Non-fermi liquid behaviour in uranium-based heavy-fermion compounds

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8. ■ Concluding remarks

8.1. U_2Pt_2In

In this thesis, it has been shown that U_2Pt_2In is the first *stoichiometric* U-based compound that exhibits non-Fermi liquid (NFL) behaviour *at ambient pressure*. Research on stoichiometric compounds prepared in a single-crystalline form is highly desirable in the study of NFL behaviour because the effects of disorder may not dominate the physics. This provided a strong motivation to carry out an extensive study on U_2Pt_2In in order to determine its NFL properties. Nevertheless, the rather difficult metallurgy of U_2Pt_2In imposed its restrictions on the research carried out.

The main results obtained on U_2Pt_2In are summarized below.

• Polymorphism was found in U_2Pt_2In [1]: while polycrystalline samples form in the U_3Si_2-type of structure, single-crystals form in the Zr_3Al_2-type of structure (superstructure of the U_3Si_2 type with a doubling along the c-axis).

• When tracing the magnetic ordering temperatures of the U_2T_2X family of compounds versus the square of the conduction-electron - f-electron hybridization matrix element in a Doniach-like diagram, one finds that U_2Pt_2In is located at the border line between magnetic and non-magnetic compounds, which suggests that U_2Pt_2In is near a magnetic instability.

- The magnetic susceptibility of U_2Pt_2In is weakly anisotropic and follows a (modified) Curie-Weiss behaviour at high temperatures. At low temperatures, however, an anomalous behaviour is observed: χ_c goes through a maximum at 7.9 K, which is attributed to the stabilization of short-range antiferromagnetic (AF) correlations, while χ_a increases as $1-bT^{0.7}$ for 2 K $\leq T <$ 10 K. This is at variance with the standard Fermi liquid (FL) behaviour, which predicts χ to attain a constant value at low temperatures.

- The resistivity of U_2Pt_2In is highly anisotropic below about 80 K with $\rho_c > \rho_a$. At the lowest temperatures, the resistivity does not follow the FL quadratic temperature dependence, instead $\rho_a \sim T^{1.25}$ and $\rho_c \sim T^{0.9}$. The residual resistivities are unusually high: $\rho_{0,a} \approx 115\ \mu\Omega cm$ and $\rho_{0,c} \approx 210\ \mu\Omega cm$. However, structure refinements from X-ray [1] and neutron-diffraction [2] experiments indicate a high sample quality. Also, the large difference between $\rho_{0,a}$ and $\rho_{0,c}$ shows that ρ_0 is largely determined by other scattering mechanisms than impurity or defect scattering. It should also be noted that the higher resistivity values are found for a current along the c-axis, which is the axis along which the AF fluctuations stabilize.

- Magnetoresistance (MR) experiments show a gradual increase of the resistivity exponent α (defined as $\rho \sim T^\alpha$ as $T \to 0$) with increasing magnetic field strength. At 8 T, $\alpha = 2$ as expected for a FL. There are two different contributions to the MR: a negative one (associated with spin effects) and a positive one (associated with orbital effects). At low fields, the negative contribution is dominant, except in the case $B \parallel I \parallel a$, whereas at high fields the positive contribution becomes more and more important. The negative contribution to the MR is more important for $I \parallel c$ than for $I \parallel a$. However, no satisfactory explanation for this anisotropy can be offered at the moment. Additional evidence that the high residual resistivity in U_2Pt_2In is not determined by crystallographic disorder is provided by the strong field dependence of ρ_0.

- The specific heat of U_2Pt_2In provides solid evidence for the classification of this heavy-fermion compound as a NFL. The specific heat shows a pronounced diverging behaviour of the type $c/T \sim -\ln(T/T_0)$ over almost two decades of temperature. The low-temperature specific heat measured in a magnetic field is dominated by a strong contribution of the In nuclear moments. The same logarithmic divergency is observed in the specific heat measured on polycrystalline samples. As these crystallize in the simpler U_3Si_2-type of structure, the NFL behaviour is not directly related to the presence of two inequivalent U positions, as present in the single crystals with the Zr_3Al_2-type of structure.

• The thermal-expansion coefficient of U_2Pt_2In becomes anisotropic below about 12 K, in a way that the c-axis shrinks more rapidly than the a-axis upon formation of the heavy-fermion bands.

