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

de Lemos Correia Estrela, P.M.

Citation for published version (APA):
de Lemos Correia Estrela, P. M. (2000). Non-fermi liquid behaviour in uranium-based heavy-fermion compounds

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: http://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.
6. Probing $U_2Pt_2In$ in the Doniach phase diagram

In order to probe the (Doniach) phase diagram of $U_2Pt_2In$, resistivity experiments under pressure and Th-doping studies were carried out. The main role of replacing small amounts of U by Th is an expansion of the unit cell, while applying hydrostatic pressure gives the opportunity to study the effects of unit-cell volume reduction. This in turn changes the conduction-electron-$f$-electron hybridization and therefore the exchange interaction $J$. Assuming that $U_2Pt_2In$ is close to a quantum critical point, it is of interest to study the eventual emergence of magnetic order with Th doping or the recovery of the Fermi-liquid state under pressure.

Experiments under pressure were also performed on $U_2Pd_2In$ in order to study the suppression of the antiferromagnetic ground state. As mentioned in Section 4.1, $U_2Pd_2In$ is an antiferromagnetic heavy-fermion material. On the magnetic side of the Doniach diagram (see Figure 6.17a in Section 6.5), it is the compound closest to $U_2Pt_2In$.

The magnetic ground states of some of the 2:2:1 compounds, prepared in single-crystalline form, were investigated, which confirmed their location in the Doniach diagram (Section 4.1.2). The results are presented in Section 6.4.
6.1. Pressure effects on the resistivity of U$_2$Pt$_2$In

Resistivity measurements under pressures up to 1.8 GPa and in magnetic fields up to 8 T. were carried out in the temperature range 0.3-300 K. The data were taken on bar-shaped single-crystals of U$_2$Pt$_2$In by using a standard four-probe method. Experiments under pressure were carried out by using the CuBe piston cylinder-type clamp cell described in Section 2.3.1. The pressure values presented in this chapter were corrected for an empirically established efficiency of 80%. For each pressure, the resistance curves were normalized to 1 at room temperature. In this way, possible changes in the geometrical factor (mainly in the distance between the voltage contacts) are taken into account.

As shown in Section 4.2.2, the zero-pressure resistivity curve of U$_2$Pt$_2$In for $I \parallel c$ follows a power law of the type $\rho \sim T^\alpha$ with $\alpha \approx 0.9$ at the lowest temperatures. Upon increasing the pressure, $\alpha$ gradually increases for $p < 1.0$ GPa (Figure 6.1). However, as the pressure is increased above 1.0 GPa, a minimum in the resistivity develops, which becomes more pronounced at the highest pressures.

![Figure 6.1 - Low-temperature dependence of the resistivity of U$_2$Pt$_2$In for $I \parallel c$ at different pressures.](image-url)
In order to investigate whether the observed minimum has a magnetic origin, measurements of $\rho(T)$ in magnetic fields ($B \parallel I$) were performed at $p = 1.8$ GPa (Figure 6.2a). $T_{\text{min}}$ decreases smoothly with the strength of the applied magnetic field: $T_{\text{min}} \sim 4.8$ K for $B = 0$ and $T_{\text{min}} \sim 2.2$ K for 8 T. Also, the minimum becomes less pronounced with increasing field. Due to the limited range of temperatures and fields available and the relative uncertainty in determine $T_{\text{min}}$, it is not possible to clearly trace the evolution of $T_{\text{min}}$ with $B$. Figure 6.2b shows that the data can be fitted with a linear dependence, $B = B_0(1 - T_{\text{min}}/T_0)$ with $B_0 = 14.2(7)$ T and $T_0 = 4.7(1)$ K. A typical antiferromagnetic-like field dependence of the ordering temperature, $B = B_0[1 - (T_{\text{min}}/T_0)^2]^{\beta}$ with $\beta = 0.8$, $B_0 = 9.2(8)$ T and $T_0 = 4.4(2)$ K, cannot be excluded. However, the latter produces a poorer fit. The field effect on the resistivity curves suggests that $T_{\text{min}}$ has a magnetic origin. Whether $T_{\text{min}}$ is associated with magnetic ordering remains unresolved.

