inferred from Figure 3, the Ce–Ce inter-site interactions are not fully negligible, the compression of the volume is the key factor in determining $T_K$. This is supported by the fact that the single-ion Kondo temperature of the AF KL ($T_N = 3.9$ K) CeAl$_2$ is $T_K \approx 5$ K, as determined from, e.g. the residual magnetic quasi-elastic neutron line width [46]: in fact, for CeAl$_2$ ($\nu \approx 0.3$) one reads off Figure 3 $T_K = (5 \pm 2)$ K. This value exceeds $T_K = 0.35$ K of dilute (La, Ce)Al$_2$ by a factor of $\approx 13$.

A similar change of $T_K$ was recently concluded from specific-heat measurements on (Ce,La)Ni$_2$Ge$_2$ alloys [47]. As shown in Figure 4, $T_K$ varies from $\approx 1.5$ K for (Ce$_{0.01}$La$_{0.99}$)Ni$_2$Ge$_2$ to $\approx 28$ K for the stoichiometric compound CeNi$_2$Ge$_2$. These $T_K$ values are compared in the figure with the temperatures $T_{max}$ at which a peak occurs in the low-$T$ thermoelectric power (TEP) of (Ce$_{1-x}$La$_x$)Ni$_2$Ge$_2$. While $T_{max}$ agrees well with $T_K$ in both the local Fermi liquid regime ($0.98 \leq x \leq 1$) as well as for CeNi$_2$Ge$_2$, it is smaller than $T_K$ for $0.05 \leq x \leq 0.5$. This inequality holds, in particular, between 60
and 95 at% Ce, where the specific-heat data indicate coherent Fermi liquid behaviour well below $T_K$. The results of [47] can be summarized as follows:

1. The position of the low-$T$ TEP peak is determined by the dominating low-energy scale, e.g. the single-ion $T_K$ for isolated Kondo impurities.
2. When spatial coherence develops at low temperatures upon the onset of (2D) percolation, $T_{\text{max}}$ is related to the coherence temperature, $T_{\text{coh}}$. Upon cooling to below $T_{\text{coh}}$, a (partial) hybridization gap forms within the renormalized (quasi-particle) DOS near $E_F$, reflected by a peak in the TEP, $S(T)$.
3. In the presence of alloying-induced disorder which weakens the coherence, $T_{\text{coh}} < T_K$, whereas in the stoichiometric KL system CeNi$_2$Ge$_2$ both energy scales are almost degenerate – as also observed for the isostructural compound CeCu$_2$Si$_2$ [48].

It is interesting to also study the transport properties of the (Lu$_{1-x}$Yb$_x$)Rh$_2$Si$_2$ system [49]. For all $x$, $\rho(T)$ and $S(T)$ exhibit a broad maximum at $T_S^{\text{high}} = (80 - 100)$ K. At smaller Yb concentrations, $0 < x < 0.5$, additional maxima occur at lower temperature, cf. Figure 5(a). The positions of the low-$T$ maxima, $T_K$, are plotted as a function of the Yb-concentration $x$ in Figure 5(b). While $T_S^{\text{high}}(x)$ is found to be $(90 \pm 10)$ K,
independent of \( x \), the Kondo temperature \( T_K \) (referring to the CF Kramers doublet ground state) shows a linear increase, from \( T_K \approx 10 \text{ K} \) for dilute \((\text{Lu}, \text{Yb})\text{Rh}_2\text{Si}_2\) to \( T_K \approx 30 \text{ K} \) for \( \text{YbRh}_2\text{Si}_2 \).

Figure 6 displays results of Scanning Tunneling Spectroscopy (STS) on \( \text{YbRh}_2\text{Si}_2 \) [50]. Apart from the almost symmetric conductance minimum at zero bias which shows up below \( T \approx 100 \text{ K} \) and is due to the on-site Kondo screening of \( \text{Yb}^{3+} \), implying all possible CF states, maxima are found at \(-43, -27 \text{ and } -15 \text{ meV} \). They indicate the CF splitting, i.e. the energetic differences between the excited Kramers doublets and the
lowest-lying one. In addition, when cooling to below $T \approx 30$ K another maximum occurs at $-6$ meV, which cannot be ascribed to either the single-ion Kondo effect or CF splitting [50]. This has been considered the signature of the (partial) hybridization gap which forms below $T_{\text{coh}}$. It was concluded that in undoped YbRh$_2$Si$_2$, $T_{\text{coh}}$ coincides with the single-ion $T_K$ (again referring to the CF doublet ground state) [50], as also observed for the isostructural HF metals CeCu$_2$Si$_2$ [48] and CeNi$_2$Ge$_2$ [47].

