Non-fermi liquid behaviour in uranium-based heavy-fermion compounds

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Link to publication

Citation for published version (APA):
de Lemos Correia Estrela, P. M. (2000). Non-fermi liquid behaviour in uranium-based heavy-fermion compounds

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Download date: 07 Jan 2019
2. Non-Fermi liquid behaviour in heavy-fermion compounds

2.1. Heavy-fermion compounds and Fermi liquid theory

Heavy-fermion (HF) systems are predominantly found in cerium and uranium compounds where the $4f$- and $5f$-electron states are relatively close to the Fermi level. Near room temperature (RT), the $f$-moment sublattice has properties resembling those of weakly (Curie-Weiss) interacting magnetic moments. The electronic transport properties are dominated by incoherent scattering of the conduction electrons by the local moments. As the temperature is lowered, local-moment behaviour gives way to electronic properties that are consistent with those of a narrow band of conduction electrons. The crossover temperature is the coherence temperature $T_{coh}$.

In Landau's theory of Fermi liquids (FL), a one-to-one mapping of non-interacting electron states to interacting electron states is assumed close to the Fermi energy. If the interactions are turned adiabatically, the states can be described in terms of quasiparticles, which have an enhanced effective mass due to interactions with other quasiparticles in the surrounding medium.

At sufficiently low temperatures (much lower than the Fermi temperature), a useful tool to describe the thermodynamic properties of a system with itinerant electrons is the effective mass
of the electrons. In normal metals, the effective mass $m^*$ is of the order of the free-electron mass $m_e$, while in heavy-fermion systems $m^*$ can attain values as large as $10^3 m_e$.

A single quasiparticle has an energy

$$\varepsilon^* = \frac{\hbar^2 k^2}{2m^*} (|k| - k_f) .$$

(2.1)

where $k$ is the wave-vector and $k_f = (3\pi^2 n_e)^{1/3}$ is the Fermi wave-vector ($n_e$ is the number of electrons per unit volume). This expression defines the effective mass $m^*$. When a quasiparticle is added to the system, it will have an energy

$$\varepsilon_n = \varepsilon^*_k + \frac{1}{\Omega} \sum f(k,k') \delta n(k') .$$

(2.2)

where $\Omega$ is the volume of the system and $f(k,k')$ is the quasiparticle interaction function. From this, it follows that the energy of the added quasiparticle is not just the bare quasiparticle energy $\varepsilon^*_k$, but also depends on the presence of other quasiparticles. $\delta n(k)=1$ represents an excited quasiparticle and $\delta n(k)=-1$ an excited quasi-hole. Notice that $k$ denotes $(k,\sigma)$ with $\sigma$ the spin index ($\uparrow$ or $\downarrow$). The function $f(k,k')$ can be transformed into spin-symmetric and spin-antisymmetric functions:

$$f(k,\uparrow,k',\uparrow) = f^+(k,k') + f^-(k,k')$$

$$f(k,\uparrow,k',\downarrow) = f^+(k,k') - f^-(k,k') .$$

(2.3)

A restriction of the Fermi-liquid theory is that all involved particles have a momentum very close to the Fermi surface: $|k| = |k_f|$. The $f^{\pm\pm}$ functions can then be expanded in a series of Legendre polynomials $P_l$:

$$f^{\pm\pm}(k,k') = \sum_{l=0}^{\infty} f_l^{\pm\pm} P_l(\cos \theta) .$$

(2.4)

with $\cos \theta = (k \cdot k')/k_f^2$. A dimensionless form of the coefficients $f_l^{\pm\pm}$ is given by the Landau parameters

$$F_l^{\pm\pm} = \frac{m^* k_f}{\pi^2 \hbar^2} f_l^{\pm\pm} .$$

(2.5)

The thermodynamic and response functions of the electronic liquid can now be calculated and shown to be smooth functions of temperature. The density of states at the Fermi energy is given by
Non-Fermi liquid behaviour in heavy-fermion compounds

\[ N(0) = \frac{m^* k^2_{\text{B}}}{\pi^2 \hbar^2} \]  

(2.6)

where the effective mass of the quasiparticles \( m^* \) is related to the bare mass \( m_c \) by a symmetric Landau parameter

\[ \frac{m^*}{m_c} = 1 + \frac{F_0}{3} \]  

(2.7)

The temperature independent Pauli susceptibility has the form

\[ \chi = \frac{\mu_n \mu_n^2 m^* k^2}{\pi^2 \hbar^2} \frac{1}{1 + F_0} \]  

(2.8)

with \( F_0 \) an antisymmetric Landau parameter. \( \chi \) is enhanced with respect to the Pauli susceptibility of the non-interacting system by a factor \( m^*/m_c (1 + F_0) \).

The specific heat \( c_v \) in the FL theory is given by

\[ \frac{c_v}{T} = \gamma = \frac{m^* k^2_{\text{B}}}{3 \hbar^2} \]  

(2.9)

which is enhanced with respect to the specific heat of the non-interacting system by a factor \( m^*/m_c \). Therefore, the specific-heat coefficient gives direct information about the effective mass \( m^* \). The Wilson ratio \( R_W \) relates the Pauli susceptibility to the electronic specific-heat coefficient

\[ R_W = \frac{\gamma}{\chi} = \frac{1}{1 + F_0} \]  

(2.10)

In the case of a non-interacting system, \( R_W = 1 \).

The electrical resistivity behaves as

\[ \rho = \rho_0 + AT^2 \]  

(2.11)

where \( \rho_0 \) is the residual resistivity due to impurities and defects and \( A \) is a constant.

