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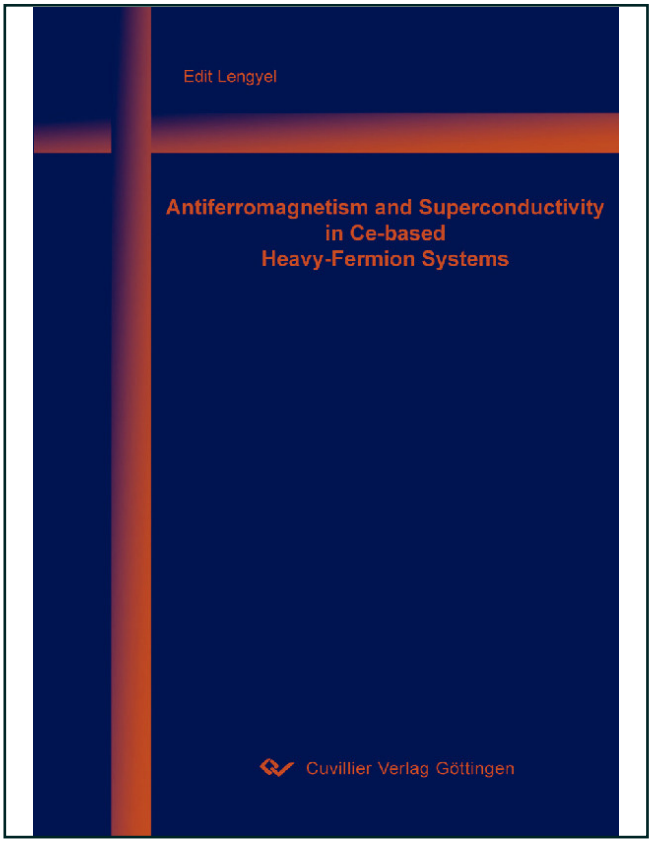
Antiferromagnetism and Superconductivity in Ce-based Heavy-Fermion Systems



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Introduction

Within the last three decades various intermetallic compounds have been found to posses "heavy" electrons. Known under the name of heavy-fermion (HF) materials, they cover an important research area in the field of solid state physics. HF systems usually contain rare-earth elements such as Ce, Yb or actinides like U. Ce-based HF systems are the most numerous among rare-earth-based HF systems. In this case Ce is known to be in the Ce^{3+} valence state with one $4f$ electron. Depending on the occupation number of the $4f$ state of Ce, a multitude of different ground states can be expected for these compounds. In order to tune a Ce-based system through the several different ground states, pressure can be considered the cleanest available tool. With increasing pressure the interaction between the conduction electrons and the $4f$ electron of Ce is gradually increasing. The Kondo interaction, considered to be the basic ingredient in the formation of a HF state, seems to play an important role in the region where the $4f$ electron of Ce is slightly interacting with the conduction electrons. As a result of the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, leading to the formation of a magnetic ground state, and the Kondo interaction, leading to the formation of a non-magnetic singlet ground state, an antiferromagnetically (AFM) ordered state usually occurs at low temperatures in this low hybridization state. At pressures high enough to lead to the transition of the $4f$ electron of Ce into the conduction band a state, called intermediate-valence (IV) state, is formed. Superconductivity (SC), found to be unconventional and most likely mediated by the different fluctuations associated to the specific instabilities present nearby, exists in many of the Ce-based compounds. AFM spin fluctuations are supposed to mediate the formation of SC in the vicinity of the AFM ordered state, while strong valence fluctuations, related to the delocalization of the $4f$ electron of Ce, are considered responsible for the SC state existing in the proximity of a valence instability of Ce.

In this work we have studied the effect of pressure on three different tetragonal Ce-based systems. All three compounds are, at ambient pressure, in a HF state. $CeCoIn_5$, a HF SC at ambient pressure [Petrovic 2001a], is located in the close vicinity