The message to be learned from these three “122”-type KL systems is therefore: the on-site Kondo screening predominates the physical properties in a wide temperature range as was recently also demonstrated for CeCu$_2$Si$_2$ by Nernst effect studies [51]. Upon cooling, it sets in at some elevated temperature of order (80–100) K, where all CF levels are still populated, according to the Boltzmann distribution function. Further cooling results in an increasing depopulation of the excited CF states, reflected by a decrease of the electrical resistivity $\rho(T)$. Once at $T \approx T_K$ all Kondo scattering centres are identical (i.e. residing in the CF-derived Kramers doublet ground state), spatial coherence starts to develop at $T_{\text{coh}} \approx T_K$.

4. Quantum criticality in heavy fermions

As already mentioned, the rare-earth-based HF metals can be considered KL systems. The latter are well described by the “Doniach phase diagram” [52], shown in Figure 7. There exists a critical value $J_c$ of the exchange constant, at which long-range magnetic order (in most cases AF order) disappears as a function of a suitable external control parameter, like pressure. If the $T = 0$ phase transition at $J = J_c$ driven by this control parameter, is continuous, $J_c$ denotes a QCP. It creates an abundance of violent quantum critical fluctuations, which are the source of NFL-type temperature dependences in thermodynamic and transport properties [4].

Two different variants of AF QCPs have been established theoretically as well as experimentally with HF metals, depending on the behaviour of the composite, heavy charge carriers upon approaching the QCP. If the heavy quasi-particles, defined on the paramagnetic side of the Doniach phase diagram well below $T_K$, exist also in the AF state, if only in the very vicinity of the QCP, there cannot be local-moment order.
Rather itinerant, i.e. spin-density-wave (SDW), order occurs which derives from a nesting instability of the renormalized Fermi surface. Since at this SDW QCP, which had been explored by theorists in great detail [53–55], the composite quasi-particles stay intact, the only degrees of freedom that matter are the fluctuations of the magnetic order parameter. Further on, as these charge carriers contain 4f states, the latter contribute to a large Fermi surface. The 3D SDW QCP scenario may be called “conventional”, since the Sommerfeld coefficient $\gamma (\sim m^*)$ stays finite at the QCP, see below.

In the alternative QCP scenario the composite quasi-particles apparently exist only on the paramagnetic side of the Doniach phase diagram, where the Fermi surface is large (Figure 7). Upon approaching the AF QCP, the Kondo singlets break up: the magnetic order is then due to (strongly Kondo screened) local moments, in the presence of a small Fermi surface – formed by the (s, p, d) conduction electrons only.

Theory predicts an overlap of a continuous low-dimensional AF quantum phase transition with an abrupt Fermi-surface reconstruction to occur at such a “Kondo breakdown” QCP, or (T = 0) 4f-selective Mott transition [56,57]. In the subsequent chapters, we shall discuss the evidence for either of the two scenarios and the possibility of unconventional SC in the vicinity of the QCP.

5. SDW QCP in CeCu$_2$Si$_2$

A substantial fraction of the 40 known HF superconductors show a low-$T$ NFL normal state – due to a nearby AF QCP. Neutron-diffraction studies on an A-type CeCu$_2$Si$_2$ single crystal (cf. Chapter 2) revealed HF-SDW order in the vicinity of the QCP [58]. Pronounced NFL temperature dependences of the resistivity and specific heat in the field-driven low-temperature normal state of S-type CeCu$_2$Si$_2$ [59] are consistent with 3D SDW fluctuations [53–55]. This was recently confirmed by inelastic-neutron-scattering (INS) results. They highlight a universal scaling for the dynamical
susceptibility at \( \tilde{q} = \tilde{Q} \), the incommensurate ordering wave vector, following 
\[ \chi T^{3/2} = f \left[ \hbar \omega/(k_B T)^{3/2} \right] \] \[60\].

In order to investigate the interplay between SC and AF order \[61\], neutron-scattering measurements were performed on an A/S-type CeCu_{2}Si_{2} single crystal in which the AF and the superconducting transitions almost degenerate, i.e. \( T_N \approx T_c \approx 0.7 \) K \[62\]. These measurements illustrate that AF order and SC (deriving from the same 4f electrons) compete and do not coexist in the same volume. Rather one finds phase separation into superconducting and AF volumes.

