



Monica Elena Macovei

**Magnetism in Yb- and
Ce-based heavy-fermion
metals under pressure**



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List of Symbols and Abbreviations

a	crystallographic axis; lattice constant
A	cross-section area
\AA	Angstrom unit; $1\text{\AA} = 0.1 \text{ nm} = 10^{-10} \text{ m}$
a.c.	alternating current
AFM	antiferromagnetic
b	crystallographic axis; lattice constant
B	magnetic field
B_c	critical magnetic field where the magnetic ordering temperature becomes zero
c	crystallographic axis; lattice constant; coefficient of the logarithmic increase of the resistivity
C_{el}	electronic specific heat
CEF	crystalline electric field
Cu:Be	copper-beryllium alloy
DAC	diamond anvil cell
d.c.	direct current
DOS	density of states
DWM	domain wall movement
F_0^S	antisymmetric Landau parameter
F_1^S	symmetric Landau parameter
FM	ferromagnetic
HF	heavy fermion
J	total angular momentum; exchange-coupling constant
k_B	Boltzmann constant; $k_B = 1.3807 \cdot 10^{-23} \text{ JK}^{-1}$
LFL	Landau Fermi-liquid
m^*	effective mass
m_e	free electron mass; $m_e = 9.1095 \cdot 10^{-31} \text{ kg}$
n	resistivity exponent; $\Delta\rho(T) \propto T^n$

$N(E_F)$	density of states at the Fermi level
NFL	non-Fermi-liquid
p	pressure
p_c	pressure at which the magnetic ordering temperature becomes zero
PM	paramagnetic
PPMS	Physical Property Measurement System
QCP	quantum critical point
QPT	quantum phase transition
RKKY	Ruderman-Kittel-Kasuya-Yosida interaction
RRR	residual resistivity ratio; $\rho_{300\text{K}}/\rho_0$
s	spin of the conduction electron
S	spin of the magnetic impurity; entropy
SC	superconducting; superconductivity; superconductor
SDW	spin-density-wave
T	temperature
T_C	Curie temperature
T_c	superconducting transition temperature
T_K	Kondo temperature
T_{LFL}	upper temperature limit of the LFL region
T_{max} ,	
T_{max}^{low} , T_{max}^{high}	temperatures corresponding to the maxima in the resistivity
T_N	Néel temperature
T_{RKKY}	characteristic temperature of the RKKY interaction
V	unit-cell volume
x	concentration
γ	electronic specific-heat coefficient
δ	non-thermal control parameter
δ_c	control parameter at which the magnetic-ordering temperature becomes zero
θ_D	Debye temperature
μ_B	Bohr magneton; $\mu_B = 9.2741 \cdot 10^{-24} \text{JT}^{-1}$
μ_{eff}	effective magnetic moment
ρ	electrical resistivity
ρ_0	residual resistivity corresponding to $\rho(T = 0)$
χ	magnetic susceptibility

Introduction

For more than 30 years the investigation of heavy-fermion (HF) metals has been one of the most fascinating and interesting fields in condensed-matter physics both experimentally and theoretically. The HF phenomenon is observed in compounds based on elements such as, e.g., Ce or Yb and manifests itself in the existence of quasiparticles with very large effective mass, m^* , at low temperatures. The ground state of these systems is considered to result from a competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. On one hand, the RKKY mechanism accounts for the indirect interaction between neighboring magnetic moments via the conduction electrons favoring magnetic order. On the other hand, the Kondo effect screens the f moment by spin polarizing the conduction electrons. The balance of these interactions is set by the local exchange interaction, J , between the f shell and conduction electrons. Thus, depending on the strength of J , the ground state of a HF compound can be either paramagnetic (PM) or magnetically ordered. Even interplay between magnetism and superconductivity is observed in some systems. The anomalous properties for which the competition between Kondo effect and RKKY interaction plays an important role are still far from being fully understood. High-pressure experiments are of particular interest because for a given system J can be tuned by external pressure (p). For Ce-based Kondo-lattice compounds, increasing pressure is known to increase J and, thereby, favoring a non-ordered ground state, while for Yb-based Kondo-lattice systems J is expected to decrease with increasing p and, thus, to favor long-range magnetic order.

The Landau Fermi-liquid (LFL) theory has been outstandingly successful in describing the low-temperature properties of normal and HF metals. However, since the 1990's an increasing number of HF systems has been reported to show strong deviations from the LFL theory; the so called non-Fermi-liquid (NFL) behavior. Up to now, a theoretical model which yields an universal description of NFL behavior is not available. However, different mechanisms have been proposed to describe the NFL properties, i.e., the proximity to a quantum critical point (QCP) at which the magnetic-transition temperature is continuously tuned to zero. The properties in the vicinity of this second-order zero-temperature quantum phase transition (QPT) are determined by quantum fluctuations rather than thermal fluctuations, leading to anomalous temperature dependencies of the thermodynamic and transport properties even at finite temperatures.