of an AFM instability. Though never detected in $CeCoIn_5$, an AFM ordered state is expected to exist at a slightly negative pressure. Application of pressure moves $CeCoIn_5$ even further away from the AFM ordered state. Ce_2RhIn_8, a compound belonging to the same family of $Ce_nT_mIn_{3n+2m}$ (T: Co, Rh, Ir) HF compounds as $CeCoIn_5$, orders AFM at ambient pressure [Thompson 2001]. Application of pressure continuously suppresses the AFM phase transition temperature. The appearance of pressure-induced SC is expected in this material. The third investigated compound, the A/S-type $CeCu_2Si_2$, already at ambient pressure offers the possibility to study the intimate interaction between AFM and SC [Steglich 1979]. The AFM ordered state, present above the temperature where SC is formed, is gradually suppressed by application of pressure, while the SC state is stabilized upon increasing pressure. With further increasing pressure, $CeCu_2Si_2$ approaches the region where the degeneracy of the $4f$ state is increased by a collapse of the crystalline electric field (CEF) splitting and a transition to an IV state takes place. However, SC survives in the system over a very broad pressure range. While the role of the AFM order in the formation of a SC state is relatively easy to study in a large number of Ce-based HF systems, the influence of the unstable $4f$ state of Ce on the formation of a SC state is still a problem difficult to approach experimentally. $CeCu_2Si_2$ is nearly unique among Ce-based HF compounds by the fact that SC is present at low temperatures over a wide pressure range and typical instabilities, such as AFM and valence, are both accessible by application of moderate pressure and are in the same time sufficiently separated in pressure to be distinguishable.

Most of this thesis is based on results obtained from low-temperature ($0.26\ \mathrm{K} \leq T \leq 7\ \mathrm{K}$) heat-capacity measurements under hydrostatic pressure ($p < 2.1$ GPa) and in magnetic field ($B \leq 8$ T). In addition, results from pressure-dependent low-temperature ($T \geq 0.26$ K) a.c.-susceptibility and magnetocaloric measurements are presented. Even though difficult to measure under extreme conditions as high pressure and low temperatures, the specific heat represents the thermodynamic property of matter which gives important information about bulk properties. Application of pressure can produce structural, electronic or other kind of phase transitions in matter. Compared to other control parameters, such as chemical doping or magnetic field, pressure is the cleanest tool to tune a system by only reducing its unit-cell volume.

The SC state, when present, and its interplay with the AFM instability was investigated in the three compounds studied in this work. Predictions of the AFM spin-fluctuation theory, as well as of theories promoting magnetic-fluctuation mediated SC in HF systems, have been probed by our measurements. The influence of a possible valence instability on the high-pressure SC state of $CeCu_2Si_2$ has been

studied as well. The possible existence of quantum critical points (QCPs) in regions where different types of instabilities are suppressed to $T = 0$ K was also addressed in this work. Landau-Fermi-liquid (LFL) type behavior at low temperatures, suited to describe the normal state of certain HF materials, is not expected to be observed in systems located in the vicinity of a QCP. Therefore, deviations from the predictions of the LFL theory, e.g. a logarithmic divergence of the low-temperature electronic specific-heat coefficient, were used to verify the proximity of a system to a QCP.

The thesis is divided into six chapters. Theoretical concepts, related to the different physical phenomena observed in the studied materials and considered necessary for the understanding of the experimental results, are contained in the first chapter. Chapter 2 describes the experimental techniques employed in this thesis. An important issue addressed in this chapter is related to the way accurate specific-heat data can be obtained by measuring under extreme conditions as high pressures and low temperatures. Therefore, a new type of pressure cell, used to achieve higher pressures, developed during this thesis is as well described. The way the experimental data were analyzed and an estimation of the errors implied in the obtained results are also included in this chapter. Chapters 3 and 4 present the experimental results. In Chapter 3 results obtained on two members of the $Ce_nT_mIn_{3n+2m}$ (T: Co, Rh, Ir) family of HF compounds, $CeCoIn_5$ and Ce_2RhIn_8, are described. Presenting a layered structure, similar to the structure of high-T_c cuprate SCs, the $Ce_nT_mIn_{3n+2m}$ (T: Co, Rh, Ir) family of HF compounds are suited to study the supposed role of electronic dimensionality in the formation of a magnetically mediated SC state. A brief description of the $Ce_nT_mIn_{3n+2m}$ (T: Co, Rh, Ir) family of HF compounds is given at the beginning of Chapter 3. The remaining of this chapter is divided in two parts. The effect of pressure on the HF SC $CeCoIn_5$ is presented in the first part, while the second part describes the evolution under pressure of the HF AFM Ce_2RhIn_8. Both parts end with a discussion. Chapter 4 is devoted to A/S-type $CeCu_2Si_2$ under pressure. Accurate low-temperature heat-capacity data up to $p < 2.1$ GPa, obtained for the first time on A/S-type single-crystalline $CeCu_2Si_2$, are presented. A detailed study in the low-pressure range, while slowly increasing pressure, helped to carefully analyze the peculiar interplay of AFM and SC in A/S-type $CeCu_2Si_2$. With increasing pressure, we were able by our measurements to distinguish between two different SC regions: SC at low pressures, in the vicinity of an AFM QCP, and SC at high pressures, in a region located close to a possible valence transition of Ce. Moreover, we have observed evidence for different SC order parameters in the two distinct SC regions. Support in favor of the suggestion that different mechanisms (AFM spin fluctuations in the low-pressure SC region and valence fluctuations in the high-pressure

SC region) are implied in the formation of the two SC states has been found. At the end, Chapter 5 summarizes the thesis.

