Strong electron correlations in magnetic systems

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Heavy-Fermion Metals

Strongly correlated electron systems can be well studied with certain rare-earth—or actinide-based intermetallics. At high temperatures, independent local magnetic moments are formed due to the strong repulsive Coulomb force between electrons on the partially filled 4*f*/5*f*-shells of these materials. This local interaction competes with the (upon cooling increasingly strong) coupling between the localized *f*-electrons and the delocalized ligand states. As a result, the local *f*-moments become progressively screened below the characteristic Kondo-lattice temperature T_K, a process that causes the formation of strongly renormalized ("composite-fermion") quasiparticles, consisting of a local *f*-part and some admixture of itinerant conduction-electron contributions. These "heavy electrons" or "heavy fermions" (HFs) exist in extended regions of parameter space and resemble the conduction electrons of simple metals. However, because of the strong intra-atomic Coulomb correlations, they acquire huge effective carrier masses m^* ($\approx 100 - 1000\ m_{electron}$) [1].

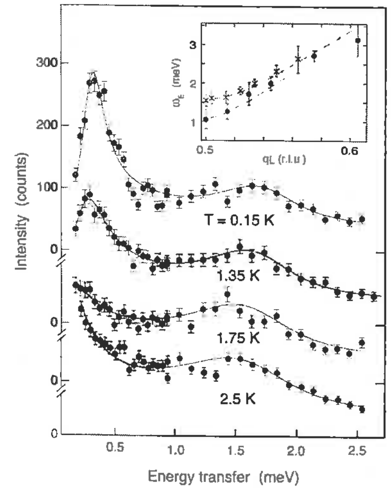
Most of the HF metals adopt a symmetry-broken ground state, either a superconducting (SC) or an antiferromagnetically ordered one [1]. In some of the U-based HF metals, like UPt_3 and UPd_2Al_3, superconductivity is found to coexist with long-range antiferromagnetic (AF) order (sec.2). On the other hand, for several Ce-based materials superconductivity has been observed to form in the vicinity of an AF instability, at which the Néel temperature, T_N, vanishes continuously as a function of a control parameter like external or internal pressure. Exemplary systems are the pressure-induced superconductors $CePd_2Si_2$ and $CeIn_3$ [2] as well as $CeCu_2Si_2$, the first HF metal for which superconductivity at ambient pressure was observed in 1979 [3] (sect. 2). The low-temperature normal (n)-state properties of these latter materials are displaying pronounced non-Fermi-liquid (NFL) phenomena commonly related to critical fluctuations associated with a quantum phase transition between the antiferromagnetically ordered and a disordered (spin-liquid) state [4]. When applying a sufficiently strong magnetic field, NFL properties usually give way to those of a heavy Landau Fermi liquid (LFL). To study NFL phenomena in more detail, it is highly desirable to search for an AF-QCP in low magnetic fields, i.e., *in the absence* of HF superconductivity. This appears to be realized in both $CeCu_{5.9}Au_{0.1}$ [5] and $YbRh_2(\underline{Si}, Ge)_2$ [6] (sec. 3).

Superconductivity

The discovery of superconductivity in the tetragonal compound $CeCu_2Si_2$ containing, at $T > T_K \approx 15$ K, a dense lattice of local magnetic Ce^{3+} moments came as a big surprise, given the antagonistic nature of superconductivity and magnetism. On the other hand, it was found that HF superconductivity is phenomenologically related to the superfluidity of ^3He and cannot be explained by the conventional electron-phonon pairing mechanism [3]. In fact, evidence for a magnetic, rather than phononic, Cooper pairing was provided ten years later by inelastic neutron scattering (INS) experiments on UPt_3 where AF order with an extremely small moment ($0.02\mu_B$/U) forms below $T_N \approx 5$ K [7].

For the hexagonal compound UPd_2Al_3 which enters an antiferromagnetically ordered state with a large saturation moment $\mu_s = 0.85\mu_B$/U at $T_N = 14.3$K, HF superconductivity forms and homogeneously coexists with magnetic order below $T_c = 2$K. UPd_2Al_3 may be called a "magnetic HF superconductor", in which magnetism derives from *localized* 5*f* states while Cooper pairs are formed by more hybridized (*delocalized*) 5*f* states, i.e., heavy (composite) fermion quasiparticles. Based upon quasiparticle tunneling [8] and INS [9] work, the acoustic magnon at the center of the Brillouin zone with an excitation energy $\omega_E \approx 1$ meV could be identified [9] as the "exchange boson" forming Cooper pairs in this strong-coupling HF superconductor. This magnon, which, because of the induced singlet-ground-state type of AF order is called "magnetic exciton", may be viewed as a local crystal-field excitation propagating through the U-lattice via inter-site interactions. It thus replaces the optical phonons in classical strong-coupling superconductors like Pb. Thus, UPd_2Al_3 is the first and so far only superconductor for which a non-phononic pairing mechanism could be demonstrated by the same experimental means as the phononic counterpart in Pb.