The FL theory gives a good description of the low-temperature properties of metals (above any magnetic or superconducting transition). In heavy-fermion compounds, the high-temperature local-moment behaviour gives way to a low-temperature coherent state where the FL theory is valid with a strongly enhanced effective mass \( m^* \). The specific heat, susceptibility and resistivity follow the temperature dependencies \( c(T) = \gamma T \), \( \chi(T) = \text{const} \) and \( \rho(T) = \rho_0 + AT^2 \), respectively. Values of the Wilson ratio in the range 2-5, as normally found in HF systems, can be accounted for by a negative Landau parameter \( F_0 \). The coefficient \( A \) is related to \( \gamma \) by the empirical Kadowaki-Woodruff relation: \( A/\gamma^2 \sim 10 \mu \Omega \text{cmK}^2\text{mol}^{-1} \text{J}^2 \) [1].
Within the FL theory, spin fluctuations in the HF system give rise to a correction term in the specific heat of the form \( c(T) = \gamma T + \delta T^3 \ln(T/T^*) \), where \( T^* \) is a characteristic spin-fluctuation temperature.

Most HF compounds exhibit the Kondo effect [2], manifested in measurements of the electrical resistivity from room temperature down to low temperatures. The Kondo Hamiltonian describes the exchange interaction of a single magnetic impurity (with spin \( S \)) with a conduction electron (with spin \( s \)):

\[
\mathcal{H} = -2J_s s \cdot S . \tag{2.12}
\]

For a negative coupling parameter \( J \), the impurity spin is completely compensated at low temperatures and a Kondo singlet is formed. As a result, the resistivity obeys a \(-\ln(T/T_K)\) behaviour. The binding energy of a Kondo singlet is

\[
k_BT_K \approx \frac{1}{N(0)} \exp\left(-\frac{1}{JN(0)}\right) . \tag{2.13}
\]

HF materials may be considered as Kondo lattices with a periodic array of magnetic "impurities". In Kondo lattices, scattering at low temperatures may be coherent, resulting in a fast drop of the resistivity (as the temperature is lowered) and a \( T^2 \) behaviour at the lowest temperatures. In general, the compensation of the \( f \)-moments by means of the Kondo effect leads to the formation of a non-magnetic ground state.

On the other hand, antiferromagnetic interactions between the \( f \)-moments are provided via the conduction electrons by the Rudermann-Kittel-Kasuya-Yosida (RKKY) interaction. This interaction tends to form a magnetic ground state in HF systems. The energy associated with the RKKY interaction is

\[
k_BT_{\text{RKKY}} \approx J^2 N(0) . \tag{2.14}
\]

Considering the scales defined by \( T_K \) and \( T_{\text{RKKY}} \), Doniach proposed that the low-temperature ground state of the system is a direct consequence of the competition between the Kondo scattering and the RKKY interaction [3]. A (Doniach) phase diagram can then be constructed (Figure 2.1)\(^1\). Heavy-fermion compounds are in general located close to the magnetic

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\(^1\) Strictly speaking, the Doniach phase diagram is of the form \( T/W \) versus \( J/W \). However, for HF compounds and close to the magnetic instability, external parameters like pressure and doping will more effectively influence the exchange-coupling parameter \( J \) than the bandwidth \( W \). Therefore, and also for simplicity, \( T/J \) diagrams will be used throughout this work.
instability, where the competition between the Kondo effect and the RKKY interaction is most important.

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\usepackage{amsmath}

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\section{2.2. Non-Fermi liquid behaviour}

