Ferromagnetism, superconductivity and quantum criticality in uranium intermetallics

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Summary

In recent years, quantum phase transitions (QPTs), defined as second-order phase transitions occurring at absolute zero temperature $T = 0$ K, have attracted much attention. In contrast to classical phase transitions which are driven by thermal fluctuations at finite temperatures, QPTs are driven by quantum fluctuations and occur as a function of a non-thermal control parameter, such as pressure, magnetic field or chemical substitution. Strongly correlated metals, notably heavy fermion systems (HFSs) based on the *f*-electron elements Ce, Yb, or U, are very suitable to investigate magnetic-to-nonmagnetic QPTs due to their low magnetic ordering temperatures and an exchange interaction which can be tuned relatively easily using a (non-thermal) external control parameter. Near the quantum critical point (QCP) where the QPTs takes place, these systems exhibit a novel ground state, the so-called non-Fermi liquid (NFL) state, which exhibits strong departures from the standard Landau Fermi liquid state commonly observed in strongly correlated metals. Moreover, the study of QCPs has led to the discovery of novel unconventional superconducting states located at the borderline of antiferro- or ferromagnetic order. These superconducting states cannot be explained by the standard Bardeen-Cooper-Schrieffer (BCS) theory. Evidence is at hand that these unconventional superconducting states are mediated by antiferro- or ferromagnetic spin fluctuations. Interestingly, superconductivity (SC) and ferromagnetism (FM) coexist in the itinerant magnets UGe_2, URhGe, UCoGe (this thesis) and possibly UIr when tuned to a QCP. This discovery came as a big surprise as normally SC and FM are competing ground states. Explanation of these new phenomena is a challenging task for both experimental and theoretical condensed matter physicists.

In this thesis, we have presented our search for ferromagnetic QCPs in the uranium-based intermetallics U*TX* where *T* is the transition metal Rh, Ru or Co, and *X* is the *p*-electron element Ge or Si. One of the results of special interest is that we have discovered a new ferromagnetic superconductor at ambient pressure UCoGe. In the next paragraphs we summarize the main results presented in this thesis.

In Chapter 4, evidence for a FM QCP in the polycrystalline $URh_{1-x}Ru_xGe$ series is presented. Upon alloying URhGe with Ru, ferromagnetic order (for $x = 0$ the Curie temperature $T_C = 9.5$ K), after an initial strengthening, is depressed and vanishes at the critical Ru concentration $x_{cr} = 0.38$. At x_{cr}, the thermal and transport properties show NFL temperature dependencies: (*i*) the specific heat $c/T \sim \ln(T/T_0)$ over one and a half decades in T, and the $\gamma(x)$-value (*i.e.* $c/T_{0.5K}$) attains a concomitant maximum; and (*ii*) the exponent n of the resistivity $\rho \sim T^n$ attains a minimum value with $n(x) = 1.2$. Additionally, the ordered moment m_0 is smoothly suppressed with increasing x. These observations provide convincing evidence that $URh_{1-x}Ru_xGe$ is the first 5*f*-electron system that can be tuned through to a continuous FM QCP at ambient pressure by doping. Upon application of a large magnetic field ($B < 10$ T) the $x_{cr} = 0.38$ compound is pushed away from the QCP and the Fermi-liquid state is recovered ($\rho \sim T^2$).

In Chapter 5, the magnetic instability in the $URh_{1-x}Ru_xGe$ series is further studied by investigating a single-crystalline sample with $x \approx x_{cr} = 0.38$ in high-magnetic field. In zero field, $\rho \sim T^n$ with $n \approx 1.2$ for a current applied along the principal axes, and the specific heat divided by temperature c/T shows a (quasi)logarithmic T dependence. The field dependence of the magnetization measured in magnetic fields up to 50 T at $T = 4.2$ K reveals a strong anisotropy of the easy-(*bc*) plane type with the *a*-axis as the hard-magnetization direction. The specific-heat data in a magnetic field ($B < 12$ T) applied along the *c*-axis show that the system can be tuned away from the FM QCP and that the Fermi-liquid state is recovered.