The discovery of superconductivity in the tetragonal compound... came as a big surprise



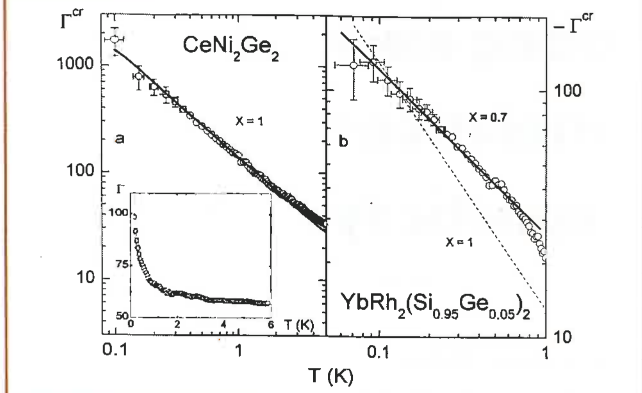
▲ **Fig. 1:** T-evolution of the INS spectra for a UPd_2Al_3 single crystal at the AF ordering wave vector $\vec{Q}_0 = (0,0,q_L{=}0.5)$ through $T_c = 1.8$ K. Solid lines represent fits using the microscopic "two-component" model described in Ref. 9. This procedure may be repeated for other wave vectors $\vec{q}$ close to $\vec{Q}_0$. There is only one fit parameter at given values of T and $\vec{q}$: The magnetic exciton energy $\omega_E(\vec{q})$ shown in the inset for T = 2.5 K (crosses) and 0.15 K (circles). The low-energy peak, inelastic at $T < T_c$ and quasielastic at $T > T_c$, is related to the itinerant HF quasiparticles, the broad hump at higher energies to the magnetic exciton.

The generic phase diagram of $CeCu_2Si_2$ presented in Fig. 2a contains, at small 4*f*-conduction electron coupling constant g, a low-moment spin-density-wave (SDW) or "A"-phase [10] which coexists in a homogeneous way with weak HF superconductivity below $T_c < T_A$, the magnetic ordering temperature. At moderate coupling, the A-phase is fully replaced by strong HF superconductivity, but can be recovered if superconductivity is suppressed by applying an overcritical magnetic field. Beyond a critical coupling constant g_c, at which T_A vanishes continuously characterizing the QCP of this material, pronounced NFL effects were found in the low-temperature n-state properties of "S-type" single crystals [11].

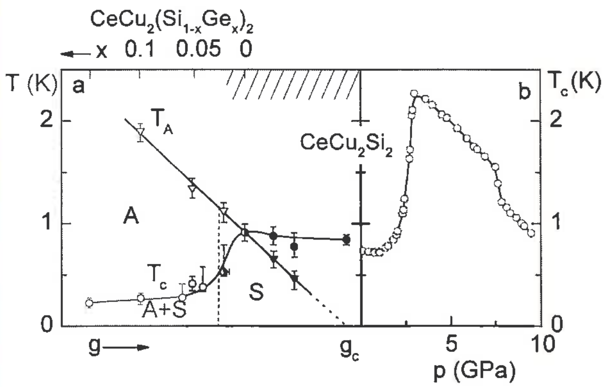
Fig. 2b displays the unusual pressure dependence of the superconducting critical temperature of $CeCu_2Si_2$, highlighting a maximum value of 2.3 K at $p \approx 2.5$ GPa. By deliberately shortening the quasiparticle mean free path through moderate Ge-substitution for Si, Yuan *et al.* could clearly separate two different SC regimes [12]. The low-pressure dome (corresponding to the low-pressure plateau in Fig. 2b) is centered around the AF-QCP, where virtual (high-frequency) fluctuations of the staggered magnetization of the A-phase might be the source to forming massive Cooper pairs [13]. On the other hand, the high-pressure dome (around the maximum T_c in Fig. 2b) appears to coincide with a weak first-order $Ce^{3+} \rightarrow Ce^{3+\delta}$ valence transition [12], giving rise to speculations about charge-fluctuation mediated superconductivity.