During the past decade, a new class of heavy-fermion systems that exhibit strong deviations from Fermi-liquid theory has attracted much interest [8]. These non-Fermi liquid (NFL) materials are U-, Ce- and Yb-based intermetallics that, with a few exceptions, have been doped with a non-magnetic element. The main macroscopic properties related to NFL behaviour are a diverging specific heat divided by temperature \( c/T \sim -\ln(T/T_0) \) or \( c/T \sim \gamma_0(1/T)^{1/2} \), a
```

Figure 2.1 - Doniach phase diagram (FL = Fermi liquid, HF = heavy fermion). The dotted lines represent \( T_{\text{RKKY}} \) and \( T_{\text{K}} \). The full line represents the ordering temperature and the dashed line the temperature below which FL behaviour is attained.

Reviews on heavy-fermion compounds are given in Refs. 4 and 5. The Fermi-liquid theory is extensively described in Refs. 6 and 7.
diverging magnetic susceptibility ($\chi \sim 1/bT^3$ or $\chi \sim -\ln(T/T_0)$) and a non-quadratic electric resistivity ($\rho \sim aT^\alpha$ with $\alpha < 2$ and $a$ can be positive or negative).

The divergency of $c/T$ is the hallmark of NFL behaviour. Within the FL theory, a temperature dependent coefficient $\gamma$ that diverges as $T \to 0$ would imply a diverging density of states at the Fermi level. Therefore, FL theory cannot be applied in its simple form. This is why the name *non-Fermi liquid* was first given to this behaviour. Nevertheless, in some cases, models based on a local FL description can account for this new behaviour.

At present, no theoretical models are at hand which yield an universal description of NFL behaviour. However, a common starting point may be found in the physics of quantum critical points. This results from the recognition that NFL properties emerge at or close to the magnetic instability in a typical Doniach phase diagram for HF compounds (Figure 2.2).

![Figure 2.2 - NFL region close to a magnetic instability for HF systems.](image)

Within the Doniach phase diagram, magnetism vanishes when the single-ion Kondo scattering becomes more important than the RKKY interaction. In renormalization group language, antiferromagnetism (AF) and FL behaviour can be considered as two competing fixed points. As the temperature is lowered, the system evolves from the high-temperature local-moment behaviour to one of these fixed points, as represented in the flow diagram in Figure 2.3. The trajectories represented correspond to different values of $T_K/T_{\text{RKKY}}$. When a material is tuned to the critical value of $T_K/T_{\text{RKKY}}$, it is forced to evolve to the quantum critical point (QCP) [9].
The existence of an antiferromagnetic QCP implies that the two fixed points (AF and FL) are linked by a new unstable fixed point. For a wide range of materials, $T_K/T_{RKKY}$ is close to its critical value. These materials will evolve towards the FL or AF fixed points, passing close to the new fixed point. Over a large temperature range their properties, excitations and interactions will be dominated by the physics of this QCP.

However, since a number of $f$-systems exhibiting NFL properties does not seem to be at a QCP, other models have been put forward in order to describe the microscopic mechanisms that lead to NFL behaviour. Single-ion Kondo models have been applied, with relative success. Also models have been proposed where disorder plays a crucial role, as a number of NFL systems are chemically-substituted or diluted compounds and, therefore, disordered.

The most relevant routes that have been proposed to lead to non-Fermi liquid behaviour in $f$-electron systems are:

i) a two-channel Kondo effect [10], where the $f$-electron impurity spin is overscreened by the spins of the conduction electrons, giving rise to an antiferromagnetic superexchange interaction with electrons off the impurity site (see Section 2.3.1);

ii) a distribution of Kondo temperatures [11], where the Kondo effect on each $f$-electron impurity sets a different temperature scale, resulting in a broad range of effective Kondo temperatures; averaging over such a distribution gives rise to thermodynamic properties which follow the NFL expressions (see Section 2.3.2);
iii) a proximity to a QCP [12], where a magnetic or superconducting phase transition occurs at $T = 0 \text{ K}$ (either spontaneously or the transition might be tuned to $0 \text{ K}$ by an external parameter, like hydrostatic or chemical pressure): here, the thermodynamic properties are determined by collective modes corresponding to fluctuations of the order parameter in the vicinity of the critical point (see Section 2.3.3):

iv) a Griffiths phase model [13] where, due to disorder, magnetic clusters appear in the paramagnetic phase close to a QCP (see Section 2.3.4).

NFL properties have also been found in $d$-transition-metal systems. For instance, Ni$_x$Pd$_{1-x}$ has a ferromagnetic QCP at $x = 0.025$ [14]. The specific heat, resistivity and magnetic susceptibility strongly deviate from the standard FL behaviour and can be fully accounted for by a proximity to a ferromagnetic QCP (see Section 2.3.3).

NFL behaviour may also be found in one-dimensional (1D) systems, where it is described theoretically by the Luttinger liquid model [15]. In these 1D systems, the electron-electron interaction is much stronger than in a FL, which may lead to spin-charge separation [16]. Quantum wires [17] and some organic 1D conductors [18] have been described as Luttinger liquids.

The normal state of high-$T_c$ cuprates is also known to exhibit properties that deviate strongly from FL behaviour. A so-called marginal-Fermi liquid model [19] has been proposed as a phenomenological approach to the behaviour of high-temperature superconductors. Here, it is assumed that the spin and charge susceptibilities have an unusual form, in that they are approximately momentum independent and vanish linearly in $\omega/T$ for low frequencies. In contrast, spin and charge of a FL are strongly momentum dependent and have a low-frequency behaviour that becomes independent of temperature as $T \to 0$. This assumption gives rise to a scattering rate linear in temperature and an effective mass that diverges logarithmically as the Fermi energy is approached.