![Figure 6.2](image)

Figure 6.2 - (a) Temperature dependence of the resistivity of $\text{U}_2\text{Pt}_3\text{In}$ for $I \parallel c$ and $p = 1.8$ GPa at different longitudinal fields. (b) Field dependence of $T_{\text{min}}$.

It is of interest to note, however, that the structure in $\rho(T)$ at $p = 1.8$ GPa resembles, to a certain extent, the one of an antiferromagnetic phase transition of the spin-density wave type, as observed e.g. in the heavy-fermion antiferromagnets $\text{URu}_2\text{Si}_2$ ($T_N = 14$ K) [1] and $\text{U(Pd}_{0.95}\text{Pd}_{0.05})_3$ ($T_N = 5.8$ K) [2]. In these compounds, $\rho(T)$ increases below $T_N$ because of the opening of an
energy gap. At about 0.9\(T_N\), \(\rho(T)\) develops a local maximum, below which the resistivity drops because of the ordered structure. The data in Figure 6.1 for \(U_2Pt_2In\) under pressure, show no local maximum at temperatures \(T > 0.15T_{\text{min}}\). If \(T_{\text{min}}\) is indeed associated with magnetic ordering, the absence of a local maximum is possibly due to the large value of the residual resistance of the samples.

The emergence of a magnetic component to \(\rho(T)\) upon applying pressure is quite surprising. Applying pressure on a compound at the magnetic instability normally leads to an increase of the control parameter \(J\) and brings the compound in the non-ordering Fermi-liquid regime. A possible explanation for this unusual behaviour might be offered by assuming that the control parameter \(J\) is not governed by the volume, but by the \(c/a\) ratio of the tetragonal unit cell. This is suggested by the comparison of the unit-cell volumes and \(c/a\) ratios of \(U_2Pt_2In\) and the antiferromagnets \(U_2Pd_2In\) \((T_N = 37\,\text{K})\) and \(U_2Pt_2Sn\) \((T_N = 15\,\text{K})\) [3]. Whereas the unit-cell volume of \(U_2Pt_2In\) is smaller than that of \(U_2Pd_2In\) and larger than that of \(U_2Pt_2Sn\), the \(c/a\) ratio is always smaller (for simplicity the doubling of the crystal structure along the \(c\)-axis in single-crystalline \(U_2Pt_2In\) has not been taken into account) [4]. Thus the appearance of magnetic ordering under pressure could be the result of an increase of the \(c/a\) ratio. This in turn requires the compressibility to be anisotropic.

In order to determine the compressibility of \(U_2Pt_2In\), X-ray diffraction experiments were performed under pressure at RT by T. Naka (NRIM-Tsukuba, Japan). The resulting lattice parameters under pressure are presented in Figure 6.3. Both the \(c\) and \(a\) parameters decrease almost linearly with pressure. The compressibility values are almost identical for both axes: \(\kappa_a = 0.220 \, \text{Mbar}^{-1}\) and \(\kappa_c = 0.242 \, \text{Mbar}^{-1}\), while the volume compressibility amounts to \(\kappa = 0.682 \, \text{Mbar}^{-1}\). This means that between \(p = 0\) and 1.8 GPa, the \(c/a\) ratio does not change significantly (it actually decreases 0.04%). This invalidates the hypothesis that the \(c/a\) ratio is the control parameter for \(U_2Pt_2In\) in the Doniach diagram.

Resistivity experiments under pressure were also carried out for the current along the \(a\)-axis (Figure 6.4). Interestingly, the results differ very much from the results for \(I \parallel c\). In Section 4.2.2 it was shown that the zero-pressure resistivity curve for \(I \parallel a\) follows a power law \(\rho \sim T^\alpha\) with \(\alpha = 1.25\) at the lowest temperatures. Upon increasing the pressure, \(\alpha\) gradually increases and no minimum in \(\rho(T)\) is observed up to \(p = 1.8\, \text{GPa}\).
Figure 6.3 - Pressure dependence of the lattice parameters of U$_2$Pt$_3$In.

Figure 6.4 - Low-temperature dependence of the resistivity of U$_2$Pt$_3$In for $I \parallel a$ at different pressures.