Quantum Criticality

Most of the unconventional properties of the low-T n-state not only of $CeCu_2Si_2$ [11], but also of, e.g., $CeNi_2Ge_2$ [14] and UBe_{13} [15] can be explained by low-frequency, spatially extended spin-fluctuations which exist in a nearly AF Fermi liquid forming in the vicinity of a 3D-SDW QCP [4]. On the other hand, thorough INS experiments on the quantum critical material $CeCu_{6-x}Au_x$ ($x = 0.1$) have highlighted [5] the importance of *local* (rather than *extended*) quantum critical fluctuations which are addressed by new theories of a *local-moment* QCP [16]. Recently, Zhu *et al.* [17] proposed that measurements of the Grüneisen ratio $\Gamma \sim \beta/C$ (where β and C are the volume thermal expansion and specific heat, respectively) may well be suited to distinguish between different QCP scenarios. As these authors argue, Γ(T) must diverge at any QCP [17]. This has indeed been observed [18] for both paramagnetic $CeNi_2Ge_2$ (inset of Fig. 3a) and the isostructural weakly antiferromagnetically ordered compound $YbRh_2Si_2$ ($T_N = 70$ mK). In the main part of Fig. 3a the critical Grüneisen ratio, $\Gamma^{cr}(T)$, as given by the singular parts of both β(T) and C(T), is shown to diverge $\sim T^{-x}$ with $x = 1$ for $CeNi_2Ge_2$. This agrees well with the prediction for the 3D-SDW scenario [17]. By contrast, recent investigations of $YbRh_2Si_2$ revealed $\Gamma^{cr}(T) \sim T^{-x}$ with $x = 0.7$ [18] as expected in the presence of *local* AF quantum critical fluctuations (Fig. 3b).

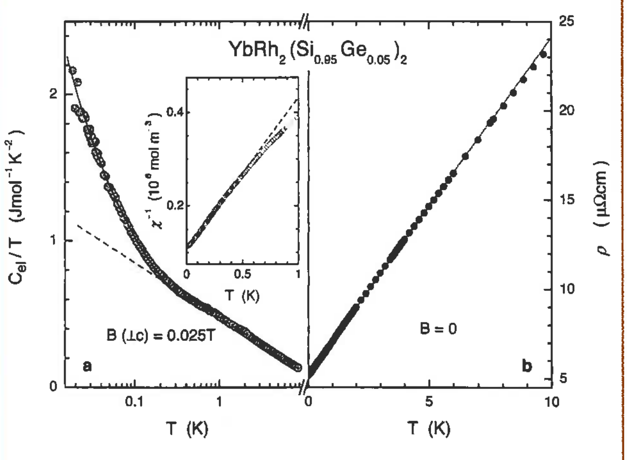


▲ Fig. 3: Critical Grüneisen ratio $\Gamma^{cr} = (V_m/\kappa_T)\,(\beta^{cr}/C^{cr})$, as $(-)\Gamma^{cr}$ vs T in double-logarithmic plots for $CeNi_2Ge_2$ (a) and $YbRh_2(Si_{0.95}Ge_{0.05})_2$ (b) [V_m: molar volume; κ_T: isothermal compressibility; $\beta^{cr} = \beta(T) - bT$ and $C^{cr} = C(T) - (\gamma T + \delta/T^2)$ are the critical components to the volume thermal expansion and specific heat, respectively, cf. Ref. 18]. Inset shows $\Gamma(T) \sim \beta(T)/C(T)$ as measured (on linear scales) for $CeNi_2Ge_2$.



▲ Fig. 2: a. Generic phase diagram of $CeCu_2Si_2$ combining data obtained with undoped polycrystals from the homogeneity range of the ternary chemical phase diagram (hatched) and from Ge-doped ones, $CeCu_2(Si_{1-x}Ge_x)_2$. Since the temperature T_A of the SDW transition increases linearly with the Ge-concentration x, the 4f-conduction electron coupling constant g was assumed to be linear in (1-x), i.e., $(1-T_A)$. b. Pressure dependence of T_c up to p = 8.5 GPa for $CeCu_2Si_2$, see Ref. 12. The different T_c values at p = 0 in a and b relate to two polycrystalline samples with slightly different stoichiometry (Cu content).