2.3. Routes to non-Fermi liquid behaviour

2.3.1. Multichannel Kondo effect

As mentioned before, within the usual single-channel Kondo effect, the physics of an f-electron system can be described by a local version of Landau's FL theory [20]. However, a simple modification of this model can produce NFL behaviour: the multichannel Kondo effect [10].

In the overcompensated multichannel Kondo model, M identical spin-½ conduction bands exchange-couple to a single-impurity spin $S_I$ with the condition $M/2 > S_I$, so that there are more conduction spins than needed to fully compensate the impurity.

In the simplest case of the $S_I=½$ two-channel Kondo effect (TCKE) [21], the impurity spin is overcompensated by the presence of two conduction spins $½$. The resulting spin of the ground state will be $½$ and an antiferromagnetic superexchange will be generated with electrons off the impurity site. In renormalization group language, the kinetic energy introduced by this superexchange interaction makes the strong-coupling ($J\to\infty$) fixed point unstable, since at this fixed point the kinetic energy is zero. This maps the effective model back to the weak-coupling ($J=0$) limit. However, the weak-coupling fixed point is unstable due to the Kondo effect. Thus both weak- and strong-coupling limits are unstable. Therefore, a non-trivial fixed point at intermediate coupling must exist [22].

This non-trivial fixed point gives rise to a degenerate ground state and a NFL energy spectrum. The extra specific-heat coefficient and spin susceptibility per mole of impurity diverge for $T\to0$ as [23]

$$\frac{c(T)}{T} = -\frac{A'}{T_k} \ln \frac{T}{b'T_k} + B'$$  \hspace{1cm} (2.15a)

$$\chi(T) = -\frac{1}{T_k} \ln \frac{T}{b'T_k} .$$  \hspace{1cm} (2.15b)

where $A' = 0.251R$, $b = 0.41$, $b'T_k$ is of the order of 1 and $B'$ is a temperature independent electronic or crystal-field background in $c/T$. The resistivity, on the other hand, behaves as [24]
\[ p(T) \sim 1 - a \left( \frac{T}{T_k} \right)^{1/2}. \]  
(2.15c)

where \( a \) is of the order of 1. However, while the logarithmic divergence of \( c/T \) and \( \chi \) appears for \( T < 0.5T_k \), \( p \sim 1 - aT^{1/2} \) should be observable only for \( T < 0.05T_k \) [10]. In the intermediate temperature range \( 0.05T_k < T < T_k \), \( p \sim 1 - aT \).

The two channels of conduction spin and the impurity have an effective spin \( 1/2 \). Therefore, the degeneracy of the impurity spin is never lifted, unlike in the ordinary Kondo problem. This residual degeneracy manifests itself in a net residual entropy of \( 1/2R \ln 2 \) per mole impurity. This residual entropy can be recovered by e.g. an external magnetic field [10], which will lift the degeneracy.

For the quadrupolar multichannel Kondo effect (QKE) [25], where the electrical quadrupolar moment of the \( f \)-ion interacts with the conduction electrons and their spins provide the two channels, the susceptibility does not diverge logarithmically but as

\[ \chi(T) = \chi(0) \left[ 1 - b' \left( \frac{T}{T_k} \right)^{1/2} \right] \] 
(2.16)

with \( b' \sim 1 \).

### 2.3.2. Kondo disorder model

A distribution of Kondo temperatures \( T_K \) can arise if a material has large disorder. Around each single magnetic impurity, antiferromagnetically coupled to conduction electrons (assuming an effective spin-\( 1/2 \) impurity magnetic moment), the Kondo effect will occur at a different value of \( T_K \). Averaging over such a distribution can produce thermodynamic and transport properties with NFL-like dependencies due to the broad range of effective Fermi temperatures. Essentially, the unquenched moments contribute to the NFL physics.

In this scenario, a NFL state is generated as a consequence of the interplay of disorder and strong correlations. The main idea of this model is that moderate bare disorder in a lattice model of localized moments is magnified due to the strong local correlations between the \( f \)-moments and the conduction electrons. In particular, a broad distribution of local energy scales (Kondo temperatures) is generated. A few local sites with very low Kondo temperatures are unquenched
at low temperatures and dominate the thermodynamics and transport, giving rise to a dilute gas of low-lying excitations above the disordered metallic ground state. The presence of these unquenched moments leads to the formation of a NFL phase [11].

In UCu$_5_x$Pd$_x$ ($x = 1, 1.5$), Cu NMR studies revealed the presence of a strong inhomogeneous broadening of the NMR line width [26]. This broadening can be explained within the Kondo disorder model assuming a collection of completely uncorrelated spins, each coupled to the conduction-electron bath by a Kondo-coupling constant $N(0)J$, which is allowed to be randomly distributed in the sample. This distribution is supposed to originate from the local disorder induced by Pd substitution at the Cu sites. The thermodynamic response is then calculated by taking an average over the response of a single Kondo spin with a distribution of coupling constants. Because of the exponential dependence of $T_K$ on $N(0)J$, a broad distribution of Kondo temperatures results, as shown in Figure 2.4.

![Figure 2.4](image)

**Figure 2.4** - Distribution of Kondo temperatures in UCu$_5$Pd (dashed line) and in UCu$_{1.5}$Pd$_{1.5}$ (dotted line). The shaded area below $T$ represents the low-$T_K$ spins which remain unquenched at that temperature. Taken from Ref. 11.

The Kondo disorder model also leads to an incoherent nature of the transport properties with sufficient disorder strength. Due to local Kondo physics at each $f$-site, the effective disorder generated from a bare distribution of local $f$-shell parameters is strongly renormalized up to scales of the order of the conduction electron bandwidth. Although clean systems have low resistivities due to the onset of coherence at low temperature, moderate amounts of $f$-element