In this work, we focus on the study of the putative pressure-induced magnetic QCPs in YbIr_2Si_2 and CeRuPO . In recent years YbRh_2Si_2 has attracted considerable

attention as being the first ordered Yb-based HF metal situated in the vicinity to a QCP exhibiting pronounced NFL behavior (e.g., Trovarelli 2000a, Trovarelli 2000b, Gegenwart 2002, Custers 2003). The close vicinity to a QCP is evidenced by a magnetic transition at $T_N = 70$ mK, which can be suppressed either by the application of a small magnetic field (Gegenwart 2002) or a small negative chemical pressure (Mederle 2002). Thus, a drawback of YbRh_2Si_2 is that the QCP can only be reached by applying an external magnetic field or by chemical substitution. Both are known to affect the behavior in the quantum critical regime; the first one by breaking the time-reversal symmetry and the latter one by introducing additional disorder. The ambient-pressure thermodynamic and transport properties of YbIr_2Si_2 indicate that this system is placed on the PM side of a QCP, in contrast to its Rh-homolog which is just on the magnetic side (Hossain 2005). Since the magnetic trivalent Yb ion is smaller than the non-magnetic divalent one, pressure favors the magnetic state in Yb compounds. Application of pressure on YbIr_2Si_2 is expected to tune the system through a QCP, providing a unique opportunity to investigate the physical properties at and around a magnetic QCP in a clean stoichiometric Yb system. Previous measurements on YbIr_2Si_2 under pressure suggest that a ferromagnetic (FM) QPT might exist in this system under sufficiently high pressures (Yuan 2006). Therefore, we performed a detailed study on the effect of pressure on YbIr_2Si_2 by means of electrical resistivity and X-ray powder-diffraction measurements.

The heavy-fermion compound CeRuPO is a rare example of a FM Kondo-lattice system with an ordering temperature of about $T_C = 14$ K at ambient pressure and a Kondo temperature on the order of $T_K \approx 10$ K (Krellner 2007). So far, the behavior at a FM QCP in a Kondo-lattice system is not settled. Therefore, we investigated the effect of pressure on the FM order in CeRuPO, where pressure is expected to suppress the FM ordering temperature. Furthermore, CeRuPO crystallizes in the same type of structure as the compound series RTPnO (R: rare earth, T: transition metal, Pn: P or As) which has started to attract considerable attention because of the discovery of superconductivity with a transition temperature exceeding 50 K (e.g., Chen 2008, Kamihara 2008). Starting from these premises, pressure studies on CeRuPO are important and are expected to shed new light on quantum critical phenomena.

This thesis is divided into five chapters. After this introduction, Chapter 1 summarizes the theoretical concepts related to the different physical phenomena observed in the studied materials and which are important for the understanding of the experimental results. Chapter 2 describes the experimental techniques employed in this work. In Chapter 3 the effect of pressure on YbIr_2Si_2 studied by electrical resistivity and X-ray powder-diffraction measurements is discussed. Results of electrical-resistivity measurements on $\text{Yb}(\text{Rh}_{0.94}\text{Ir}_{0.06})_2\text{Si}_2$ under pressure are also included in this chapter. High-pressure investigations on CeRuPO by means of electrical resistivity and a.c. susceptibility are presented in Chapter 4. At the end, Chapter 5 summarizes and concludes this thesis.

1 Theoretical concepts

1.1 Heavy-fermion systems

Heavy-fermion (HF) systems are intermetallic compounds containing rare-earth or actinide elements with partially filled $4f$ - and $5f$ -electron shells, respectively. The name of this class of materials is connected to the high effective mass, m^* , of their conduction electrons. This heavy mass manifests itself, for example, in a large electronic specific heat or Pauli susceptibility at low temperatures. An interesting aspect of the HF systems is that a variety of unusual low-temperature properties can occur as a result of different ground states. In this chapter, the presentation of the complex properties of this class of materials is restricted to those that are important for the understanding of the materials studied in this work. For a more detailed discussion on HF compounds the reader is referred to review articles, e.g., Stewart 1984, Fulde 1988, Grewe and Steglich 1991, and Stewart 2001.

At elevated temperatures, HF metals exhibit properties resembling those of conventional metals with weakly interacting magnetic moments immersed in a sea of conduction electrons. The electronic transport properties are dominated by incoherent scattering of the conduction electrons off the local moments. As the temperature is reduced below a characteristic temperature, a crossover to a coherent scattering at low temperatures is observed. The low-temperature properties of HF metals display similarities to those of normal metals. Thus, the thermodynamic properties of the HF systems may be described in terms of the Landau Fermi-liquid (LFL) theory.

