

1 Introduction

Nowadays, many modern materials, such as high-temperature superconductors, geometrically frustrated oxides and low-dimensional magnets, are discussed in terms of strong correlations in complex systems. Common to these materials is that their unique properties arise from many-body effects. In particular, in this field strongly correlated electron materials represent one of the major topics. Here, to obtain more insight in this - on behalf of theory - still poorly understood research area, model systems are needed in order to advance our knowledge on correlated electron materials in particular and correlation physics in general. In this situation, the physics of heavy fermion systems, as archetypical strongly correlated electron systems, is one key to better understand such behavior.

As a result of electronic correlations, heavy fermion systems display a wide range of exotic features, such as

- quantum phase transitions and non-Fermi liquid behavior, *e.g.* in $\text{CeCu}_{5.9}\text{Au}_{0.1}$ [1–5];
- unconventional superconductivity, *e.g.* in UPt_3 [6];
- "hidden order" phases, as observed in URu_2Si_2 [7].

Inherent to heavy fermions is the vicinity to a magnetic instability as a result of the hybridization J between conduction and localized f electrons. The physical ground state properties of heavy fermions as function of the strength of hybridization is schematically summarized in the so-called Doniach phase diagram. With decreasing hybridization these materials transform from an intermediate valence state for large J values into a stable f -shell system, and in between crossing the Kondo regime. In the Kondo regime, the hybridization between conduction and f -electrons is the source for strong correlations between the electrons. As long as no magnetic order occurs, this behavior can be described in terms of Fermi liquid behavior.

Upon further reduction of the hybridization, at a critical hybridization value J_c magnetic order sets in. At J_c the physics of heavy fermion systems is determined by quantum fluctuations, hence the name quantum critical point (QCP). At the QCP exceptional physical properties are observed, such as non-Fermi liquid behavior with a resistivity deviating from Fermi liquid behavior. Experimentally, in most cases magnetically ordered heavy fermion system can be tuned by external control parameters such as pressure or magnetic fields through such a quantum critical point.

On the low hybridization side of the Doniach phase diagram are systems with localized f -electrons. These f -electrons can potentially order, resulting in magnetically ordered structures composed of dipolar moments. Further, higher multipole moments, in particular quadrupolar moments, can also exhibit ordered structures and control the physical ground state properties.

One of the main open topics concerning heavy fermions is the interplay between crystallographic disorder and the correlated electron state. Enhanced disorder effects are arising from correlations between the charge carriers. Furthermore, non-Fermi liquid behavior is predicted and observed for moderately structural disordered systems in the vicinity of a quantum critical point.

In contrast, for strongly localized or completely delocalized systems pronounced disorder effects are not expected. An investigation of the effect of disorder on the degree of itinerancy appears to be of interest. Therefore, in this thesis materials with a varying level of itinerancy are investigated with respect to the physical ground state properties. In particular, materials studied here are ranging from Ce intermetallics like CePt₃B, U heavy fermion systems such as UPd₂Sb, and UPd₂Sn, or systems previously considered to be strongly localized, *viz.*, UPt₂Si₂ and UPd₃. Finally, a truly localized f -electron material, PrB₆, is studied regarding its properties.

The outline of this thesis is as follows:

In **Chapter 2** a brief introduction in the theory of heavy fermion systems is provided. The competition between magnetic RKKY interaction and Kondo interaction is summarized in the Doniach phase diagram. The properties of the Fermi liquid model are summarized and concepts accounting for non-Fermi liquid behavior are pointed out. Finally, a brief introduction into quadrupolar ordering will be provided.

