Ferromagnetism, superconductivity and quantum criticality in uranium intermetallics

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1. Introduction

1.1. General introduction

The physics of strongly correlated metals, especially heavy fermion systems (HFSs) [1-4], has been studied intensively. Heavy-fermion behavior is mainly found in strongly correlated *f*-electron materials, notably Ce-, Yb- and U-intermetallic compounds. At low temperatures, heavy fermion systems are described by the Landau Fermi liquid (FL) theory [5,6]. The main characteristic is a large enhancement of the effective mass as derived from the large value of the linear coefficient γ in the specific heat $c = \gamma T$, the enhanced Pauli susceptibility χ_0 and the large coefficient A of the electrical resistivity $\rho \sim A^{\mathrm{n}}$ (with $n = 2$). However, in the past decade many correlated *f*-electron materials were discovered, which show unusual properties that strongly deviate from those of a FL. This anomalous behavior, observed for instance in the low-temperature magnetic, thermodynamic and transport properties (*i.e.* for $T \rightarrow 0$ γ diverges logarithmically and the exponent of the resistivity $n < 2$), has led to the identification of a new ground state of correlated metals, the so-called non-Fermi liquid (NFL) state (for a review see [7-9]). In the different scenario's proposed as origin of NFL behavior, the mechanisms which are associated with quantum phase transitions (QPTs) form a timely and challenging research subject in condensed matter physics for both experimentalists [7-10] and theorists [11,12].

In *f*-electron HFSs, QPTs are in general transitions between a magnetic and non-magnetic ground state at absolute zero of temperature, which is driven by a non-thermal control parameter, such as pressure p [13-16], magnetic field B [17-21] or chemical doping x [22-27]. In the case of QPTs, the thermal fluctuations are no longer relevant and quantum fluctuations dominate. The singular point of the tuning parameter at which the QPT takes place is called the quantum critical point (QCP). In a simple model, the QCP results from the competition of the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, as in the Doniach-like picture [28,29]. Here the Kondo interaction [30], which quenches the local moment of the *f*-electrons by conduction electron screening, competes with the RKKY interaction [31], which enhances long-range magnetic order.

Interestingly, in the vicinity of the QCP, not only unusual properties (NFL behavior) are observed, but also new collective states may emerge, particularly unconventional superconductivity (SC). The formation of such a superconducting state cannot be explained by the standard Bardeen-Cooper-Schrieffer (BCS) theory for *s*-wave superconductivity [32].

Unconventional SC states have been found to emerge near pressure-induced QCPs in antiferromagnetic (*e.g.*, $CePd_2Si_2$ and $CeIn_3$ [33]) and ferromagnetic (*e.g.* UGe_2 [34]) systems. Evidence is at hand that Cooper pairing is mediated by antiferromagnetic or ferromagnetic spin fluctuations [35], resulting in *d*-wave pairing for antiferromagnetic (AF) and *p*-wave pairing for ferromagnetic (FM) systems.

Because ferromagnetism and superconductivity (SC) are mutually exclusive, it came as a large surprise that their coexistence is nevertheless realized in a few materials: the ferromagnetic superconductors UGe_2 [34], URhGe [36] and (possibly) UIr [37]. These systems currently attract much attention. Of special interest is URhGe in which itinerant magnetism, with a Curie temperature $T_C = 9.5$ K, coexists with SC, with a superconducting transition temperature $T_S = 0.25$ K, *at ambient pressure* [36]. Moreover, re-entrant SC is induced by applying a large magnetic field ($B \sim 12$ T) [38] along the orthorhombic *b*-axis. These findings prompted the important question whether it would be possible to study the magnetic spin fluctuations of a FM superconductor, by tuning URhGe to a QCP.

Motivated by the above idea, we inspected the ordering temperatures of U*TX* compounds (*T* = transition metal Rh, Ru, Co, and *X* = *p*-electron element Ge, Si) [39,40]. According to literature, among the neighboring compounds of URhGe, URhSi also orders ferromagnetically, while URuGe, UCoGe, URuSi and UCoSi did not show magnetic order. Therefore we decided to study the evolution of ferromagnetism of URhGe by doping with Ru and Co in order to search for a FM quantum critical point. The results are reported in this thesis. In the course of this work we found that UCoGe also orders ferromagnetically (see Fig. 1.1). Moreover, we discovered that UCoGe is a new ambient pressure FM superconductor. Our results on the U(Rh,Ru)Ge system probably provide the first example of an 5*f*-electron systems that can be tuned to a FM QCP, while UCoGe might be the first ambient pressure triplet superconductor, where the pairing interaction is due to critical ferromagnetic spin fluctuations directly associated with a FM QCP.

1.2. Outline

This thesis comprises seven chapters, after this introductory chapter, and is followed by a summary, as below.

In Chapter 2, the main experimental techniques are presented. We first describe the sample preparation method and characterization. Next we give details of the in-house experimental set-ups, used to investigate the macroscopic physical properties of the compounds, like magnetization, resistivity and specific heat.