To probe the magnetic response in S-type CeCu_{2}Si_{2} by high-resolution INS both in the normal and the superconducting state \[48,60\], the momentum and energy dependence of the spin excitations were measured in the neighbourhood of the ordering wave vector \( \mathbf{Q} \), as determined in an anti-ferromagnetically ordered A-type CeCu_{2}Si_{2} single crystal \[58\].

Energy scans have been recorded at the ordering wave vector \( \mathbf{Q} \) (in the vicinity of the nuclear Bragg reflection (0 0 2)) in the normal as well as superconducting states of S-type CeCu_{2}Si_{2} \[48,60\]. The measurements in the superconducting state were conducted at \( B = 0 \) and \( T \) well below \( T_c \) (0.07 K). At this same temperature, the normal state could be studied by applying a magnetic field of \( B = 2 \) T which exceeds \( B_{c2}(0) \approx 1.7 \) T. The response consists of a strongly field-dependent, incoherent elastic signal and the magnetic response, cf. Figure 8(a). The normal state fluctuations reveal a magnetic quasi-elastic line, which is well described by a simple Lorentzian, multiplied by the Bose factor (dashed line). The response in the superconducting state turns out to be gapped, i.e. spectral weight to be transferred from lower energies to higher energies above the gap of the spin excitations, \( \hbar \omega_{\text{gap}} \). The latter is \( \hbar \omega_{\text{gap}} \approx 0.2 \) meV at \( T = 0.07 \) K, which corresponds to \( \hbar \omega_{\text{gap}} \approx 3.9 \) k_{B}T_{c} and is roughly 10% below the value of the superconducting gap in the electronic DOS, predicted for a weak-coupling

Figure 8. (colour online) Inelastic neutron scattering of S-type CeCu_{2}Si_{2}. (a) Magnetic response \( S_{\text{mag}} \) (on an absolute intensity scale) at the AF ordering wave vector in the superconducting and normal states at \( T = 0.07 \) K (from \[48\]). (b) Spin-fluctuation dispersion in the normal state (at \( B = 0, T > T_c \) and \( B = B_{c2} = 1.7 \) T, \( T \ll T_c \)) as well as in the superconducting state (\( B = 0, T \ll T_c \)) (from \[60\]).
d-wave superconductor [63], and about 20% smaller than $5 \kBT_c$ as obtained from Cu-NQR results [64,65]. The spin gap becomes smaller upon warming and closes at $T_c$. The symmetry of the superconducting order parameter has been determined by in-plane isothermal ($T = 0.04$ K) magneto-resistance measurements, performed on an S-type CeCu$_2$Si$_2$ single crystal, to be of $d_{xy}$ type [66]. However, this has been questioned by recent high-precision measurements of the low-T specific heat of S-type CeCu$_2$Si$_2$; they reveal a fully gapped superconducting ground state [67].

Figure 8 shows the wave-vector dependence of the magnetic response in the superconducting state [60]. The spin excitations near the AF ordering wave vector $Q$ belong to an overdamped dispersive mode with a velocity being smaller by almost an order of magnitude than the extremely small renormalized Fermi velocity [14].

Clearly, the coupling of the HFs to the spin excitations is retarded, which implies that the direct Coulomb repulsion between the even charges of the composite quasi-particles can be avoided. Therefore, these spin excitations are well suited to act as superconducting glue in CeCu$_2$Si$_2$. This conclusion is corroborated by the difference in magnetic exchange energy between the superconducting and the normal state, which was calculated in [48]. The resulting gain in exchange energy is much larger (by more than a factor of 20) compared to the superconducting condensation energy [48]. This underlines that the spin excitations are driving the formation of Cooper pairs in CeCu$_2$Si$_2$. It also implies that there must be a correspondingly large loss of kinetic energy, which has been interpreted as the result of Mott-like physics associated with the Kondo effect [48,61].

To be able to study the spin fluctuations in the normal state of CeCu$_2$Si$_2$ when tuning the system towards the QCP, SC was suppressed in a magnetic field $B = 1.7$ T ($\approx B_{c2}$). When the magnetic response in the normal state at the ordering wave vector $Q$ is studied upon increasing temperature, it is found to remain quasi-elastic, but to broaden and decrease in intensity [60]. This means that the magnetic response shows a pronounced slowing down upon cooling. The temperature dependence of the spin fluctuations is in full agreement with the expectations for a 3D SDW QCP [48].