The pressure dependence of the exponent $\alpha$ is shown in Figure 6.5a. The Fermi-liquid value of $\alpha = 2$ is reached at $p \sim 1.0$ GPa. Upon further pressure increase, there is an increase of
the temperature $T_{FL}$ below which a $\rho \sim T^2$ behaviour is observed. The pressure dependence of $T_{FL}$ is shown in Figure 6.6b. According to the theory of Rosch [5] for the resistivity of heavy-fermion materials close to an antiferromagnetic quantum critical point (see Section 2.4), the temperature below which a $\rho \sim T^2$ behaviour is observed should vary initially as $T_{FL} = a_1 (p-p_c)$ and at higher pressures as $T_{FL} = a_{1/2} (p-p_c)^{1/2}$, where $p_c$ is the pressure at which the QCP occurs. The region where the linear behaviour is observed depends on the amount of disorder $x$ in the system ($x \approx 1/RRR$). The $T_{FL}(p)$ behaviour in U$_2$Pt$_2$In for $I \parallel a$ is consistent with a linear dependence with $p_c = 0$. A $\rho \sim T$ region is predicted to occur for $x < T/\Gamma < x^{1/2} (x < 1)$, where $\Gamma$ defines the temperature scale where the spin fluctuations are destroyed ($\Gamma \sim T_K$ or $T_{coh}$). Defining a region where $\rho \sim T$ for the different pressure values (see Figure 6.6a for $p = 1.8$ GPa), the diagram of Figure 6.6b can be constructed, from which it follows that $x = 0.34$ and $\Gamma = 8.1$ K. The fact that $x < 1/RRR$ indicates that the residual resistivity observed in U$_2$Pt$_2$In for $I \parallel a$ is not uniquely due to impurity or defect scattering. The distance to the QCP is given by $r = \zeta p$ with $\zeta = 0.11$ GPa$^{-1}$.

Figure 6.5 - Pressure dependence of the resistivity exponent $\alpha$ for $I \parallel a$. 
Therefore, the resistivity data under pressure for $I \parallel a$ are consistent with the location of U$_2$Pt$_2$In at or close to a QCP. Pressure increases the exchange parameter $J$, shifting the compound towards the non-ordering Fermi-liquid regime. These results for $I \parallel a$ indicate that the minimum in $\rho_c(T)$ is most probably not related to magnetic order.

At high temperatures, the resistivity curves also have different characteristics for the two crystallographic directions. The anisotropy in the transport properties increases with pressure in the whole temperature range studied, as shown in Figure 6.7. As the pressure is increased, $\rho_a$ tends to decrease, while $\rho_c$ increases, thus the difference $\rho_c - \rho_a$ increases (Figure 6.7b). Assuming that the difference $\rho_c(T) - \rho_a(T)$ is due to an extra resistivity component only present for $I \parallel c$, the effect of pressure is to enhance this component, without significantly changing its temperature dependence. This implies that the minimum in $\rho_c(T)$ is probably a consequence of the enhancement of a resistivity component for $I \parallel c$. 

Figure 6.6 - (a) Temperature dependence of $\rho_a - \rho_{a,0}$ for U$_2$Pt$_2$In under $p = 1.8$ GPa. (b) Pressure dependence of the temperature below which $\rho - T^2$ and the temperature interval where $\rho - T$. The lines are defined in the text.
Probing \( U_2Pt_2In \) in the Doniach phase diagram

The temperature \( T_m \) at which a maximum in the resistivity occurs increases strongly for \( I \parallel a \), but decreases slightly for \( I \parallel c \). According to the theory of Yoshimori-Kasai for the dense Kondo system [6], the temperature \( T_m \) is approximately proportional to the Kondo temperature \( T_K \). The volume dependence of \( T_K \) is given by a Grüneisen parameter defined as

\[
\Gamma_K = -\left. \frac{\partial \ln T_K}{\partial \ln V} \right|_{V=V_0},
\]

where \( V_0 = V(p=0) \). Since \( T_m \propto T_K \), it follows that [7]

\[
\Gamma_K = -\left( \frac{V-V_0}{V_0} \right)^{-1} \ln \left( \frac{T_m(P)}{T_m(0)} \right).
\]

