Unconventional Normal-State Properties and Superconductivity in Heavy-Fermion Metals

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We discuss (i) UPd_2Al_3, for which local-moment antiferromagnetism ($T_N = 14.3\,K$) coexists with heavy-fermion (HF) superconductivity below $T_c \approx 2\,K$, (ii) the HF superconductor $CeCu_2Si_2$ ($T_c \approx 0.6\,K$) which, for $B > B_{c2}$, shows pronounced "non-Fermi-liquid" (NFL) effects near a quantum-critical point at which the spin-density-wave "phase A" disappears, and (iii) the NFL $CeNi_2Ge_2$, a "clean-limit" HF superconductor below $T_c \approx 0.1\,K$.

1. Introduction

Unlike high-T_c cuprates, heavy-fermion superconductors (HFS) show a distinctly non-universal behavior. For example, some of them are "clean-limit" superconductors, which may loose their superconductivity if, upon doping [1], the mean free path, ℓ, of the charge carriers is made as short as the coherence length, ξ_0. On the other hand, the prototypical HFS UBe_{13} [2] shows $\ell < \xi_0$, while superconducting (sc) $CeCu_2Si_2$ samples [3] can be either in the "clean" or in the "dirty limit". The non-universality of the sc phenomena has led to the proposal [4] that more than one Cooper-pairing mechanism may have to be considered for *all* HFS. Because in most of them some kind of antiferromagnetic (AF) order could be established, it is widely believed that electronic exchange mechanisms must play an essential role.

In this paper we discuss members of two groups of HFS with very different low-temperature (T) normal (n)-state properties: The hexagonal compound UPd_2Al_3 [5] belongs to those U-based intermetallics which exhibit coexistence of long-range AF order and a heavy Landau Fermi-liquid (LFL) phase at $T_c < T < T_N$. UPd_2Al_3 is unique among these systems because it shows large, atomic-like, saturation moments due to localized 5f-electrons in the presence of heavy-fermion (HF) quasiparticles, i.e. "itinerant 5f-electrons". In section 2, it is shown that, by using this "dual nature" of the 5f-electrons to analyze recent neutron-scattering data [6], a dominating electronic coupling mechanism for UPd_2Al_3 can be proposed.

The second group of HFS contains, mostly pressure-induced, Ce-based intermetallics whose n state is dominated by pronounced deviations from a LFL, see e. g. Refs. 7 and 8. These "non-Fermi-liquid" (NFL) effects are commonly ascribed [9,10] to the abundance of strong, extended and long-lived fluctuations of the local staggered magnetization in the vicinity of an AF quantum critical point (QCP). Among these systems the tetragonal compound $CeCu_2Si_2$ [3] is unique. A thorough study of the chemical Ce-Cu-Si phase diagram revealed a number of different groundstate properties in different sectors of the existence range of the primary 1:2:2 phase [11]. This allows one to study already at $p = 0$ (i) NFL effects near a QCP at which AF order ("phase A") disappears, (ii) competition as well as (iii) coexistence between HFS and "phase A". In section 3 we wish to focus mainly on the nature of "phase A". Section 4 is devoted to the isostructural compound $CeNi_2Ge_2$, which exhibits NFL effects in its n state and "clean-limit" HF superconductivity, also at $p = 0$ [12-15]. We will present new experimental results for $CeNi_2Ge_2$ and, in addition, address the possible origin of its unconventional n-state properties. The paper is

concluded in section 5 by a short outlook.

2. Local-moment antiferromagnet: UPd_2Al_3

The simultaneous observation of a large saturation moment $\mu_s = 0.85\,\mu_B$ [16] and a large Sommerfeld coefficient $\gamma = 140\,\mathrm{mJ/K^2mole}$ [5] in UPd_2Al_3 has led to the assumption that the average occupation number [17,18] of the U-5f shell is slightly smaller than three, with two localized 5f electrons being responsible for the magnetic properties and an additional itinerant 5f electron. The latter, being stronger hybridized to the conduction electrons, causes both the LFL n-state properties and anisotropic HF superconductivity, as inferred from the large specific-heat jump, $\Delta C = 1.2\gamma T_c$, below $T_c \approx 2\,\mathrm{K}$ [5,17].

In contrast to common attempts to describe the electronic structure of UPd_2Al_3 by using purely itinerant models, Sato et al. [6] have taken into account the fundamental "dual nature" of (localized and itinerant) 5f electrons to interpret recent neutron-scattering spectra on UPd_2Al_3 . The latter highlight a strong coupling between the localized and itinerant 5f states, which can also explain [6] recent tunneling spectra [19]. These, like the neutron-scattering data of Ref. 6, reveal the importance of an acoustic magnetic exciton. Its excitation energy $\omega_E \simeq 1$ meV (near the AF Brillouin zone center) evaluated for $T < T_c$ [6] is almost coinciding with the sc gap, $2\Delta_0 \simeq 6k_BT_c$ [20]. UPd_2Al_3, therefore, appears to be a strong-coupling magnetic-exciton-mediated superconductor, with a polar shape of its gap function [6].