However, there is no critical slowing down, as one extrapolates to a finite correlation time in the zero-temperature limit. This is due to the fact that the S-type CeCu$_2$Si$_2$ single crystal is located on the paramagnetic of, but very close to, the QCP. Therefore, the broadly distributed dynamical SDW correlations that build up the quasi-elastic response, may be called nearly quantum critical spin fluctuations.

6. Kondo destroying QCP in YbRh$_2$Si$_2$

AF quantum criticality beyond the SDW scenario has been concluded first for CeCu$_{6-x}$Au$_x$ at its critical concentration $x_c = 0.1$, above which long-range AF order develops [5]. Its dynamical susceptibility exhibits energy over temperature scaling, with a fractional exponent $\alpha \approx 0.75$ [68]. This cannot be explained by spin fluctuation theory, neither for a 2D nor 3D SDW scenario [53–55]. The results of [68] have motivated new theoretical concepts for quantum criticality, which involve a breakdown of the Kondo effect and, thus, an abrupt change of the Fermi surface at the AF QCP [56,57].

Direct studies of Fermi surface properties are usually done by either angle-resolved photoemission spectroscopy (ARPES) or measurements of magnetic quantum oscillations, e.g. the de Haas-van Alphen (dHvA) effect. At present, ARPES is confined to temperatures $T > 1$ K and cannot achieve the energy resolution necessary to explore HF
quantum criticality. dHvA experiments, on the other hand, require the application of magnetic fields of order several Teslas. In most cases, such fields severely affect the HF nature and suppress any quantum critical fluctuations. An exception is provided by CeRhIn$_5$. This compound exhibits a pressure-induced QCP at $p_c \approx 2.3$ GPa. The latter becomes visible when a magnetic field $B > B_{c2}$ ($T \rightarrow 0$) = 9.2 T is applied, at which SC is suppressed [70]. The results of the dHvA measurements, performed down to $T = 80$ mK and in the field range 10–17 T, indicate an abrupt change of the Fermi surface right at the QCP, $p_c \approx 2.3$ GPa [70], cf. Figure 9(a). Taken together with the observation of an incipient divergence of the cyclotron mass at $p = p_c$ (Figure 9(b)), these dHvA results are highly consistent with a Kondo breakdown QCP [70], although they have been interpreted in terms of a valence crossover as well [71].

A discontinuous Fermi surface reconstruction could be observed at the field-induced QCP of YbRh$_2$Si$_2$ ($B_N \approx 0.06$ T, $\perp$ c-axis). As shown in Figure 10(a) and (b) for, respectively, $B \parallel c$ and $\perp c$, the hard magnetic axis, NFL-type $T$-dependences can be studied at $B = B_N \approx 0.6$ T down to the lowest temperature $T \approx 20$ mK [72]. For an YbRh$_2$Si$_2$ single crystal, doped with a tiny amount of Ge, this holds true even at $B = 0$. Figure 11(a) and (b) show the temperature dependences of the Sommerfeld coefficient of the electronic specific heat $\gamma = C_{el}/T$ and the electrical resistivity $\rho$ for pure YbRh$_2$Si$_2$ and the Ge-doped crystal [72]. While the Néel transition at $T_N = 70$ mK is visible in both $\gamma(T)$ and $\rho(T)$ for the stoichiometric compound with residual resistivity $\rho_0 \approx 1\mu\Omega$cm, in case of YbRh$_2$(Si,Ge)$_2$ a very weak AF phase-transition anomaly is resolved at $T_N \approx 20$ mK in $\gamma(T)$ only. $\rho(T)$ exhibits linear behaviour over three decades, from $T = 10$ mK to $\approx 10$ K, for this doped crystal with substantially enhanced $\rho_0$ ($\approx 5 \mu\Omega$cm). With both samples, $\gamma(T)$ is found to diverge logarithmically upon cooling to below $T = 10$ K, as expected – in view of $\Delta \rho \sim T$ – in case of a wave-vector independent self energy [73]. However, below $T \approx 0.3$ K, the divergence of $\gamma(T)$ becomes

![Figure 9](https://example.com/figure9.png)

**Figure 9.** (colour online) Changes of Fermi surface properties across a likely Kondo destroying QCP in CeRhIn$_5$. Pressure dependencies of the dHvA frequencies (a) and cyclotron mass (b) (after [70]).
Figure 10. (colour online) Temperature-magnetic field phase diagram of YbRh$_2$Si$_2$ for $B \parallel c$ (a) and YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$ for $B \perp c$ (b) (from [72]).