The volume change is \( \Delta V/V_0 = -\kappa p \). The values of \( \ln[T_m(p)/T_m(p=0)] \) are plotted versus \( \Delta V/V_0 \) in Figure 6.8. The slope of the straight lines through the data points yields the value of \( \Gamma_K \). Not considering the point for \( I \parallel a \) at \( p = 1.8 \) GPa, where the maximum is not well defined, a value of \( \Gamma_{K,a} = 49.7 \pm 7.1 \) can be estimated for \( I \parallel a \). This value is close to the values of 59 and 65 reported for the heavy-fermion compounds CeInCu_2 and CeCu_6, respectively [7]. This indicates that the strong pressure dependence of \( T_m \) observed in \( U_2Pt_2In \ (I \parallel a) \) is not unusual. The increase of \( T_K \) reflects the increase of the conduction electron - \( f \)-electron hybridization and, therefore, the
increase of the exchange parameter $J$. This is in agreement with the appearance of a FL $\rho \sim T^2$ behaviour at low temperatures.

The slight decrease of $T_m$ for $I \parallel c$, on the other hand, is unexpected. A similar analysis of the pressure dependence of $T_m$ as performed for $I \parallel a$, yields a Grüneisen parameter $\Gamma_{K,c} = -6.8(7)$. Since $T_K$ should not depend on the current direction, this value of $\Gamma_{K,c}$ indicates that the maximum observed in $\rho_c(T)$ is not due uniquely to the Kondo effect. The enhancement of an extra anisotropic resistivity component under pressure (mainly present for $I \parallel c$) occurs even at high temperatures (Figure 6.7b). Therefore $T_m$ cannot be taken to be proportional to $T_K$ for this current direction.

![Graph](image)

**Figure 6.8** - Values of $\ln(T_m(p)/T_{m,0})$ as a function of volume change $[-V(p)/V(0)]$ for $U_2Pt_2In$.

Resistivity measurements under pressure were also performed on other single-crystals of $U_2Pt_2In$. Although all the crystals were cut from the same batch, there is a slight sample dependence of some of the features. The data under pressure presented so far were all measured on one single crystal (sample #1). It had a platelet shape such that the current could be applied along the a- and c-axis. Measurements on a second crystal (sample #2) with $I \parallel c$ and on a third crystal (sample #3) with $I \parallel a$, confirm the overall behaviour of the resistivity curves under
pressure: an increase of the transport anisotropy, the development of a minimum in $\rho_\perp(T)$ at low temperatures and the recovery of a $T^2$ term in $\rho_\parallel(T)$.

However, the values of $T_{\text{min}}$ are different for samples #1 and #2. For sample #2, the minimum shows up for $p > 1.2$ GPa and attains values of $T_{\text{min}} \approx 1.2$ K at $p = 1.5$ GPa and $T_{\text{min}} \approx 2.1$ K at $p = 1.8$ GPa [8]. The field dependence of $T_{\text{min}}$ at $p = 1.8$ GPa has the same form as shown in Figure 6.2b for sample #1. Also, at high temperatures, the absolute values of the maximum $T_{m}$ are slightly different, but the pressure dependence of $T_{m}$ remains the same. The Grüneisen parameters obtained are $\Gamma_{K,\parallel} \approx -4.5$ (sample #3) and $\Gamma_{K,\perp} \approx -2$ (sample #2). These values do not differ significantly from the values obtained for sample #1.

### 6.2. Studies of Th doping in $U_2Pt_2In$

Resistivity studies on polycrystalline $(U_{1-x},\text{Th}_x)_2Pt_2In$ samples ($0 \leq x \leq 1$) reported in the literature [9,10], indicate that the low-temperature behaviour of $\rho(T)$ gradually changes, from linear in $T$ as observed for undoped $U_2Pt_2In$ towards a quadratic behaviour as the Th content increases. The resistivity of the sample with $x = 0.1$ shows a knee at about 19 K, which suggests magnetic ordering. However, it should be noted that a small amount of UPt impurity phase, possibly present in the sample, could also give rise to a similar anomaly, as UPt has two ferromagnetic transitions at 19 K and 27 K [11].