disorder are capable of destroying this low-temperature coherence, leading to characteristic incoherent Kondo scattering behaviour.

The predictions of the model for the specific heat, susceptibility and resistivity are [11]

\begin{align}
\hat{c}(T) & \sim -T \ln(T/T_c) \\
\chi(T) & \sim -\ln(T/T_c) \\
\rho(T) & \sim 1 - aT
\end{align}

(2.17a) (2.17b) (2.17c)

2.3.3. Proximity to a quantum critical point

A quantum critical point (QCP) occurs when a critical point such as that associated with a ferro- or antiferromagnetic transition is tuned to \( T = 0 \) by some external parameter \( \delta \), such as pressure or dopant concentration. At the QCP (\( \delta = \delta_c \)), the low-temperature thermodynamics is determined by collective modes corresponding to fluctuations of the order parameter, rather than by single-fermion excitations as in a FL. Therefore, NFL properties arise. NFL behaviour can also occur near quantum spin-glass [27] or superconducting [28] transitions.

Like its finite-temperature counterpart (thermal or classical phase transition), a quantum phase transition is characterized by a diverging correlation length \( \xi \) and a diverging relaxation time \( \tau \). However, the critical fluctuations that lead to these diverging length and time scales are quantum fluctuations rather than thermal ones. Contrary to the situation for a classical critical point, the dynamic and static behaviour of a QCP are coupled together. A system at a QCP will be affected in the same way by either a finite frequency or a finite temperature. The system is characterized by the dynamical scaling exponent \( \epsilon \) that describes the divergence of \( \tau \). The value of \( \epsilon \) affects strongly the static critical behaviour [29]. The dynamical exponent \( \epsilon \) takes the value of 2, 3 and 4 for an antiferromagnet, a clean ferromagnet and a dirty ferromagnet, respectively. A \( d \)-dimensional quantum system is related to a classical one with an effective dimension \( d_{\text{eff}} = d + \epsilon \).

When studying the effect of non-zero temperatures on the QCP in itinerant-fermion systems using renormalization-group theory, the diagram of Figure 2.5 applies [12,30]. Different regions close to the QCP must be considered. Region I is the disordered quantum regime where
the FL picture applies, region II is the perturbative classical regime and region III is the classical Gaussian regime. The different regimes are separated by the lines

\[ T_1 \sim (\delta - \delta_c)^{1/2} \]  
\[ T_{\Pi} \sim (\delta - \delta_c)^{1/(d + z - 2)} \]  
\[ T_m \sim (\delta_c - \delta)^{1/(d + z - 2)} \]  

**Figure 2.5** - Phase diagram of the temperature versus the control parameter $\delta$. Region I is the disordered quantum regime, region II is the perturbative classical regime and region III is the classical Gaussian regime. The lines $T_1$, $T_{\Pi}$ and $T_m$ are defined in the text. After Ref. 12.

Calculations of the specific heat give the same expressions for regions II and III, while the correlation length $\xi$ is the same for regions I and II. For the classical regime, i.e. just above the QCP, and for $d = 3$ the specific heat and resistivity near an antiferromagnetic ($z = 2$) QCP have the temperature dependencies

\[ \frac{c(T)}{T} = \gamma_0 - \alpha T^{3/2} \]  
\[ \rho(T) \sim T^{3/2} \]  

while for a ferromagnetic ($z = 3$) QCP,

\[ \frac{c(T)}{T} \sim -\ln(T / T_0) \]  
\[ \rho(T) \sim T^{5/3} \]
The same predictions are obtained within the self-consistent renormalization (SCR) theory of spin fluctuations [31]. The SCR theory takes into account the couplings among the different modes of spin fluctuations in a self-consistent way. Although initially developed for itinerant $d$-electrons, the theory can be modified to nearly localized $f$-electron systems [32]. It assumes that around the magnetic phase boundary there are weakly and nearly ferro- and antiferromagnetic regimes with various anomalous properties, which are predominantly due to exchange-enhanced spin fluctuations. Within this theory, the $d=3$ predictions for $c(T)$ and $\rho(T)$ are the same as in equations 2.19 and 2.20. The pressure dependence of $T_N$ or $T_C$ is also the same as in equation 2.18c. However, at very low temperatures, $c(T)$ and $\rho(T)$ should attain temperature dependencies as in the FL theory.

A phenomenological description of NFL systems at a QCP [27] gives the following scaling relations of the magnetization and specific heat:

$$M = \frac{B}{T^\gamma} \left( \frac{B}{T^{\beta \gamma}} \right)^{\alpha} \quad \text{(2.21a)}$$

$$\frac{c(B,T)}{T} - \frac{c(0,T)}{T} = g \left( \frac{B}{T^{\beta \gamma}} \right) \quad \text{(2.21b)}$$

where $f(x)$ and $g(x)$ are non-singular functions. These scaling relations can also be applied to a field-induced QCP by replacing $B$ and $c(0,T)$ by $\Delta B = B - B_c$ and $c(B_c,T)$, respectively [33].

Another type of QCP, that has been considered theoretically, is the quantum Lifshitz point [34]. A "classical" Lifshitz point is a critical point that, in addition to the onset of magnetic ordering, is characterized by the disappearance of stiffness in one or several directions, i.e., a tricritical point where a disordered phase, a spatial uniformly ordered phase and a spatially modulated ordered phase meet [35]. In the quantum Lifshitz point model, NFL behaviour occurs in the classical Gaussian region near a quantum Lifshitz point in a three-dimensional itinerant antiferromagnet. The Néel temperature is predicted to follow the pressure dependence

$$T_N \sim (P_c - P)^{1/5}.$$  \quad \text{(2.22)}

The specific heat coefficient and resistivity are predicted to vary as

$$c(T)/T \sim T^{1/4} \quad \text{(2.23a)}$$

$$\rho(T) \sim T^{5/4} \quad \text{(2.23b).}$$

Although no NFL system has been found to obey these relations, the concept of loss of stiffness near a QCP might have its relevance in systems like CeCu$_{6-x}$Au$_x$ [36].