Figure 11. (colour online) Disparate behaviours of Sommerfeld coefficient $C_v/T$ vs. $T$ (a) and electrical resistivity $\rho$ vs. $T$ (b) for YbRh$_2$(Si$_{1-x}$Ge$_x$)$_2$. Results for pure YbRh$_2$Si$_2$ are also shown (from [72]).
stronger than logarithmic. It follows a power law, \( \gamma \sim T^{\epsilon} \), \( \epsilon \approx 1/3 \). The disparate \( T \)-dependences of \( \rho(T) \) and \( \gamma(T) \) below \( T = 0.3 \) K were considered evidence of a breakup of the composite quasi-particles at the AF QCP [72].

The finite-\( T \) signature of this breakdown of the Kondo effect could be identified for YbRh\(_2\)Si\(_2\) by isothermal measurements of the initial Hall coefficient \( R_{\text{H}}(B) \), with \( R_{\text{H}} \) probing solely Fermi surface properties below \( T = 1 \) K [74]. At \( B(T = T^*) \), \( R_{\text{H}}(B) \) undergoes a thermally broadened, substantial change [74]. More recent results of isothermal measurements of \( R_{\text{H}}(B) \) and the longitudinal magneto-resistivity on YbRh\(_2\)Si\(_2\) crystals of very different quality clearly revealed that the crossover line \( T^*(B) \) extrapolates (as \( T \to 0 \)) to \( B = B_N \), the AF QCP [75] (see Figure 12(a)). In addition, the width of the Hall crossover was found to be proportional to temperature, \( FWHM \sim T \) [75] (Figure 12(b)), indeed demonstrating an abrupt jump of \( R_{\text{H}}(B) \) at the AF QCP (Figure 12(c)). These results verify the theoretical prediction [56,57] of a Kondo destroying QCP to concur with an AF one. Further support for this stems from low-temperature measurements of the thermoelectric power, which shows a change of sign, from hole to electron like, when increasing the magnetic field across \( B(T = T^*) \) [76].

Thorough investigations of various thermodynamic and transport properties of YbRh\(_2\)Si\(_2\) confirmed \( T^*(B) \) to signal an intrinsic energy scale, hitherto unknown in KL systems [77]. This scale is distinct from both the AF phase boundary \( T_N(B) \) and the Fermi liquid crossover scale \( T_{\text{FL}}(B) \), below which \( \Delta \rho = (\rho - \rho_0) \sim T^2 \).

According to the \( R_{\text{H}}(B) \) results at the lowest accessible temperature, \( T = 20 \) mK, the Hall coefficient changes from a relatively large positive value at \( B < B_N \) to a small negative one at elevated fields [74]. When comparing this observation with results from LDA and renormalized band-structure calculations [78], it turns out to be indeed consistent with a field-induced change from a smaller to a larger charge-carrier concentration – as expected [56,57].

One fundamental question left open in the theoretical and experimental literature cited above concerns the dynamical processes associated with the breakup of the Kondo singlets at the AF QCP of YbRh\(_2\)Si\(_2\). For example, how do the composite quasi-particles (defined in the low-\( T \) paramagnetic state) behave when approaching the Kondo destroying QCP? A well-established way to study the behaviour of quasi-particles is to perform combined measurements of charge and heat transport: In a metal at \( T = 0 \) (where all scatterings are elastic), the Wiedemann Franz (WF) law holds. This means that the Lorenz number \( L = \rho/(\kappa \pi T^2) \) becomes \( L_0 = (\pi k_B)^2 / 3 e^2 \) as \( T \to 0 \). Here \( \kappa(T) \) is the thermal conductivity, \( k_B \) Boltzmann’s constant, \( e \) the elementary charge and \( L_0 \) Sommerfeld’s constant. If one defines the thermal resistivity \( w(T) \) such that it is measured in the same unit as its electrical counterpart \( \rho(T) \), i.e. \( w = L_0 T / \kappa \), the WF law states that for \( T \to 0 \), \( L(T)/L_0 = \rho(T)/w(T) = \rho_0/w_0 = 1 \), where \( \rho_0 \) and \( w_0 \) denote the residual electrical and thermal resistivities.