• Absence of weak magnetic order, at least down to 0.05 K, is confirmed by means of muon spin relaxation and rotation (μSR) experiments. Besides a static magnetic component originating from the In nuclear moments, the μSR spectra below 10 K reveal the presence of magnetic fluctuations. No evidence was found for Kondo disorder.

• Resistivity experiments under hydrostatic pressure indicate a recovery of the FL $\rho \sim T^2$ behaviour at low temperatures for $I \parallel a$. This is consistent with predictions from a transport theory for heavy-fermion compounds near an AF quantum critical point (QCP) [3]. The anisotropy in the resistivity is strongly enhanced under pressure, as follows from the increase of the ratio ρ_c/ρ_a measured for $0.3 \text{ K} \leq T < 300 \text{ K}$. Due to the enhancement of the anisotropy, the resistivity curves for $I \parallel c$ do not show a $\rho \sim T^2$ behaviour but a low-temperature minimum.

One of the main issues that arise when discussing the properties of NFL compounds is the responsible mechanism for this behaviour. Although in the current state-of-the-art NFL physics no definite answers can be provided, one most important distinction can often be made: whether the NFL behaviour is due to a single-ion or a cooperative mechanism.

The location of U_2Pt_2In at the border line between magnetic and non-magnetic compounds in the Doniach diagram for the U_2T_2In family of compounds, suggests a proximity to an AF QCP. Resistivity measurements under pressure carried out on U_2Pt_2In and U_2Pd_2In yield strong support for this hypothesis. Considering the absence of magnetic order down to 0.05 K (as evidenced from the μSR experiments) and the important finding that pressure leads to the recovery of the FL behaviour, one cannot exclude that U_2Pt_2In is even located *at* the AF QCP. In order to investigate this further, specific-heat experiments under pressure would be most welcome. As the specific-heat coefficient γ is related to the coherence temperature T_{coh} [4], the observation and evolution of the FL γ coefficient with pressure would provide valuable information on the recovery of the FL state near the QCP.

It is important to notice that the observed divergency of the specific heat, $c/T \sim -\ln(T/T_0)$ is not consistent with an AF QCP, but rather indicates a ferromagnetic (FM) QCP [5]. However, a logarithmic divergency of c/T is allowed for an AF QCP in a two-dimensional (2D) system. This appears to apply to compounds like $CeCu_{5.9}Au_{0.1}$ [6] and $CeNi_2Ge_2$ [7], where inelastic neutron

scattering (INS) experiments have provided evidence for a spectrum of strongly anisotropic magnetic fluctuations, with a quasi-2D nature. INS experiments on U_2Pt_2In could possibly elucidate the nature of the magnetic fluctuations further.

In order to investigate the evolution of magnetic order near U_2Pt_2In in the Doniach phase diagram, the study of single-phase samples of e.g. $(U_{1-x}Th_x)_2Pt_2In$ and $U_2(Pt_{1-x}Pd_x)_2In$ is helpful If Pd or Th doping results in the emergence of magnetism, strong evidence for U_2Pt_2In being at a QCP is obtained. One could then also study such doped samples under pressure, which should lead to the suppression of magnetic order and to the appearance of the FL state.

The origin of the strongly anisotropic character of the resistivity of U_2Pt_2In remains puzzling. Its enhancement under pressure indicates that the anisotropy is not inherent to the crystal structure since the compressibility is almost isotropic. Resistivity measurements on single crystals of the non-magnetic compound Th_2Pt_2In would be useful to address this issue. These could also serve to obtain an estimate for the phonon contribution to the resistivity of U_2Pt_2In. In addition, specific-heat experiments on Th_2Pt_2In should be carried out in order to estimate the phonon contribution to $c(T)$. This would enable a more precise determination of the temperature up to which a logarithmic divergency is present in the electronic specific heat of U_2Pt_2In.

The high residual resistivity of U_2Pt_2In raises the question of the role of disorder in the NFL properties. The dependence of ρ_0 on the current direction, as well as its field and pressure variations, indicate that impurity and defect scattering are not the dominant mechanisms leading to the high values of ρ_0. Even though X-ray and neutron-diffraction experiments indicate a good single-crystalline quality, the use of a local probe to measure the near-neighbour bond-length distributions, like (synchrotron radiation) X-ray absorption fine-structure (XAFS) experiments, could be helpful to determine the exact amount of disorder. For instance, neutron-diffraction experiments carried out on UCu_4Pd were inconclusive regarding the presence of disorder [8]. However, XAFS experiments on the same sample revealed the presence of Pd/Cu site interchange [9]. It was concluded that the amount of disorder observed was sufficient for the Kondo disorder model to apply.