2.3.4. Griffiths phase model

Another model, proposed recently, takes into account the effects of disorder near a quantum critical point. In the Griffiths phase model [13], the presence of disorder is considered to lead to the coexistence of a metallic paramagnetic phase and a granular magnetic phase. These coexisting phases are equivalent to the Griffiths phase [41] of a dilute magnetic system.

The "classical" problem of a Griffiths phase occurs in a lattice of magnetic atoms diluted with non-magnetic atoms. Long-range order is lost at the percolation threshold when the last infinite cluster of magnetic moments ceases to exist. Above the threshold, the system is composed of finite clusters of magnetic atoms. When a magnetic field is applied to the percolation lattice, there is a non-analytic contribution from rare large clusters to the free energy [41].

For the Griffiths phase model for NFL compounds, a similar picture can be drawn. Two electronic liquids coexist: in one of them, the magnetic moments are quenched by the Kondo interaction, giving rise to a FL, while the other is dominated by the RKKY interaction giving rise to ordered regions. This inhomogeneous situation is energetically favoured by disorder, due to the entropy contribution to the free energy.

For a generic magnetic HF compound, which exhibits a QCP upon alloying, a phase diagram can be constructed within this model. For small amounts of doping, the RKKY interaction dominates and the system orders magnetically. With increasing doping, the quantum fluctuations grow due to the Kondo effect and the critical temperature decreases until it vanishes for the critical value of doping. At this QCP, the system percolates. For larger values of doping, i.e. in the paramagnetic phase, only finite clusters of magnetic atoms can be found. Among these clusters, there are some rare ones that are large and strongly coupled, in which the spins behave coherently as a giant spin or a magnetic grain. In this phase, the thermodynamic functions show essential singularities with strong effects at low temperatures. The specific heat coefficient and the static susceptibility diverge as

\[
\frac{c(T)}{T} \sim T^{-1+\epsilon} \quad (2.24a)
\]

\[
\chi(T) \sim T^{-1+\frac{1}{2}} \quad (2.24b)
\]
with \( \lambda < 1 \). Notice that \( \lambda = 1 \) corresponds to the FL expressions. The parameter \( \lambda \) also characterizes the temperature dependence of the mean square deviation of the susceptibility due to the distribution of susceptibilities in the system and of the non-linear static susceptibility, as well as the frequency dependencies of the local susceptibility and the NMR relaxation rate [13].

In general terms, within the Griffiths phase model, the NFL behaviour can be observed over an extended region in the paramagnetic phase next to a QCP (Figure 2.6).

\[ \text{Figure 2.6 - Phase diagram for the Griffiths phase model.} \]

**2.4. Magnetotransport in nearly antiferromagnetic metals**

A magnetotransport theory has recently been developed to explain the transport properties of NFL compounds near an antiferromagnetic QCP [42,43]. Under such conditions, the low-energy excitations of a HF system below a characteristic temperature \( T_K \) (see Figure 2.7a) can be assumed to be due to heavy quasiparticles and their collective excitations. The resistivity near the QCP is then determined by scattering of quasiparticles by spin fluctuations. These scattering processes are most important near hot lines, i.e. points on the Fermi surface connected by the
magnetic ordering vector $Q$. In the remaining cold regions, inelastic scattering is weak (Figure 2.7b).

![Diagram](image)

**Figure 2.7** - (a) Phase diagram for an antiferromagnetic QCP. (b) Fermi surface with hot lines where scattering is enhanced - $Q$ is the ordering vector of the AF phase. Taken from Ref. 42.

The theory presented in Ref. 42 predicts the behaviour of the resistivity and magnetoresistance of compounds in the paramagnetic phase near an antiferromagnetic QCP. Considering that spin fluctuations are destroyed at the temperature scale $\Gamma$, where $\Gamma$ is typically of the order of $T_{\text{coh}}$ or $T_K$, the resistivity is universal for $t < x^{1/2}$ and $r < 1$ in the scaling limit $t, x, r \rightarrow 0$ and $t/x, r/x \rightarrow \text{const}$, where $t = T/\Gamma$ measures the temperature, $x = \rho_0/\rho_M \approx 1/\text{RRR}$ measures the amount of disorder and $r \approx \delta - \delta_c$ measures the distance to the QCP in the paramagnetic phase. Here, $\rho_0$ is the residual resistivity, $\rho_M$ is a typical high-temperature ($t \sim 1$) resistivity value, $\delta$ is a control parameter like pressure and $\delta_c$ is its critical value.

Three different regimes are predicted for the resistivity $\Delta \rho = \rho - \rho_0$:

$$\Delta \rho \rho_M = \begin{cases} t^{d/2}, & r < t < x^{2/d-1} \\ x^{2(d-1)} r^{(4-d)/(5-d)}, & \max\left[x^{2/(d-1)} r^{(5-d)/4}, r^{(5-d)/4}\right] < t < x^{1/2} \\ r^{-2} t^{2-d/2}, & t < \min\left[r, x^{1/2} r^{(5-d)/4}\right] \end{cases}$$ (2.25)

For three dimensions ($d = 3$), the diagram of Figure 2.8 results with
\[
\Delta \rho \sim \begin{cases} 
  t^{3/2}, & \text{regions I} \\
  t^{1/2} x, & \text{regions II} \\
  t^2 r^{-1/2}, & \text{region III}
\end{cases}
\] (2.26)

The temperature ranges where the different regimes occur depend on the amount of disorder in the system. In the very dirty limit \( x \to 1 \), region II is not observed, i.e. no \( \rho \sim T \) regime occurs. Regions I (\( \rho \sim T^{3/2} \)) and III (\( \rho \sim T^2 \)) extend over large ranges in this limit and therefore are called the disorder-dominated regime and the disorder-dominated FL regime, respectively.

The \( \rho \sim T^{3/2} \) behaviour predicted by the theories of Millis [12] and Moriya [32] for the AF QCP (see Section 2.3.3) is only observed in a small region close to the QCP. For very clean systems, this behaviour will only be observed at ultra-low temperatures. In the immediate vicinity of the QCP, the scattering process at the hot lines is short-circuited by quasiparticles at the remaining cold regions of the Fermi surface giving way to \( T^2 \) behaviour.