3. Competing groundstates: $CeCu_2Si_2$

Neutron diffraction has, so far, not been able to resolve signatures of "phase A" which, according to resistivity results [21], has the outward appearance of a (perhaps slowly fluctuating, $\tau_A \simeq 10^{-7}$ sec [22]) spin-density wave (SDW). Below we discuss recent results from dc-magnetization (M) experiments which support the SDW nature of "phase A".We used a high-quality single crystal of the "AS variety" where upon cooling, a transition into "phase A" is followed by a sc one at T_c slightly below T_A. This sc transition is associated with a diamagnetic signal, both after zero-field and field cooling, cf. inset of Fig. 1a.

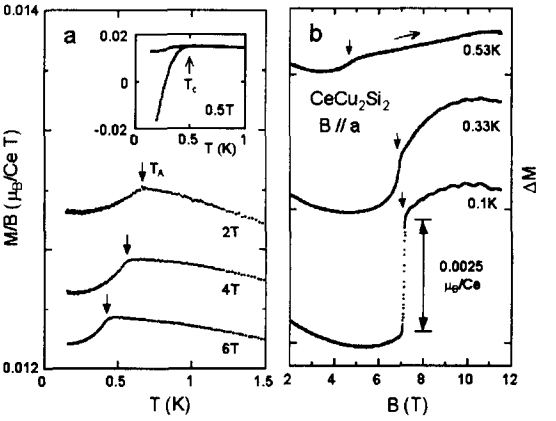

Figure 1. dc-magnetization of an "AS-type" $CeCu_2Si_2$ single crystal for $B \parallel a$ as (a) $M/B\,vs\,T$ for various fields and (b) $\Delta M(B,T) = M(B,T) - M(B, T = 0.95\,\mathrm{K})$ at different temperatures. The curves in (b) are shifted by a constant value for clarity. Inset shows $M/B\,vs\,T$ at $B = 0.5\mathrm{T}$ after zero-field (lower) and field cooling (upper curve).

The main panel of Fig. 1a displays on an enlarged scale $M(T, B = \mathrm{const})$ data taken in overcritical fields $B > B_{c2} \approx 1.5\,\mathrm{T}$. Obviously, cooling through T_A is accompanied by a distinct cusp in the magnetization. In Fig. 1b we show results of $M(B, T = \mathrm{const})$ measurements, from which (background) data taken at $T = 0.95\,\mathrm{K}$ have been subtracted [23]. At $T = 0.1\,\mathrm{K}$ a sharp, first-order metamagnetic-like transition is found at $B = 7\,\mathrm{T}$. This is attributed to the transition from "phase A" to the related high-field "phase B" [24]. Note that albeit the jump anomaly in $\Delta M(B)$ corresponds to only $2.5 \cdot 10^{-3}\mu_B$/Ce it can be clearly resolved in our high-resolution experiment. Upon warming, this phase transition shifts to lower fields and changes its character from first order at $T = 0.1\,\mathrm{K}$ and $0.33\,\mathrm{K}$ to second order at $0.53\,\mathrm{K}$, in accordance with the phase diagrams collected for other "AS-type" crystals [24,25]. For fields in excess of 7 T, de Haas-van Alphen oscillations become visible in the $M(B, T = \mathrm{const})$ curves indicating the high quality of our single crystal.

The above magnetization measurements clearly reflect the magnetic character of "phase A". Both the reduction of the magnetization below T_A as well as the mean-field (mf) type phase-transition anomaly in the specific heat at T_A (not shown) are consistent with a SDW on parts of the renormalized Fermi surface (FS). The FS fraction involved can be estimated from the size of the discontinuity in the specific heat at T_A, ΔC_A, normalized to the specific heat in the paramagnetic state, γT_A. Assuming a mf ratio of $\Delta C_{SDW}/\gamma T_{SDW} = 1.43$ for a fully gapped FS, our result of $\Delta C_A/\gamma T_A = 0.55$ indicates that about 40% of the FS takes part in the SDW formation.