In Chapter 6, the evolution of ferromagnetism in polycrystalline samples of the $URhGe_{1-x}Si$ and $URh_{1-x}Co_xGe$ series is investigated via magnetization and resistivity measurements. For all samples, the resistivity data show a Fermi-liquid temperature dependence $\rho(T) \sim T^2$. In the case of Si doping on the Ge site, ferromagnetism is not significantly changed: $T_C = 9.5$ K is constant for $x \leq 0.2$ and increases slightly up to 10.4 K for $x = 1$. However, in the case of Co doping on the Rh site, T_C shows an increase up to 20 K for $x = 0.6$, is then depressed for larger Co concentration, and attains a value $T_C = 3$ K for $x = 1$. Surprisingly, superconductivity below 0.8 K is found to coexist with ferromagnetism at ambient pressure for the end-member of the Co-doping series, UCoGe.

Chapter 7 is devoted to a thorough characterization of the new ferromagnetic superconductor UCoGe. The magnetic, thermal and transport data taken on polycrystalline

UCoGe provide proof of the coexistence of bulk FM and SC. The magnetization data indicate polycrystalline UCoGe to be a weak itinerant ferromagnet with a Curie temperature T_C = 3 K and a small ordered moment of 0.03 μ_B. The bulk nature of the FM order is demonstrated by the large step in the coefficient of linear thermal expansion at T_C and the observation of a spontaneous muon precession frequency (ν = 2 MHz for $T \rightarrow 0$) in the whole sample volume below T_C. Proof for bulk SC is obtained by broad peaks in the specific heat and in the coefficient of thermal expansion at T_S^{bulk} = 0.66 K. Moreover, the thermal expansion and μSR data demonstrate coexistence of the FM and SC. The proximity to a magnetic instability, the absence of Pauli limiting in the upper critical field and the defect sensitivity of T_S indicate UCoGe is a triplet superconductor, with Cooper pairing most likely mediated by critical magnetic fluctuations. This is further corroborated by experiments on single-crystalline samples.

Experiments on single-crystalline UCoGe show that the magnetic and superconducting properties are strongly anisotropic. Magnetization measurements show that UCoGe is a uniaxial ferromagnet with the ordered moment m_0 = 0.06 μ_B directed along the orthorhombic c-axis. The upper-critical field $B_{c2}(0)$ determined in transport measurements has a value $B_{c2}(0) \approx 5$ T for $B \parallel a, b$ which is much larger than the value $B_{c2}(0) \approx 0.5$ T obtained for $B \parallel c$. The large anisotropy in B_{c2} supports the ideal of unconventional superconductivity in UCoGe with an axial SC gap function with nodes along the moment direction m_0. In the B_{c2} curves, a pronounced upward curvature or kink for $B \parallel b$ or $B \parallel a$ is observed. These data from UCoGe may be the first experimental indication for the existence of a two-band ferromagnetic superconductor.

In Chapter 8, we have reported the depression of FM and SC in the polycrystalline $UCoGe_{1-x}Si_x$ series via magnetization, ac-susceptibility, and transport measurements. Upon Si doping, ferromagnetic order and superconductivity are both depressed and vanish at the same critical concentration $x_{cr} \approx 0.12$. Near x_{cr}, a NFL temperature dependence of the electrical resistivity is observed and the exponent $n(x)$ attains a minimum value $n \approx 1$. The magnetization data reveal the ordered moment is depressed smoothly, which points to a continuous FM quantum phase transition at x_{cr}. Superconductivity is confined to the ferromagnetic phase, which provides further support for the existence of magnetically-mediated superconductivity. These results offer a unique route to investigate the emergence of superconductivity near a FM QCP at ambient pressure.