In region IIB (see Figure 2.8), the thermodynamic functions show FL characteristics, although the resistivity rises linearly with temperature.

**Figure 2.8** - Resistivity scaling regimes for a compound near an antiferromagnetic QCP \((d = 3)\). \( t \) measures the temperature, \( x \) the amount of disorder and \( r \) the distance from the QCP in the paramagnetic phase. After Ref. 42.
In the presence of a magnetic field, the resistivity near a QCP is influenced by spin and orbital effects. The spin contribution typically suppresses the antiferromagnetic order. In the paramagnetic phase, AF fluctuations will be suppressed. Within the diagram of Figure 2.8, this corresponds to an increase of $r$ (the distance to the QCP). The suppression of the fluctuations reduces the amount of scattering and the resistivity drops. Therefore, a negative magnetoresistance is expected. Due to the orbital effects, the resistivity increases in field. This positive magnetoresistance originates because $B$ smears out the quasiparticle distribution, minimizing the effect that cold regions short-circuit the hot lines.

The field dependence of the resistance due to the orbital effects is different for the different regions in Figure 2.8. Defining $b = B/B_0$, where $B_0$ is the typical magnetic field necessary to observe Shubnikov-de Haas oscillations at $t = 1$, the following dependencies have been predicted [43]:

- region I (disorder-dominated regime)

$$\frac{\Delta \rho}{\rho_{\text{st}}} \sim \frac{b^2 t^2}{x^2 g_{t,x}} \quad b < g_{t,x}$$
$$\sim \frac{b t}{x} \quad g_{t,x} < b < t^{1/2} x$$
$$\sim \frac{t^{5/2}}{x} \quad t^{1/2} x < b < t^{1/2} x^{1/2}$$

(2.27a)

with $g_{t,x} = \max\{t^{1/2}, 1^{1/2} x^{1/2}\}$.

- region II (clean systems)

$$\frac{\Delta \rho}{\rho_{\text{st}}} \sim \begin{cases} \frac{t x^2 + b^2}{t x^{3/2}} & b < t^{1/2} x \\ \frac{b}{t^{1/2}} & t^{1/2} x < b < h_x \\ \frac{h_x}{t^{1/2}} & h_x < b < \min\{t^{1/2}, t^{1/2} x^{1/2}\} \end{cases}$$

(2.27b)

with $h_x = \min\{t^{1/2}, t^{1/2} x^{1/2}\}$.

- region III (disorder-dominated FL regime)

$$\frac{\Delta \rho}{\rho_{\text{st}}} \sim \begin{cases} \frac{t^2}{r^{1/2}} - \frac{t^4}{x r^{3/2}} & b < x r^{1/2} \\ \frac{r^2}{x r^{3/2}} & x r^{1/2} < b < t x^{1/2} r^{-1/2} \end{cases}$$

(2.27c)
Therefore, this theory predicts a magnetoresistance with an initial $B^2$ behaviour and a crossover towards a linear dependence in $B$ before saturation. In the disorder-dominated FL regime (region III), $\Delta \rho \sim B$ is not observed. Besides this positive orbital contribution to the magnetoresistance, spin effects give a negative magnetoresistance in all regions.

### 2.5. Examples of non-Fermi liquid compounds

A characteristic of most (but not all) HF compounds exhibiting NFL behaviour is the presence of a QCP in the phase diagram. For the majority of the known NFL compounds, the QCP is reached by changing the composition. This introduces disorder in the system and its effect on the critical behaviour may be crucial. A distinction can be made between systems where the $4f$- or $5f$-atom is partially substituted and systems where the ligand configuration is changed. In the first case, a "Kondo hole" introduced by dilution may lead to substantial scattering and loss of coherence, while in the second case the $f$-atoms may experience different local environments and possibly different local Kondo temperatures. Therefore, even though a QCP is present in the phase diagram, the mechanism responsible for the NFL behaviour might be of the single-ion type like Kondo disorder or a multichannel Kondo effect.

NFL properties are also found in a few stoichiometric compounds. The advantage of stoichiometric compounds is that the role of disorder may not be dominant. Usually, hydrostatic pressure can be applied in weakly magnetic HF compounds in order to reach the QCP. Examples of stoichiometric compounds with NFL properties at ambient pressure are $\text{U}_2\text{Pt}_3\text{In}$ [44], $\text{CeNi}_2\text{Ge}_2$ [45] and $\text{YbRh}_2\text{Si}_2$ [46]. Evidence for NFL has also been found in the normal state of $\text{CeCu}_2\text{Si}_2$ [47], which has a complex phase diagram with competition between magnetism and superconductivity, which relates to an intricate metallurgy.

One of the best studied NFL systems is $\text{CeCu}_6\text{Au}$, [48]. $\text{CeCu}_6$ is a non-magnetic HF compound with intersite antiferromagnetic fluctuations, as was shown by inelastic neutron-scattering experiments. The low-temperature properties are characteristic of a FL. Upon alloying with Au, the lattice expands. This leads to a decrease of the hybridization between the $4f$-orbitals and Cu $3d$-orbitals and, therefore, to a decrease of the exchange interaction $J$. For $x > 0.1$, RKKY
interaction between the localized moments leads to antiferromagnetic order. At the critical concentration, $x_c = 0.1$, NFL behaviour is observed (Figure 2.9). Applying pressure has an effect opposite to Au substitution. The antiferromagnetic phase ($x > 0.1$) can be tuned to $T_N = 0$ by pressure: e.g., $p_c = 0.41$ GPa (= 4.1 kbar) for $x = 0.2$ and $p_c = 0.82$ GPa for $x = 0.3$. At these pressure values NFL behaviour is observed, as demonstrated by the logarithmic divergence of the specific heat shown in Figure 2.9. Above the critical-pressure value, FL behaviour is recovered.

Inelastic neutron-scattering studies on CeCu$_{6-x}$Au$_x$ have revealed the presence of quasi 2-dimensional (2D) magnetic critical fluctuations coupled to quasiparticles with 3D dynamics for $x = 0.1$ [36]. These 2D fluctuations can be viewed as precursors to the 3D ordering for $x > 0.1$. Further support for a 2D character of the critical fluctuations is provided by the fact that the temperature dependencies of the specific heat and the resistivity of CeCu$_{5.9}$Au$_{0.1}$ are in agreement with the predictions for a 2D antiferromagnetic QCP: $c/T \sim -\ln(T/T_0)$ and $\rho \sim T$ [12]. Also the dependencies of $T_N$ on the pressure and the Au content are consistent with a 2D AF QCP: $T_N \sim |\delta |$.