In this section, results are presented on polycrystalline samples of $(U_{1-x},\text{Th}_x)_2Pt_2In$ with $x = 0, 0.03, 0.08$ and 0.1. The samples were prepared by L.C.J. Pereira (Technological and Nuclear Institute, Portugal) by arc-melting the constituents under a purified Ar atmosphere. The crystallographic structure and the lattice parameters were determined by X-ray diffraction [12]. It was found that all polycrystalline samples crystallize in the simple $U_2Si_2$-type of structure. The lattice parameters and unit-cell volume are given as a function of $x$ in Figure 6.9. The unit-cell expansion is almost isotropic. The volume increases linearly with $x$. However, the prepared materials were not single phase. All samples have small amounts of UPt and/or UPtIn as impurity phases. UPtIn is an antiferromagnet with $T_N = 15$ K [13]. Due to the non-single phase
character of the Th-doped samples, the results presented here should be considered as preliminary.

![Graph](image)

**Figure 6.9** Lattice parameters and unit-cell volume of polycrystalline \((U_{1-x}Th_x)_2Pt_2In\). Data taken from Ref. 12.

As mentioned in Section 4.3.3, specific-heat measurements on polycrystalline \(U_2Pt_2In\) show the same logarithmic divergence of \(c/T\) as observed on single crystals. However, in the polycrystals small amounts of UPtIn and UPt impurity phases were detected by X-ray diffraction and magnetization measurements. These small amounts of impurity phases (less than 5 w.%) were not detected in the specific heat. Due to the high electronic specific heat of \(U_2Pt_2In\), the low-temperature contribution from the impurity phases is unobservable.

Magnetization measurements were performed on \((U_{0.95}Th_{0.05})_2Pt_2In\) in the temperature range 2-300 K and in fields up to 5 T. Both the temperature and field dependence of the magnetization \(M\) reveal the presence of a ferromagnetic component with \(T_C \approx 25\) K, consistent with an UPt impurity phase. The \(M(B)\) curves are shown in Figure 6.10a. Below 30 K, the saturation of the ferromagnetic contribution of the UPt impurity phase in low fields is clearly visible. At higher fields \(M(B)\) is almost linear. Assuming that the high-field linear behaviour \((B > 1\) T\) is only due to the main phase, the susceptibility \(\chi_{DC}\) of \((U_{0.95}Th_{0.05})_2Pt_2In\) can be extracted by taking the slopes of the \(M(B)\) curves at high fields. The low temperature values of
the magnetization obtained by extrapolating the linear high-field behaviour to $B=0$ are consistent with the presence of about 3 w.\% of UPt, in agreement with the value estimated from the X-ray structure refinement. The results of $\chi_{dc} = \mu_0 dM/dB$ are shown in Figure 6.10b. A maximum in $\chi_{dc}(T)$ appears at low temperatures. The maximum possibly indicates antiferromagnetic ordering at $T_N \sim 8(2)$ K. However, it is also possible that the maximum is related to the presence of short-range antiferromagnetic correlations, as in the case of U$_2$Pt$_2$In with $B \parallel c$. The high-temperature ($T \geq 25$ K) $\chi_{dc}(T)$ curve follows a modified Curie-Weiss law with $\chi_0 = 1.4(1) \times 10^{-8}$ m$^3$/molU, $\mu_{\text{eff}} = 1.8(4) \mu_B/\text{U}$ and $\theta = -29(2)$ K. However, it should be stressed that due to the presence of impurity phases, the $\chi_{dc}(T)$ curve should be considered as indicative and its analysis should be taken with care.

$$\chi = \frac{C}{T - \theta}$$

Figure 6.10 - (a) Field dependence of the magnetization of (U$_{0.95}$Th$_{0.05}$)$_2$Pt$_2$In. (b) Temperature dependence of the high-field slope of the $M(B)$ curves. The line represents a modified Curie-Weiss law.

Zero- and transverse-field $\mu$SR experiments were carried out on the same (U$_{0.95}$Th$_{0.05}$)$_2$Pt$_2$In sample in the temperature range 2-200 K. Reasonable fits to the zero- and 0.01 T transverse-field spectra can be obtained with a Gaussian or exponentially damped depolarization function. An important feature of the data is that part of the asymmetry is missing. In the TF-$\mu$SR spectra with $B = 0.01$ T, the full asymmetry $A \sim 0.24$ is observed above 15 K.
This value of the asymmetry is determined by the experimental geometry and sample size. Below about 15 K the asymmetry drops to a value of \( A \sim 0.15 \), which implies that an additional fast component to the \( \mu \)SR signal appears, which has not been accounted for in the fits. A fast component, i.e., a component with a high depolarization rate, must have its origin in a magnetically ordered phase. The drop in the asymmetry near 15 K (\( \Delta A \sim 0.09 \)), is too high to be attributed to the presence of \( \sim 3 \) wt.% of UPt impurity phase.

