Evolution of Magnetism and its Interplay with Superconductivity in Heavy-Fermion U(Pt,Pd)3

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Chapter 1

Introduction

In this booklet the interplay between magnetism and superconductivity in the heavy-fermion system U(Pd,Pt)$_3$ is investigated. We use neutron-diffraction and μSR experiments to study the magnetic properties, while the superconducting properties are investigated by (magneto)resistance, specific heat, thermal expansion and magnetostriction. The main chapters in this thesis are written in the form of a journal publication. As a consequence some items are present in more than one chapter. In this introductory chapter we first present a short review of the properties of UPt$_3$, followed by the motivation of our research. After this we present the outline of the thesis.

1.1 General introduction

In the past fifteen years a host of experiments has been carried out on the heavy-fermion superconductor UPt$_3$. In spite of this large experimental effort, there are still many open questions concerning the magnetism and superconductivity in the system. UPt$_3$ is regarded as exemplary amongst the heavy-fermion superconductors, because of the coexistence of superconductivity and weak antiferromagnetic order. The non-standard BCS properties of the antiferromagnetic heavy-fermion superconductors provide strong evidence for an unconventional Cooper pair state and have led to speculations upon electron-electron mediated superconductivity.

At low temperatures the normal state of UPt$_3$ is characterised by pronounced spin fluctuations ($T^* \sim 20$ K), which gives rise to an effective mass of 200 times the free electron mass [1,2]. Below $T_N = 6$ K an unusual type of antiferromagnetic order has been detected by neutron diffraction [3] and μSR [4]. The size of the ordered moment is unusually small, $m = 0.02\ \mu_B$/U-atom. Incipient magnetic order in UPt$_3$ was first detected by substitution studies [2]. By replacing Pt by iso-electronic Pd , pronounced phase-transition anomalies in thermal and transport properties provided evidence for an antiferromagnetic transition of the spin-density-wave type.

The unconventional superconducting properties of UPt_3 are reflected in particular in the observation of two consecutive superconducting phase transitions with the second transition located approximately 55 mK below the first one. When a magnetic field is applied a third superconducting phase appears. Substitution studies are a powerful method to study the unconventional superconducting properties. In these studies Pd takes a special place, because it is so far the only substitution for which the distance between the two superconducting transitions (ΔT_c) increases [5]. Most other substitutions, either on the U- or the Pt-site, tend to smear out and to decrease both superconducting transitions at the same rate, leaving ΔT_c approximately constant.

Different scenarios have been proposed [6-10] to explain the unusual phase diagram of UPt_3 in an external magnetic field [11,12] and under pressure [13,14]. Almost all theories of the superconducting phase diagram involve unconventional superconductivity, i.e. the symmetry of the superconducting gap function is lower than that of the underlying Fermi surface [15]. These scenarios have almost exclusively been discussed on the basis of generalised Ginzburg-Landau (GL) theories of superconductivity, where the free energy is purely derived by symmetry arguments.

The scenarios for the UPt_3-phase diagram can be divided into at least two different classes. There is either a symmetry breaking field (SBF) required to lift the degeneracy of the gap or an accidental near-degeneracy of the superconducting gap function. The main objective of the present study is to investigate whether the weak antiferromagnetic order is the symmetry breaking field or not. We performed neutron-diffraction and μSR experiments in order to investigate whether the antiferromagnetic order correlates with the superconducting properties.

An other important issue is the nature of the weak antiferromagnetic order. By substituting Pd or Au on the Pt site or Th on the U site anomalies in thermal and transport properties provided evidence for an antiferromagnetic transition with substantial magnetic moments. By exploring the development of the magnetic order for Pd substitution we expect to arrive at a better understanding of the weak antiferromagnetic order in pure UPt_3.

1.2 Outline

The outline of this thesis is as follows. In chapter 2, the details of several experimental techniques are discussed. For the in-house experimental techniques we restrict ourselves to a basic description. In this chapter also the sample preparation is presented. After that a detailed description of the μSR technique will follow, while also some elements of the neutron-scattering technique will be discussed.

Chapter 3 is a theory chapter, dealing first with the unconventional superconducting properties of UPt_3. The superconducting state is described within several Ginzburg-Landau models in which the symmetry of the superconducting gap plays an important role. The second part deals with the principles of μSR. The most important issue here is how to interpret the muon depolarisation function.

In chapter 4 the evolution of the antiferromagnetic order in UPt_3 doped with Pd is studied by neutron-diffraction. The development of the small-moment antiferromagnetic order (SMAF) in $U(Pt_{1-x}Pd_x)_3$ single crystals is studied for x= 0.000, 0.001, 0.002 and 0.005. The interplay between small-moment magnetism and superconductivity is discussed. The large-moment antiferromagnetism (LMAF) is measured for single crystals with x= 0.01, 0.02 and 0.05. The differences between small-moment and large-moment antiferromagnetic order are reviewed.

In chapter 5 the evolution of the antiferromagnetic order in $U(Pt_{1-x}Pd_x)_3$ is studied by μSR. The μSR experiments provide important evidence that there is a fundamental difference between large-moment and small-moment antiferromagnetic order. While the SMAF state is not observed by zero-field μSR in polycrystalline samples with x= 0, 0.002 and 0.005, the LMAF shows up as pronounced oscillations in the muon depolarisation function for x= 0.01, 0.02 and 0.05.

In chapter 6 the superconducting properties of $U(Pt_{1-x}Pd_x)_3$ are studied by resistivity, specific heat, thermal expansion and magnetostriction. Combined thermal expansion and magnetostriction data are used to construct the multicomponent superconducting phase diagram of $U(Pt_{0.998}Pd_{0.002})_3$ as a function of temperature and magnetic field. The constructed phase diagram is compared with the phase superconducting diagram of pure UPt_3.

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