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

Keizer, R.J.

Publication date
1999

Summary

For more than a decade now it has been realised that the intermetallic compound UPt_3 is an exemplary system for the study of unconventional magnetic and superconducting properties. The unconventional superconducting properties ($T_c \approx 0.5$ K) of UPt_3 are most clearly evidenced by the multicomponent superconducting phase diagram in the B-T plane. Notably in zero-field two consecutive superconducting phase transitions are observed at a distance $\Delta T_c = T_c^+ - T_c^- = 0.055$ K. The magnetic properties are unconventional in the sense that pronounced antiferromagnetic spin fluctuation phenomena coexist with antiferromagnetic order ($T_N \approx 6$ K) with an extremely small ordered moment ($m \approx 0.02\ \mu_B$/U-atom). By substituting Pt by small amounts of isoelectronic Pd the superconducting and magnetic properties are strongly influenced. In this thesis we report a study of the magnetic properties of the $U(Pt,Pd)_3$ system by means of neutron-diffraction and μSR experiments, while the superconducting properties are investigated by (magneto)resistance, specific heat, thermal expansion and magnetostriction techniques. In this way we are able to probe the interplay of magnetism and superconductivity in the $U(Pt,Pd)_3$ system.

Chapter 1 gives a short general introduction, followed by the motivation of our research. The experimental techniques used to study the superconducting and magnetic properties of $U(Pt,Pd)_3$ are described in chapter 2. The in-house techniques are only briefly presented, while the principles of the μSR and the neutron-diffraction techniques are discussed in more detail.

In chapter 3 we present the theoretical aspects of our research. This chapter consists of two distinct parts. In the first part the theory of unconventional superconductivity in UPt_3 is discussed, while in the remaining part the interpretation of the muon depolarisation function as measured in a μSR experiment is presented. As regards unconventional superconductivity in UPt_3, we predominantly focus on Ginzburg-Landau models. In these models the symmetry of the superconducting gap function plays an important role. The Ginzburg-Landau models presented in chapter 3 are: (i) triplet superconductivity with negligible spin-orbit coupling, described by a 1D representation, (ii) coupling of the components of a 2D superconducting vector order parameter to a symmetry breaking field. (iii) coupling of two nearly degenerate 1D superconducting order parameters. In scenarios (i) and (ii) a symmetry breaking field (SBF) is required to lift the degeneracy of the spin or the 2D order parameter, respectively. In the case that the weak antiferromagnetic order acts as the SBF it is predicted $\Delta T_c \propto m^2$.

In chapter 4 neutron-diffraction experiments on a series of $U(Pt_{1-x}Pd_x)_3$ single crystals ($x \leq 0.05$) are presented. It was found that the small-moment antiferromagnetic order

(SMAF), previously reported for pure UPt_3 is robust upon doping with Pd and persists till at least $x = 0.005$. The ordered moment grows from 0.018 ± 0.002 μ_B/U-atom for pure UPt_3 to 0.048 ± 0.008 μ_B/U-atom for $x = 0.005$. For the SMAF the Néel temperature, T_N, is approximately 6 K and, most remarkably, does not vary with Pd contents. The order parameter squared has an unusual quasi-linear temperature variation. For $x \geq 0.01$ a second antiferromagnetic phase with much larger ordered moments is found. For this phase at optimum doping ($x = 0.05$) T_N attains a maximum value of 5.8 K and the ordered moment equals 0.63 ± 0.05 μ_B/U-atom. $T_N(x)$ for the large-moment antiferromagnetic (LMAF) order follows a Doniach-type phase diagram. From this diagram we infer that the antiferromagnetic instability for the LMAF in $U(Pt_{1-x}Pd_x)_3$ is located in the range 0.5-1 at.% Pd.

In chapter 5 we report μSR experiments carried out on a series of $U(Pt_{1-x}Pd_x)_3$ samples with $x \leq 0.05$. For $x \leq 0.005$ the zero-field muon depolarisation is described by the Kubo-Toyabe function. However, the temperature variation of the Kubo-Toyabe relaxation rate $\Delta_{KT}(T)$ does not show any sign of the small-moment antiferromagnetic phase with $T_N \approx 6$ K, in contrast to previous reports. The absence of SMAF in the zero-field μSR signal provides evidence that the antiferromagnetic moments fluctuate at a rate >10 MHz, i.e. too fast to be detected by μSR, but slower than the time scale of the neutron-diffraction experiment ≈ 0.1 THz. For $0.01 \leq x \leq 0.05$ the muon depolarisation in the ordered state is described by two terms of equal amplitude: an exponentially damped spontaneous oscillation and a Lorentzian Kubo-Toyabe function. These terms are associated with antiferromagnetic order with substantial moments. The Knight-shift measured in a magnetic field of 0.6 T on single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ in the paramagnetic state shows two signals for $\mathbf{B} \perp \mathbf{c}$, while only one signal is observed for $\mathbf{B} \parallel \mathbf{c}$. The analysis of the Knight shift points to the presence of one muon localisation site $(0,0,z)$.

In chapter 6 we report the effect of Pd doping on the superconducting phase diagram of the unconventional superconductor UPt_3 as measured by (magneto)resistance, specific heat, thermal expansion and magnetostriction. Experiments on single- and polycrystalline $U(Pt_{1-x}Pd_x)_3$ for $x \leq 0.006$ show that the superconducting transition temperatures T_c^+ and T_c^- both decrease, while the splitting ΔT_c increases at a rate of 0.30 ± 0.02 K/at.%Pd. The B phase is suppressed first, near $x = 0.004$, while the A phase survives till $x \cong 0.007$. We find that $\Delta T_c(x)$ correlates with an increase of the weak magnetic moment $m(x)$ upon Pd doping. This provides further evidence for Ginzburg-Landau scenarios with magnetism as the symmetry breaking field (scenarios (i) and (ii)). Only for small splittings $\Delta T_c \propto m^2(T_c^+)$ ($\Delta T_c \leq 0.05$ K) as predicted. The results at larger splittings call for Ginzburg-Landau expansions beyond 4th order. The tetracritical point in the B-T plane persists until at least $x = 0.002$ for $\mathbf{B} \perp \mathbf{c}$, while it is rapidly suppressed for $\mathbf{B} \parallel \mathbf{c}$. Upon alloying the A and B phases gain stability at the expense of the C phase.