1.1.1 Single-impurity Kondo effect

In the early 1930's a minimum of the electrical resistivity, $\rho(T)$, followed by an increase toward lower temperatures, was observed in simple metals such as gold or copper with small amount of magnetic impurities (e.g., Fe). This phenomenon was theoretically not understood before Kondo's work in 1964 (Kondo 1964). His theory

explains the upturn of the resistivity at low temperatures by considering the scattering of the conduction electrons off a single magnetic ion in an otherwise non-magnetic sea of conduction electrons. In general, the single-impurity Kondo effect is observed in diluted alloys with a small amount of $3d$ or $4f$ impurities, in which the magnetic moments do not interact, directly or indirectly, due to the large distance in between them. The important aspect of this scattering mechanism is that the resistance increases logarithmically upon lowering the temperature. The above-mentioned resistance minimum is caused by an interplay between the T^5 -dependent resistivity, due to the electron-phonon interaction dominating the resistance at high temperatures, and the logarithmically increasing spin-dependent scattering at low temperatures. It turns out that the theoretical estimations made by Kondo are valid only above a characteristic temperature, which is known as the Kondo temperature, T_K . Below it, Kondo's prediction leads to an unphysical result, namely the resistance diverges as $T \rightarrow 0$. Known as the "Kondo problem", the behavior of $\rho(T)$ at low temperatures was solved by Wilson using the renormalization-group technique (Wilson 1975). Within this framework, the exact solution at $T = 0$ consists in a non-magnetic spin-singlet state formed by an antiparallel coupling between the impurity spin and the conduction electron spins. In the simplest model, the $s - d$ model (Wilson 1975), a single impurity spin $S = \frac{1}{2}$ is coupled by an exchange interaction J to the conduction electrons of the host metal. This model is also valid for systems containing a $4f$ impurity embedded in a non-magnetic metallic host. The classical exchange Hamiltonian can be written as

$$H = -Js \cdot S, \quad (1.1)$$

where s is the conduction electron spin. The exchange-coupling constant, J , depends on the hybridization strength or matrix element between the impurity spin and the conduction electron, V_{s-f} , and the binding energy of the $4f$ level, ϵ_{4f} , as $J = -\frac{V_{s-f}^2}{\epsilon_{4f}}$. The temperature dependence of the thermodynamic properties in the single Kondo impurity case was derived applying the Bethe-Ansatz on the classical exchange Hamiltonian (Desgranges 1982, Andrei 1983). The Coqblin-Schrieffer model generalizes the $s - d$ model for effective impurity spins larger than $1/2$ (Coqblin 1969). Later, the spin-orbit coupling and crystalline electric field (CEF) effects were included in these models leading to a good agreement between the experiments and the theoretical estimations (Rajan 1983, Desgranges 1985, Desgranges 1986). Today, there exists a variety of theoretical models describing the single-impurity Kondo problem at different levels or complexity and a number of theoretical approaches have been used to solve these models (Hewson 1997).

The physical properties of diluted Kondo systems may be classified with respect to

the Kondo temperature. T_K determines the characteristic energy scale of the interaction between the magnetic impurity and the conduction electrons. It is defined as

$$k_B T_K \propto \frac{1}{N(E_F)} \exp\left(-\frac{1}{|JN(E_F)|}\right), \quad (1.2)$$

where k_B is the Boltzmann constant and $N(E_F)$ represents the electronic density of states (DOS) at the Fermi level, E_F .

- At $T \gg T_K$ the temperature dependencies of the resistivity and specific heat resemble those of normal metals. The impurity spin behaves as free magnetic moment giving rise to a magnetic susceptibility, $\chi(T)$, displaying a Curie-Weiss-type behavior. As $T \rightarrow T_K$ the resistivity follows $\Delta\rho(T) = (\rho(T) - \rho_0) \propto -\ln T$, where ρ_0 is the residual resistivity.
- At very low temperatures, $T \ll T_K$, the transport properties are well described within the Landau Fermi-liquid formalism which is addressed in Section 1.2. As soon as the magnetic moments are completely compensated, $\chi(T)$ shows a temperature-independent Pauli susceptibility.
- At $T = 0$, the properties are characteristic of a non-magnetic spin-singlet state. The magnetic susceptibility and the electronic specific-heat coefficient are enhanced compared with those in a normal metallic behavior, while $\rho(T)$ saturates at a constant value ρ_0 . Moreover, the hybridization between the $4f$ and the conduction electrons gives rise to two peaks in the DOS: one broad peak centered at the position of the $4f$ level, below E_F , and a narrow peak located at the Fermi level which has the width of the order of $k_B T_K$ and is known as the Abrikosov-Suhl or Kondo resonance peak.

1.1.2 Kondo-lattice systems and RKKY interaction

The single-impurity Kondo effect is caused by the antiferromagnetic (AFM) exchange interaction between a small amount of non-interacting magnetic impurities and conduction electrons. The situation changes, if the localized magnetic moments form a dense periodic array. Thus, the so-called Kondo-lattice systems can be viewed as a lattice of f electrons, each with a magnetic moment, embedded in a metallic host. At high temperatures the physical properties of these dense f -electron materials are similar to those of the single-impurity Kondo systems, but marked differences are observed at low temperatures. An alteration in physical properties can be clearly distinguished in the low- T resistivity, where due to the periodicity of the arrangement of the f electrons