Although no reliable fits to the spectra at low temperature were obtained, a common feature appeared in all fit attempts: as the temperature is decreased below 25 K, the zero-field depolarization rate increases and has a tendency to diverge at about 10 K, below which it regains a low value. A divergence of the depolarization rate is often associated with an antiferromagnetic transition [14], which is in agreement with the antiferromagnetic-like \( \chi_{\text{DC}}(T) \) curve.

Transverse field \( \mu \)SR experiments in 0.01 T were carried out on \((U_{0.97}Th_{0.03})_2Pt_2In\) for \( 2.5 \leq T \leq 100 \) K. In contrast to the results obtained on the \( x = 0.05 \) sample, no significant loss of asymmetry was found at low temperatures. Best fits to the spectra were obtained using a damped-Gaussian depolarization function: \( P(t) = P_G(t) \exp(-\lambda_F t) \) with \( P_G(t) = \exp(-\Delta_G^2 r^2/2)\cos(\omega t) \). Here, the temperature-independent Gaussian relaxation rate was found to be \( \Delta_G = 0.17 \mu s^{-1} \), which possibly accounts for the In nuclear contribution. This value is much reduced with respect to the one observed on \( U_2Pt_2In \) (\( \Delta_G = 0.34 \mu s^{-1} \)). It should be noted that the crystallographic structures are different and therefore different muon stopping sites might be involved, leading to a different value of \( \Delta_G \) from the distribution of In nuclear moments. The damping of the Gaussian term is observed below 10 K with \( \lambda_F \) increasing below this temperature. Whether this increase is associated with a dynamic or a static magnetic component cannot be determined from the available data set.

Recently, polycrystalline \((U_{1-x}Th_x)_2Pt_2In\) samples with \( x = 0.03, 0.05, 0.08 \) and 0.1 were prepared with lower amounts of impurity phases [12]. Specific-heat measurements carried out by G. Bonfait (Technological and Nuclear Institute, Portugal) on these samples do not confirm magnetic order above 2 K. It would be of interest to carry out \( \mu \)SR experiments on these samples as well, in order to look for weak magnetism. Clearly, single-phase samples are highly desirable to reliably determine the presence of magnetic order.
6.3. $U_2Pd_2In$ - resistivity under pressure

As mentioned in Section 4.1, $U_2Pd_2In$ is an antiferromagnet with $T_N = 37$ K [15,16]. The U magnetic moments are confined to the basal plane and form a non-collinear magnetic structure. At 10 K, $\mu_U = 1.6 \mu_B$ [15].

Resistivity measurements under pressure were performed on a single crystal of $U_2Pd_2In$ in the temperature range 0.3-300 K at pressures up to 1.8 GPa. The shape of the sample restricted the current to be applied along the [101] direction. The $\rho(T)$ curves are shown in Figure 6.11a for $p = 0.2$, 1.0 and 1.8 GPa. The shape of the curves does not change significantly with pressure. The value of $T_N$ decreases only slightly from 37.4(5) K at $p = 0.2$ GPa to 35.2(5) K at $p = 1.8$ GPa (Figure 6.11b).

![Figure 6.11](image.png)

**Figure 6.11** - (a) Temperature dependence of the resistivity of $U_2Pd_2In$ for different pressure values. (b) Pressure dependence of $T_N$. The line is a guide to the eye.

The small decrease of $T_N$ under pressure is consistent with the location of $U_2Pd_2In$ at the maximum in the Doniach phase diagram for the $U_2T_2In$ family of compounds (Section 4.1.2 - see also Figure 6.17). Upon pressure, the hybridization increases and thus also the exchange
interaction \( J \). For \( p \leq 1.8 \) GPa, the increase of the hybridization strength only shifts the position of the compound in the Doniach phase diagram around the relatively broad maximum of \( T_J \).