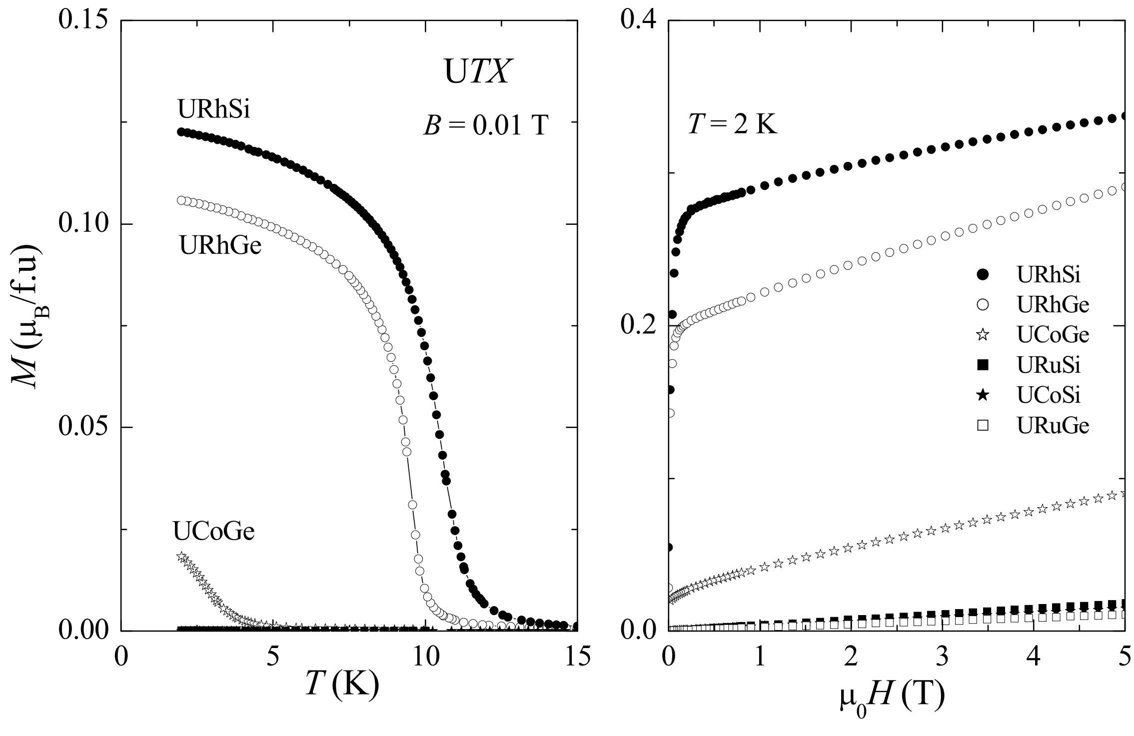

Figure 1.1 Left panel: Temperature variation of the dc-magnetization measured in a field B = 0.01 T of U*TX* compounds (with T = Rh, Ru, Co, and X = Ge, Si). This reveals the compounds URhSi, URhGe and UCoGe order ferromagnetically at Curie temperatures T_C = 10.4, 9.6 and 3 K, respectively, while UCoSi, URuGe and URuSi have a paramagnetic ground state. *Right panel*: Field dependence of the magnetization of U*TX* compounds measured in a field up to 5 T at 2 K.

Chapter 3 is about theory. An introduction to heavy fermion systems and Fermi liquid theory is given, followed by the basic concepts of quantum critical point physics and NFL behavior in *f*-electron HFSs. The chapter ends with a brief overview of theoretical aspects relevant for the explanation of the coexistence of superconductivity and ferromagnetism, as well as of for order parameter models of FM superconductors.

In Chapter 4 we report the evidence for a continuous FM QPT in the $URh_{1-x}Ru_xGe$ series. Upon alloying URhGe by Ru, ferromagnetism is smoothly suppressed and vanishes at a critical Ru concentration x_{cr} = 0.38. Magnetization, transport, specific heat and ac-susceptibility measurements are presented. The low-temperature data are analysed in terms of NFL behavior associated with a QCP.

The study of a single-crystal $URh_{1-x}Ru_xGe$ close to the magnetic instability, with $x \sim 0.38$, is the topic of Chapter 5. The data, obtained by transport, magnetization and specific heat measurements, reveal the compound is strongly anisotropic and that the FL state is recovered upon application of a high magnetic field.

In Chapter 6, we study the evolution of ferromagnetism in polycrystalline $URh_{1-x}Co_xGe$ and $URhGe_{1-x}Si_x$ samples by means of magnetization and transport measurements. In the case of Co and Si doping, a FM QCP does not appear. However, for the end compound UCoGe superconductivity was discovered, which coexists with ferromagnetism at ambient pressure.

In Chapter 7, we describe the properties of the new ferromagnetic superconductor UCoGe as measured for both poly- and single-crystalline samples. The macro- and microscopic experiments on the polycrystals provide proof for the coexistence of bulk FM and SC. The magnetization and transport data taken on a single-crystalline sample confirm UCoGe is a uniaxial ferromagnet. Upper-critical field measurements show the SC gap function has an axial symmetry, with point nodes along the direction of the magnetic moment.

A search for a FM QCP in the $UCoGe_{1-x}Si_x$ alloys is presented in Chapter 8. By measuring the magnetic and transport properties we show that FM and SC are both depressed upon Si doping. Interestingly, we observe that the SC state is always confined to the FM phase and that both states vanish at the same Si concentration. This shows SC and FM are closely tied together.