Chapter 1

Theoretical concepts

1.1 Introduction to heavy-fermion systems

Low-temperature physical properties of matter are connected to the low-entropy states and therefore to a small number of degrees of freedom. Knowledge about the ground-state properties of matter stays at the basis of understanding any physical properties which appear at higher temperatures. Additional degrees of freedom can be taken into account in the study of a system only when the ground-state properties are elucidated. HF systems are especially suited to study the ground-state properties of matter due to the increased values of their measurable physical properties at low temperatures. With their usual transition temperatures of about 10 K or below, the HF systems are unlikely to lead to immediate applications but they may help to find out and to solve the insufficiencies of the existing theories in order to better understand condensed matter physics at higher temperatures.

1.1.1 The Kondo effect

The Kondo effect arises from the interaction between a single magnetic ion, such as iron, and the conduction electrons in an otherwise non-magnetic metal (e.g. Fe in Au). When embedded in a sea of conduction electrons, the spin of this magnetic impurity interacts with the conduction electrons. As a result of this interaction, a logarithmic increase of the electrical resistivity of the metal when the temperature is decreased, $\Delta\rho(T) \propto -\ln T$, and a minimum in $\rho(T)$ right before $\rho(T)$ starts to logarithmically increase, is found. In 1964, it was shown by Kondo that this low-temperature increase of the resistivity is due to the electronic scattering from the magnetic ion which interacts with the conduction electrons, while the observed minimum in $\rho(T)$ derives from an interplay between the monotonically decreasing phonon resis-

tivity, usually $\Delta\rho_{phonon}(T) \propto T^5$, and the logarithmically increasing spin-dependent contribution [Kondo 1964]. However, though the high-temperature properties were very well reproduced, this first theoretical estimation made by Kondo was leading to the unphysical prediction that the resistance would be infinite at $T = 0$ K. Due to the extremely strong coupling at low temperatures between the impurity spin and the conduction electrons, the perturbation theory used by Kondo in solving this problem was breaking down. Known as the 'Kondo problem', the behavior at low temperatures remained unsolved for a while. Ten years later, under the name of 'single-impurity Kondo model', a new theory based on the numerical renormalization group (NRG), accounting for the low-temperature properties of the metallic systems with a low amount of magnetic impurities, was elaborated [Wilson 1975]. Replacing the perturbation theory used by Kondo by the NRG theory, Wilson has obtained ground-state and low-temperature results for the case of the isolated magnetic impurity with spin $S = 1/2$. As a result of an antiparallel type of interaction, at low temperatures, the impurity spin is completely compensated by the spins of the conduction electrons, leading to the formation of a Kondo singlet. The exchange hamiltonian can be written as: $H = -JSs$, with S representing the spin of the magnetic impurity, s the spin of a conduction electron and J the coupling. The coupling J depends on the hybridization matrix element, V, between the impurity spin and the conduction electrons as $J \propto V^2$. For a negative coupling J, typical for the antiparallel interaction, it was shown that the exact solution at $T = 0$ K consists of a non-magnetic singlet. Following the work of Wilson, exact results for the low-temperature thermodynamic properties by using the Bethe-Ansatz were obtained [Andrei 1980, Desgranges 1982]. Triggered by the experimental results, orbital degeneracy (spin-orbit coupling) and CEF splitting effects were later added to these models (by using the Coqblin-Schrieffer model [Coqblin 1969] and the N-fold degenerate Anderson model [Anderson 1981, Bickers 1987]), leading to relatively good agreement between the experimental findings and the theoretical estimates [Rajan 1983, Desgranges 1985, Desgranges 1987]. Those theoretical results are especially suited to describe Ce and Yb impurities with $N = 2j+1$, where j is the total angular-momentum quantum number associated with the degenerate ground-state multiplet of the impurity ion. The main results of the single-impurity Kondo model can be summarized as following:

1. At $T = 0$ K the properties are characteristic of a spin-singlet state (non-magnetic). The magnetic susceptibility, χ, and the electronic specific-heat coefficient, γ, are greatly enhanced and the electrical resistivity approaches the unitarity limit ρ_0. The low-temperature ($T \ll T_K$, for T_K cf. no. 3) behavior is characteristic of a local LFL state, with strongly enhanced values of χ and γ.