6.4. Magnetization studies of several \( \text{U}_2\text{T}_2\text{X} \) compounds

In order to partially confirm the general Doniach phase diagram for the \( \text{U}_2\text{T}_2\text{X} \) family of compounds presented in Section 4.1.2, the magnetization \( M_a \) and \( M_c \) where \( a \) and \( c \) denote the direction of the applied field, of several \( \text{U}_2\text{T}_2\text{X} \) single-crystals was measured for \( T \geq 2 \) K and \( B \leq 5.5 \) T. The measurements were performed after zero-field cooling (ZFC) and, in some cases, after field cooling (FC), using a SQUID magnetometer at the University of Lisbon.

6.4.1. \( \text{U}_2\text{Co}_2\text{Sn} \)

Above 25 K the magnetizations \( M_a \) and \( M_c \) of \( \text{U}_2\text{Co}_2\text{Sn} \) (Figure 6.12a) are linear functions of \( B \), which is typical of a paramagnetic state. At lower temperatures, \( M_c(B) \), and to a lesser extent \( M_a(B) \), depart significantly from linear behaviour. Over the entire temperature range and for \( B \leq 5.5 \) T, \( M_c > M_a \) which shows an important easy-axis anisotropy.

The \( M(T) \) curves do not show any sign of magnetic ordering (see Figure 6.12b for \( B \parallel c \) at 2 T). Above 20 K, the d.c. susceptibility curve, \( \chi_c(T) = M(T)/H \), follows a Curie-Weiss law with \( \theta = -24.5 \) K and a reduced effective moment \( \mu_{\text{eff}} = 1.65 \mu_B/\text{U} \), indicating the presence of hybridization effects. For all field directions, the low-temperature \( (T < 20 \) K) \( M/H \) values are higher than those given by the Curie-Weiss law extrapolated from the high-temperature range, as shown in Figure 6.12b for the \( c \) direction. The observed difference could indicate the presence of ferromagnetic spin fluctuations. However, the negative paramagnetic Curie-Weiss temperature \( \theta \) suggests antiferromagnetic correlations. The two contributions indicate the possibility of different magnetic interactions on the U and Co sublattices. It should be noted however, that the \( M(B) \) curves are not linear below 25 K. This means that the comparison of the \( M/H \) values with the Curie-Weiss behaviour should be taken with care.
In order to determine if the Co atoms carry a magnetic moment, the magnetization density distribution in the unit cell was measured at 2 K by means of polarized-neutron scattering experiments under a magnetic field of 5.5 T applied along the c-axis [17]. It was found that the major contribution to the magnetic susceptibility is located on the U atoms, $\mu_U = 0.118(3)\, \mu_B$, but a small response from the Co atoms was also present, $\mu_{Co} = 0.013(2)\, \mu_B$. At 2 K and 5.5 T, the induced magnetization along the c-axis is $0.220\, \mu_B$/f.u. to be compared with a value of $0.262(7)\, \mu_B$/f.u. determined by neutron scattering. The difference is likely due to a negative conduction-electron polarization.

### 6.4.2. U$_2$Ru$_2$Sn

Experiments carried out on polycrystalline samples have shown that U$_2$Ru$_2$Sn is a weak itinerant paramagnet [18]. This is confirmed by magnetization measurements on single crystals. A linear behaviour of the magnetization is observed as a function of applied fields up to 5.5 T (Figure 6.13a). $M(T)$ shows a weak temperature dependence (Figure 6.13b) and the
magnetization curves show no difference when measured after ZFC and FC. At room temperature, the susceptibility along the a-axis reaches a value of \(2.51 \times 10^{-8} \text{ m}^3/\text{mol}\) only. This value is further gradually reduced below 200 K to \(1.98 \times 10^{-8} \text{ m}^3/\text{mol}\) at \(T = 5\) K.

![Graph showing magnetization curves](image)

**Figure 6.13** - Magnetization of U$_2$Ru$_2$Sn for fields applied along the a-axis. (a) Field dependence at temperatures as indicated. (b) Temperature dependence at \(B = 1\) T.

Recently, resistivity data taken on polycrystalline samples were reported [19], which possibly reveal Kondo insulating behaviour. A sharp rise in the resistivity was observed at low temperatures \((T < 30\) K\), which could be an indication of semiconducting behaviour due to gap formation in the electronic density of states with an energy gap of the order of 1 K. So far, this has not been confirmed by the single-crystal magnetization data.