4. Non-Fermi liquid: $CeNi_2Ge_2$

The resistivity results shown in Fig. 2a for a high-quality polycrystal of $CeNi_2Ge_2$ reveal a NFL power-law dependence over more than a decade in temperature. The exponent $\varepsilon = 1.4$ is in accordance with $1 < \varepsilon < 1.5$ as recently calculated for a 3D nearly antiferromagnetic Fermi liquid (NAFFL) in the presence of only weak potential scattering [26]. The deviation from the $T^{1.4}$ law below 100 mK indicates "incipient superconductivity" and can be suppressed by a magnetic field of 0.1 T [12,13]. Down to $T = 0.2$ K, the specific-heat coefficient $\gamma(T) = \Delta C(T)/T$ obeys the predicted [9,10] asymptotic quantum-critical power law $\gamma(T) = \gamma_0 - \alpha\sqrt{T}$ (cf. Fig. 2b). At lower temperature, a huge upturn in $\gamma(T)$ becomes visible whose size is found to differ considerably from sample to sample. Very similar observations were made for $CeCu_2Si_2$. Here, the $\gamma(T)$ upturn appears to be especially pronounced for "S-type" samples [21], i.e. in the vicinity of a QCP ($T_A \to 0$).

Like for the latter compound, any (quadrupole or Zeeman) splitting of nuclear spin states can be excluded for $CeNi_2Ge_2$ as well. For example, very high internal fields of 34 T (91 T) acting on the nuclear spins of Ge (Ni) would have to be assumed to account for the observed $B = 0$ upturn. Since nuclear effects can be safely discarded, the feature is very likely of electronic nature. This striking disparity between the low-T, n-state $\Delta\rho$(T), following the theoretical prediction [26], and $\gamma(T)$, deviating from it [9,10], makes the applicability

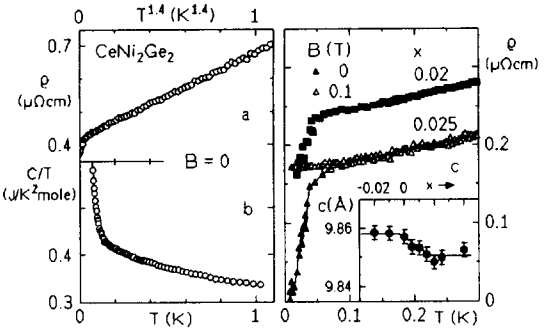

Figure 2. Results of the resistivity and specific heat for $Ce_{1.005}Ni_{2+x}Ge_{2-x}$ polycrystals: $\rho vs T$ and $C/T vs T$ (b) at $B = 0$ for $x = 0$. $\rho vs T$ at $B = 0$ for $x = 0.02$ and $x = 0.025$ as well as at $B = 0.1$T for the latter (c). Inset shows c-lattice parameter as a function of x.

of the NAFFL theory to these HF compounds questionable [21].

By scrutinizing the chemical Ce-Ni-Ge phase diagram utilizing polycrystalline $Ce_{1.005}Ni_{2+x}Ge_{2-x}$ material, we have recently found [27] that samples containing a few percent of Ni excess are sc. For the $x = 0.025$ sample in Fig. 2c a full sc transition below $T_{c,onset} \approx 100$ mK could be registered resistively [28]. Ni-excess in $CeNi_2Ge_2$ acts like Cu-excess in "S-type" $CeCu_2Si_2$ [11]: It strengthens the 4f-conduction electron hybridization (cf. inset of Fig. 2c) and favors HF superconductivity. However, unlike $CeCu_2Si_2$ where superconductivity below $T_c \approx 0.6$ K was observed in samples with large residual resistivities ($\rho_0 \leq 40\mu\Omega cm$ [29]), very low ρ_0 values ($< 1\mu\Omega cm$ [13]) are prerequesite for HF superconductivity to develop in $CeNi_2Ge_2$!

To answer the question, what the origin of the NFL effects might be in this compound, we have searched in the chemical phase diagram for additional phase transitions, different from the sc one. This way we found very recently that polycrystals slightly rich of Ni exhibit the same "B-phase" transition as established in $CeCu_2Si_2$.

Further on, several nearly stoichiometric and Ge-rich $CeNi_2Ge_2$ polycrystals show an anomaly strongly resembling the "A-phase" transition in the coppersilicide. If the existence of "phase A" in $CeNi_2Ge_2$ were established by future work, the NFL phenomena in both compounds could be explained by the same type of AF spinfluctuations near a QCP at which $T_A \to 0$.

5. Outlook

Already the few exemplary HFS discussed above illustrate the multitude of phenomena that have been found for these systems. For most of them, a conclusive experimental verification of (presumably) unconventional sc order parameters is still lacking. With the notable exception of UPd_2Al_3 [6], the specific pairing mechanisms could not be established beyond doubt either.

Finally, the striking disparities between transport and thermodynamic n-state properties discussed for both $CeCu_2Si_2$ and $CeNi_2Ge_2$, but apparently present in other stoichiometric HF metals as well [30], require more experimental and theoretical studies. These should help to find out whether or not a (generalized) Fermi-liquid theory is apt to describe the physics of HF metals near an antiferromagnetic quantum-critical point.

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