In **Chapter 3** the experimental methods used in this thesis are described and illustrated using measurements on the alloying series $\text{CePt}_3\text{B}_{1-x}\text{Si}_x$. Bulk property studies from previous works, in particular susceptibility, specific heat and electronic transport, on CePt_3B reveal a first transition into an antiferromagnetic and secondly at lower temperatures into a weak ferromagnetic phase. In addition, here the bulk properties of the alloyed samples $\text{CePt}_3\text{B}_{0.8}\text{Si}_{0.2}$ and $\text{CePt}_3\text{B}_{0.6}\text{Si}_{0.4}$ have been investigated. Using these measurements the phase diagram of $\text{CePt}_3\text{B}_{1-x}\text{Si}_x$ has been generated. In order to verify the scenario deduced from the bulk property study and to determine the magnetic structure neutron scattering experiments on CePt_3B have been carried out. Surprisingly, in our neutron scattering experiments no evidence of neither antiferromagnetic nor ferromagnetic order has been found. Subsequently, μSR measurements have been carried out to successfully prove the existence of bulk magnetism in CePt_3B . In addition, at the end of Chapter 3 an introduction into resonant x-ray scattering is given to provide the basis for the experiments presented on PrB_6 .

In **Chapter 4** two closely related material classes are investigated, namely UPd_2Sb and $\text{UPd}_{2-x}\text{Sn}$. First, a review of the properties of the Heusler compounds UT_2M (T: *d* electron element, M heavier element such as In, Sn Sb or Pb) investigated so far is presented. In the following, the heavy fermion system UPd_2Sb is investigated by means of neutron scattering experiments. From these data, it is shown that unconventional semi-conductor like behavior of the resistivity can be accounted for by structurally disordered regions in the sample. The antiferromagnetic structure has been determined and the neutron scattering experiments reveal an interplay of structural disorder and magnetism. The influence of structural disorder on the Fermi liquid properties has been investigated on a series of compounds $\text{UPd}_{2-x}\text{Sn}$, with different stoichiometric composition ($x = 0, 0.02$ and 0.04) and different heat treatments. Using these experimental results the predictions on disorder induced non-Fermi liquid behavior in the vicinity of a quantum critical point and the influence of structural disorder on the Hall effect have been investigated.

Chapter 5 is dedicated to the moderately mass enhanced compound UPt_2Si_2 . Previously, UPt_2Si_2 has been discussed as a local moment antiferromagnet, which ought to be describable by a crystal electric field level scheme. In our reinvestigation of the low temperature electronic transport properties strong

evidence has been found, that UPt_2Si_2 is better understood in terms of an itinerant system rather than a localized one. This evidence of delocalized f -electron behavior has been supported by susceptibility and resistivity measurements in high magnetic fields. Furthermore, a new phase diagram, based on high magnetic field magnetization and resistivity measurements, has been generated and new field induced phases have been observed. Altogether, these findings imply that UPt_2Si_2 has to be discussed as a delocalized f -electron system with the unusual physical properties arising from Fermi surface effects.

Chapter 6 discusses the influence of Pt doping on the properties of antiferroquadrupolar ordered intermetallic UPd_3 . The phase diagram of $\text{U}(\text{Pd}_{1-x}\text{Pt}_x)_3$ has been generated by means of specific heat, electronic transport and susceptibility measurements. We show that UPd_3 is very sensitive to Pt doping, *i.e.*, a very small amount of Pt in UPd_3 destroys the quadrupolar ordered phases. Moreover, the magnetic phase diagram of $\text{U}(\text{Pd}_{1-x}\text{Pt}_x)_3$, $x = 0.005$, has been obtained by means of susceptibility and resistivity measurements in different applied magnetic fields. Furthermore, a newly observed splitting in the susceptibility between zero-field cooled and field cooled measurements with the magnetic field applied along the crystallographic c -axis is detected, which possibly is associated to a quadrupolar and magnetic transition.

In **Chapter 7** the results of a detailed study of the rare earth hexaboride PrB_6 by means of resonant x-ray scattering is presented. PrB_6 undergoes two phase transitions, the first one from a paramagnetic into an incommensurate, antiferromagnetically ordered phase and at lower temperatures a second one into a commensurate antiferromagnetic phase. In this study, structural Bragg peaks are investigated for all three phases and evidence of a lattice distortion in the commensurate phase has been provided. Furthermore, detailed investigations on magnetic and charge Bragg peaks have been carried out. The Bragg peaks associated with charge ordering have been examined in all three phases. In addition, the type of magnetic ordering in the commensurate and incommensurate phase has been determined.