Recently, temperature sweeps of both \( \rho \) and \( \kappa \) were taken for YbRh\(_2\)Si\(_2\) (\( \rho_0 \approx 1 \) \( \mu \Omega \) cm) at \( T \leq 0.5 \) K as well as \( B = 0 \) and differing values of the control parameter \( B \) [79]. One recognizes that \( w(T) > \rho(T) \) in a wide parameter space, except at the highest field (\( B = 1 \) T) and the lowest temperatures, where \( \rho/w = 1 \) within the experimental uncertainty. As expected, the WF law is valid in the field-induced paramagnetic phase. At magnetic fields \( B \leq B_N \approx 0.06 \) T, a pronounced drop in \( w(T) \) is found below \( T \approx 0.1 \) K. For \( B = 0 \) and 0.02 T, \( w(T) \) becomes even smaller than \( \rho(T) \) below \( T \approx 30 \) mK. This clearly indicates an extra magnon contribution to the heat transport, adding to that
Figure 12. (colour online) Fermi surface collapse from isothermal crossover in magneto-transport for YbRh$_2$Si$_2$. (a) Position of crossover in temperature-field phase diagram for two different samples from crossed-field and single-field Hall coefficient as well as longitudinal magneto-resistivity. (b) Crossover width (FWHM) as a function of temperature. (c) Schematic illustration of abrupt change in $R_{\text{H}}(T)$ at $T = 0$ and thermal broadening of this jump at finite temperatures (from [75]).
by the electronic carriers. Correspondingly, the drop found in \( w(T) \) below \( T \approx 70 \) mK at \( B = 0.06 \) T was ascribed to the contribution of overdamped magnons (“paramagnons”), which exist in the paramagnetic state close to the AF order.

The identification of the extra heat carriers as magnon excitations of the AF phase appears to be justified by an earlier observation of a magnon contribution, \( C_m \sim T^3 \), to the \( B = 0 \) specific heat below \( T \approx 50 \) mK, i.e. inside the AF phase of YbRh\(_2\)Si\(_2\) [72]. In the asymptotic low-\( T \) regime, the magnon heat conductivity, \( \kappa_m(T) \), is proportional to the magnon specific heat. This means that \( \kappa_m \sim T^3 \) as to \( T \) goes to zero, where the thermal transport is exclusively due to the electronic carriers. Since the AF state of YbRh\(_2\)Si\(_2\) behaves as a heavy Fermi liquid [72], see Figure 10(a), the WF law must also hold in the zero-temperature limit at \( B < B_N \).

Figure 13 displays isothermal field scans of \( L/L_0 = \rho/w \) in the paramagnetic regime, \( 0.1 \) K \( \leq T \leq 0.4 \) K (the data at lower \( T \) are not shown because of the interfering magnon contribution). Each of these isotherms exhibits a minimum \( B_{\text{min}} \), where the violation of the WF law appears to be largest and, in turn, the inelastic scattering of the electronic heat carriers to be strongest. Upon cooling, \( B_{\text{min}} \) shifts to lower values, approaching \( B_N \). At the same time, the \( L(B)/L_0 \) minimum narrows considerably. I.o.w., this minimum tracks the evolution of the Hall crossover (cf. arrows and horizontal bars in Figure 14), and the electronic Lorenz ratio, \( L_{\text{el}}(B)/L_0 \), is concluded to extrapolate to an abrupt drop of \( \approx 10\% \) in the zero-temperature limit at \( B = B_N \) [79]. More recently, several groups have performed the same type of measurements on high-quality YbRh\(_2\)Si\(_2\) single crystals; their published data are similar to those discussed above. However, their conclusions disagree with those of Ref. 79, cf. [80].

This apparent violation of the WF law exactly at the field-induced AF QCP may be ascribed to electron-electron type of scatterings at \( T = 0 \). This implies a fermionic

![Figure 13](image-url)
nature of the scatterers which may be considered the “residue” of those inelastic scattering which, at finite temperature, cause the distinct minima in the isothermal $L(B)/L_0$ curves shown in Figure 13. These scatterings are naturally identified with the fluctuations of the Fermi surface (note that bosonic excitations show up at $T = 0$ as zero-point fluctuations only).