Recent inelastic neutron-scattering experiments on single crystals of CeNi$_2$Ge$_2$ also provide evidence for anisotropic magnetic correlations with a quasi-2D character [49].

![Figure 2.9](image)

**Figure 2.9** - Specific heat of CeCu$_{6-x}$Au$_x$ ($x = 0.1, 0.2, 0.3$) plotted as $c/T$ versus $\log T$ for different pressure values. The sharp kinks indicate $T_N$, while the $\log T$ behaviour is characteristic of a NFL. For $x = 0.1$ and $p = 6$ GPa, the FL is restored. Taken from Ref. 48.

Besides pressure and doping, an external magnetic field can also act as a control parameter. In many NFL compounds, the specific heat and the resistivity display a tendency towards FL.
behaviour under the influence of a magnetic field. In magnetic systems like CeCu$_{5.2}$Ag$_{0.8}$, where $T_N = 0.7$ K, a magnetic field decreases the Néel temperature and NFL properties are observed at a critical field value of 2.3 T where $T_N \rightarrow 0$ [33].

A very interesting aspect of the tuning of $T_N$ by pressure is that superconductivity might occur near $p_c$. Strong evidence for magnetically mediated superconductivity has been found in systems like CePd$_2$Si$_2$ and CeIn$_3$ with an unconventional normal state of the NFL type. CePd$_2$Si$_2$ is an antiferromagnet with $T_N = 10.5$ K. Upon applying pressure, $T_N$ drops to below 1.6 K around 2.5 GPa (Figure 2.10). $T_N$ extrapolates to zero at $p_c = 2.7$ GPa ($\approx 27$ kbar) if the linear $T_N(p)$ dependence is assumed to continue. Around this critical pressure, superconductivity appears with a maximum transition temperature, $T_c = 0.6$ K, for $p = p_c$. The superconducting phase extends almost symmetrically to $\pm 0.5$ GPa around $p_c$ [50]. The normal state, above the superconducting phase, exhibits NFL behaviour. The superconductivity observed at the edge of magnetic order in NFL compounds like CePd$_2$Si$_2$ is restricted to high-quality samples. A possible explanation for this is that the attractive magnetic interactions are strong enough to overcome competing interactions and create Cooper pairs. In other words, the superconducting state appears to be magnetically mediated, with the charge carriers held together in pairs by a "magnetic glue" [50]. Traces of superconductivity have also been found in high-purity single crystals of CeNi$_2$Ge$_2$ at ambient pressure [45].

![Figure 2.10 - $T$-$p$ phase diagram of CePd$_2$Si$_2$. For clarity, the values of $T_c$ have been scaled by a factor 3. Inset: $\rho$ versus $T^{-1.2}$ for $p = 2.8$ GPa. Taken from Ref. 50.](image-url)
Recently, much attention has been devoted to Yb compounds. The physics of Yb and Ce systems are comparable due to an electron-hole analogy: the missing 4f-electron in the 4f$^{13}$ configuration of Yb$^{12+}$ can be interpreted as the presence of a 4f-hole, in analogy to the 4f$^1$ electron in Ce$^{1+}$. Accordingly, Yb systems respond to doping and pressure in reverse with respect to Ce systems. For instance, pressure may drive Yb compounds towards the magnetic regime, crossing the QCP from the non-magnetic side, while in Ce compounds the opposite effect is observed. Another important property is the valence of the Yb ion: divalent Yb (4f$^{14}$ configuration) is non-magnetic, while trivalent Yb (4f$^{13}$) is magnetic. Proper substitutions of the ligand atoms in an Yb system may induce a crossover from the divalent to the trivalent state, hence inducing a crossover from a non-magnetic to a magnetic compound. This occurs e.g. in the system YbCu$_{5-x}$Al$_x$, where a gradual change of the valence of Yb is observed with increasing Al content $x$: non-magnetic Yb(4f$^{14}$) for $x=0$ and magnetic Yb(4f$^{13}$) for $x=2$. A quantum critical point occurs for $x=1.5$, where NFL properties are observed [51].

Many attempts have been made to group all NFL heavy-fermion compounds in one universal class. However, there does not seem to be a single and uniform picture of the mechanism responsible for NFL behaviour. One striking example of this diversity is the system $U_{1-x}M_xPd_2Al_3$ with $M=Th$ or Y. UPd$_2$Al$_3$ is a well known HF compound with coexistence of antiferromagnetism and superconductivity. Upon Th doping ($x<0.2$), $T_S$ decreases only slightly and $T_c\to0$ at $x\sim0.1$. This small decrease suggests that U is tetravalent, just like Th, in $U_{1-x}Th_xPd_2Al_3$ for $0\leq x<0.2$ [52]. As the Th content increases further, a crossover region ($0.2<x<0.4$) occurs where neither antiferromagnetism nor superconductivity has been observed. For $x>0.6$, NFL behaviour is observed. The NFL characteristics of $\rho$, $c$ and $\chi$ scale with $x$ and $T_K$, indicating that a single-ion mechanism could be responsible for the NFL. No QCP seems to be present in the $T$-$x$ diagram. On the other hand, the $T$-$x$ diagram of the $U_{1-x}Y_xPd_2Al_3$ system is remarkably different. Upon Y doping, $T_S$ decreases rapidly and $T_c=0$ for $x\sim0.03$. NFL behaviour occurs around the QCP at $x_c=0.7$, where $T_S$ vanishes. The characteristics of $\rho$, $c$ and $\chi$ are consistent with cooperative phenomena arising from fluctuations related to magnetic order above the QCP [52]. Therefore, substitutions with Y$^{3+}$ or Th$^{4+}$ lead to NFL regimes associated with different mechanisms: single-ion for Th and cooperative for Y.

NFL properties have been observed in many other systems. A list of some representative HF compounds exhibiting NFL behaviour is given in Table 2.1. Recent reviews are given in Refs. 8, 47, 48 and 51.
Table 2.1 - List of some HF compounds exhibiting NFL properties. References to most recent and general papers are given (for the original works see references therein).

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References
Non-Fermi liquid behaviour in heavy-fermion compounds

43. A. Rosch. preprint (cond-mat/9910432).