A QCP at zero can, in principle, be approached by moderate volume expansion of $YbRh_2Si_2$. In fact, in the case of a slightly doped $YbRh_2(Si_{1-x}Ge_x)_2$ single crystal with a nominal Ge-concentration $x = 0.05$, the Néel temperature was found to be as low as $T_N = 20$ mK [19]. By tuning the system away from its QCP, i.e., through applying a magnetic field (⊥c) larger than $B_c \approx 0.027$ T necessary to suppress the weak AF order in this Ge-doped crystal, the field dependences of both the electronic specific heat, $C_{el}(T,B) = \gamma_0(B)T$, and the electrical resistivity, $\Delta\rho(T,B) = A(B)T^2$, could be attributed to the field-induced low-T LFL state. From the divergences of both the Sommerfeld coefficient $\gamma_0(B)$ and the resistivity coefficient A(B) as $B \rightarrow B_c^+$, as well as from those of $C_{el}(T)/T$ and $A(T) = \Delta\rho(T)/T^2$ as $T \rightarrow 0$ at $B = B_c$ it was concluded [19] that field (temperature) acts as the only relevant energy scale at low temperature (field). Two striking observations made with such $YbRh_2(Si, Ge)_2$ single crystals are worth mentioning:
(i) Correlated, but unscreened large paramagnetic Yb^{3+} moments are observed in the static bulk susceptibility (inset of Fig. 4a) as well as in the low-temperature electron-spin resonance [20].
(ii) The temperature dependences of $\gamma_0(T) = C_{el}(T)/T$ and $\Delta\rho(T)$ taken at $B \lesssim B_c(0)$ behave *disparately*: $\gamma_0(T) \sim (-\log T)$ and $\Delta\rho \sim T$ are observed upon cooling from $T \approx 10$ K down to $T \approx 0.3$ K. While the linear T-dependence of the resistivity can be followed all the way down to 10 mK, the lowest temperature of the experi-



▲ Fig. 4: Electronic specific heat divided by T as a function of T (on a logarithmic scale) measured at a field (⊥c) of 0.025 T, close to $B_c(0) = 0.027$ T (a) and electrical resistivity ρ vs T (on a linear scale) measured at B = 0 (b) for a $YbRh_2(Si_{0.95}Ge_{0.05})_2$ single crystal. Dashed [solid] line in a represents $C_{el}/T \sim (-\log T)$ [$\sim T^{-(0.4\pm0.03)}$], solid line in b represents $\rho - \rho_0 = aT$ with $\rho_0 \approx 5$ μΩcm. Inset displays the zero-field ac-susceptibility as χ^{-1} vs T for the same crystal. In the same T-range (T < 0.3 K) where C_{el} (T)/T deviates from –logT, χ(T) follows a Curie-Weiss law, $\chi^{-1} \sim (T - \Theta)$ with $\Theta \approx -0.3$ K. From the slope a large paramagnetic moment, $\mu_{eff} \approx 1.4\ \mu_B/Yb^{3+}$, is derived.

ment (Fig. 4b), γ(T) at T ≤ 0.3 K is found to diverge stronger than logarithmically, i.e., $\sim T^{-\varepsilon}$, $\varepsilon = 0.4 \pm 0.03$ (Fig. 4a). This disparity hints at a break-up of the composite fermions on the approach to the QCP.

Epilogue

Strong electronic correlations on partially filled localized 4*f* or 5*f* shells, weakly hybridized with the itinerant ligand states, cause the formation of extremely heavy quasiparticles composed of a local spin part and delocalized charge-carrier contributions. They may form Cooper pairs, the SC glue presumably being of magnetic origin, at least in most cases. This could be convincingly demonstrated with the aid of tunneling and neutron scattering experiments on UPd_2Al_3.

In case of a number of Ce-based HF metals, superconductivity was found to be intimately related to the existence of an AF instability, most likely of the 3D-SDW type [4]. On the other hand, the QCP in slightly Ge-doped $YbRh_2Si_2$ appears to be of the *local-moment* variety [16], although already in undoped $YbRh_2Si_2$ the ordered moments below $T_N = 70$ mK are extremely small, in contrast to the large paramagnetic moments, cf. inset of Fig. 4a. Interestingly enough, superconductivity could not be observed in this material. To find out whether a *local-moment* QCP is indeed unfavorable for HF superconductivity has to be left for future work.

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