In YbRh$_2$Si$_2$, the multiple lines denoting the AF-ordering temperature $T_N(B)$, the Fermi liquid-NFL crossover temperature $T_{FL}(B)$ and the Fermi surface crossover temperature $T^*(B)$ merge at the same value of the control parameter: $B = B_N$. The question arises: What happens when some additional control parameter is varied? By choosing (positive and negative) chemical pressure as this second control parameter [81], the “global” phase diagram (at $T = 0$) shown in Figure 14 is obtained [82]. It displays that the AF order is stabilized (weakened) by means of volume compression (expansion), in line with the well-known fact that in Yb-based compounds magnetism is strengthened under pressure. While the Kondo breakdown critical field $B^*$ (where $T^* \to 0$) appears to follow this same trend, its volume dependence is much weaker than that of the magnetic critical field $B_N$.

In view of the substantial weakening of the Kondo interaction under volume compression [83], this minor dependence of $B^*$ on chemical pressure is quite surprising.

When the average unit-cell volume of YbRh$_2$Si$_2$ is reduced via partial substitution of Rh by the isoelectronic smaller Co, the AF QCP occurs at a $B_N$ value which exceeds $B^*$ considerably. This implies that $T^*$ is finite at the AF instability [82]. Under small volume expansion, i.e. via partial substitution of Rh by 2.5 at % of the isoelectronic larger Ir, $B_N$ and $B^*$ are still overlapping within the experimental accuracy, cf. Figure 14. Upon larger volume expansion (i.e. by doping with 6 at % Ir), a finite field range exists for $T \to 0$, in which the Yb-derived magnetic moments are neither ordering nor screened by the Kondo effect. According to [81], it defines a NFL phase with small Fermi surface, hitherto unknown in KL systems. Most likely, this new phase is of a metallic spin-liquid type.

Measurements under hydrostatic pressure revealed results which are consistent with those on the Co-doped YbRh$_2$Si$_2$ samples of similar average unit-cell volume [84]. This proves that the intersection of the $T_N(B)$ and $T^*(B)$ lines observed for 7 at %
Co-substituted YbRh$_2$Si$_2$ [81] originates in the alloying-induced volume compression rather than disorder. Recently, an abrupt Fermi surface reconstruction inside the AF order has been directly observed with the aid of dHvA measurements on CeRhIn$_5$ at high pulsed magnetic fields [85]. Very interestingly, the experimental “global” phase diagram of Co/Ir-doped YbRh$_2$Si$_2$ (Figure 14) has striking similarity to its theoretical counterpart [86,87].

7. Outlook

Many of the HF metals showing NFL phenomena at low temperatures are superconductors. As demonstrated with the prototypical HF superconductor CeCu$_2$Si$_2$, unconventional (presumably d-wave) SC develops near a conventional, i.e. 3D SDW QCP. Here, almost quantum critical SDW fluctuations could be identified as the glue which mediates the Cooper pair formation [48]. It is very likely that, in many of the NFL-type HF metals, e.g. CePd$_2$Si$_2$ [7] and UBe$_{13}$ [17,88], SC is of a similar origin as in CeCuSi$_2$.

SC in pressurized CeRhIn$_5$ [69,70] and in β-YbAlB$_4$ [89] occurs near an unconventional QCP; in the latter case, its nature has yet to be established. In case of CeRhIn$_5$, the afore-mentioned recent Fermi surface studies up to high pulsed magnetic fields [85], strongly support the pressure-induced QCP at low and moderate fields [69,70] to be of the Kondo destroying type. For YbRh$_2$Si$_2$, a canonical HF metal exhibiting a Kondo breakdown QCP, no SC exists at $T > 10$ mK [72]. Surprisingly, recent investigations at ultra-low temperatures reveal the formation of SC below $T_c = 2$ mK [90]. Exploring HF SC in the vicinity of such a ($T = 0$) t-orbital selective Mott transition in more detail offers a link to the unconventional SC in other classes of correlated materials, like the doped Mott insulators in organic charge-transfer salts and cuprates. For the nearly optimally doped cuprates, a collapse of the energy scale of the pseudo-gap and a destruction of the quasi-particles over the entire Fermi surface was found [91] – similar to what occurs at the Kondo breakdown QCP in YbRh$_2$Si$_2$. The interplay of quantum criticality and unconventional SC is of fundamental interest and will remain in the focus of condensed-matter research.

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