Although there is no evidence for Kondo disorder in U_2Pt_2In, a good test for the Kondo disorder model is obtained by a comparison of the μSR and NMR line widths [10]. As shown in this work, the small frequency shifts observed in the transverse-field (TF) μSR spectra of U_2Pt_2In, require that a full analysis of the TF line widths can only be accomplished with samples with a well-defined geometry in order to account for demagnetizing effects.

A further study of the NFL behaviour of U_2Pt_2In may hopefully be carried out on a "second generation" of single crystals, with a minimum amount of disorder. Single crystals with a lower residual resistivity should be prepared. Therefore, one should look into single-crystal growth methods other than the mineralization technique, by which possibly crystals of higher quality can be produced.

8.2. Related compounds

The presence of a QCP has also been investigated for the compounds $U_3Ni_3Sn_4$ and $U(Pt_{1-x}Pd_x)_3$.

Specific-heat measurements carried out on the stoichiometric compound $U_3Ni_3Sn_4$ show the presence of a NFL regime in the temperature range 0.5-5 K and a crossover to a FL ground state below 0.4 K. The divergency of the specific heat in the NFL regime is of the type $c/T \sim \gamma_0 - \alpha T^{1/2}$, which is consistent with the proximity to an AF QCP in a 3D system [5]. Both the NFL c/T divergence and the spin-fluctuation term in the FL specific heat, have the same characteristic temperature T_0 of 10 K. The pressure dependence of the temperature below which the FL regime in the resistivity is attained, is well described in terms of a transport theory for nearly AF metals [3]. These results indicate that by a small lattice expansion, equivalent to a negative pressure of about -0.04 GPa, $U_3Ni_3Sn_4$ may be tuned to the QCP. The isostructural compound $U_3Cu_3Sn_4$ is an antiferromagnet with $T_N = 13$ K and its unit-cell volume is about 5% larger than that of $U_3Ni_3Sn_4$. Therefore, it would be highly interesting to check the existence of an AF QCP by studying samples in which small amounts of Ni are replaced by Cu.

μSR experiments on the system $U(Pt_{1-x}Pd_x)_3$ indicate a new type of QCP in the phase diagram: at the critical concentration $x_c = 0.006$, unconventional superconductivity is suppressed and a large-moment antiferromagnetic (LMAF) phase emerges. The fact that the superconducting wave-function has odd parity suggests that doping with Pd leads to a shift of the spectral weight from FM to AF fluctuations. The observed competition between superconductivity mediated by FM fluctuations and static AF order, is in contrast with superconductivity mediated by AF interactions in materials like $CePd_2Si_2$ close to the QCP [11]. The phase diagram of $U(Pt_{1-x}Pd_x)_3$

is similar to the phase diagrams observed for high-temperature superconductors with the peculiarity that the critical points $T_N \to 0$ and $T_c \to 0$ coincide.

Resistivity measurements carried out on $U(Pt_{1-x}Pd_x)_3$ show values of the exponent α equal to 1.8 and 1.6 for samples with $x = 0.004$ and $x = 0.007$, respectively [12]. These values are inconclusive with respect to the type of QCP in the system ($\alpha = 3/2$ and $\alpha = 5/3$ are predicted for the AF and FM QCP, respectively). A systematic study of the resistivity exponents in samples with x around 0.006 is required to clearly determine the type of magnetic fluctuations at the QCP.

The origin of the NFL behaviour observed in $URh_{1/3}Ni_{2/3}Al$ might be of the single-ion type. $URh_{1/3}Ni_{2/3}Al$ shows a diverging specific heat of the type $c/T \sim -\ln(T/T_0)$ below about 6 K. Below 10 K the resistivity increases as $\rho \sim 1 - aT^n$ with α about 1. The low-temperature resistivity increase is an indication that the mechanism responsible for the NFL behaviour is of the single-ion type. In fact, a loss of coherence is predicted to occur at low temperatures within the Kondo disorder model. This model also predicts a logarithmic divergency of c/T and the linear increase of the resistivity below T_K, as observed for $URh_{1/3}Ni_{2/3}Al$. A detailed structural analysis on single-phase samples is required to establish the amount of disorder in this compound.

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