### 6.4.3. U$_2$Rh$_2$Sn

The ZFC magnetizations, \(M_a(T)\) and \(M_Q(T)\), of U$_2$Rh$_2$Sn are shown in Figure 6.14a for \(B = 0.05\) T. For both field directions a well defined peak in the susceptibility is observed, which signals an antiferromagnetic transition with a Néel temperature \(T_N = 28(2)\) K.
Over the whole temperature range studied $M_a < M_c$. This shows that the unique tetragonal axis is the easy axis for magnetization. The pronounced peak observed in $M_c(T)$ indicates that the magnetic moments are aligned along the c-axis. In a simple ideal antiferromagnet, the susceptibility measured for the field direction perpendicular to the magnetic moments should be almost constant below $T_N$. The small peak at $T_N$ observed in $M_c(T)$ is possibly due to a small misalignment of the crystal with respect to the magnetic field direction. In fact, if one considers a misalignment of only $5^\circ$ between the field direction and the a-axis, the magnetization becomes almost constant for temperatures up to $T_N$ and the peak is much reduced (Figure 6.14a).

The $M(B)$ curves (Figure 6.14b) show a linear behaviour for fields up to 5.5 T in both directions ($B \parallel c$ and $B \parallel a$). No hysteresis was observed for either field direction over the whole temperature range studied.

![Figure 6.14 - Magnetization of U$_2$Rh$_2$Sn. a) Temperature dependence for $B = 0.05$ T (the dotted line represents the corrected magnetization curve for $B \parallel a$ assuming a misalignment of $5^\circ$ - see text). b) Field dependence (open symbols: $B \parallel a$; solid symbols: $B \parallel c$).](image)

The susceptibility curves show that there is an important magnetic anisotropy even in the paramagnetic region. For $B \parallel c$, a Curie-Weiss behaviour is detected for temperatures above $T_N$ with $\theta = -105(2)$ K and an effective moment per U atom $\mu_{\text{eff}} = 2.80 \mu_B$/U. For $B \parallel a$, $\chi^{-1}(T)$ shows a positive curvature for the higher temperatures in the paramagnetic region, well described by a
modified Curie-Weiss law with $\Theta = -60(2)$ K. $\mu_{\text{eff}} = 1.34 \mu_B$ and $\chi_0 = 1.9 \times 10^{-8} \text{ m}^3/\text{mol} \cdot \text{K}$. This modified Curie-Weiss behaviour could be due to a small misalignment of the crystal with respect to the direction of the field, which leads to an additional contribution from the $c$-axis to the total magnetization measured. Therefore, the smaller value of $\mu_{\text{eff}}$ that was found for $B \parallel a$ should be treated with caution. Accurate determinations of $\mu_{\text{eff}}$ along both axes require a perfectly aligned crystal and an extension of the measurements to higher temperatures. It is likely that hybridization effects between the $f$-electrons and the ligands, as found in the UTX family of compounds [20], contribute significantly to this strong anisotropy in the paramagnetic region.

Antiferromagnetic order of the U moments was confirmed by neutron-scattering experiments on a single crystal [21]. The data reveal that $\text{U}_2\text{Rh}_2\text{Sn}$ orders in a collinear magnetic structure along the $c$-axis with $k = (0.0.1/2)$ (magnetic unit cell doubled in the $c$ direction compared to the nuclear cell). The refined value of the ordered magnetic moment at the U atom is $0.53(2) \mu_B$.

Interestingly, $\text{U}_2\text{Rh}_2\text{Sn}$ presents an exception to the empirical rule (obtained for the UTX compounds) that the $f$-moments should point perpendicularly to the shortest $f$-$f'$ bond distances, as in this compound the shortest interuranium distances are found to lie along the easy axis ($d_{yy} = 3.63 \text{ Å}$).

**6.4.4. $\text{U}_2\text{Ir}_2\text{Sn}$**

The magnetization of $\text{U}_2\text{Ir}_2\text{Sn}$ measured in fields applied along the $a$- and $c$-axis, shows the presence of a ferromagnetic phase below 50 K (Figure 6.15). Below this temperature, the $M/H$ values strongly deviate from a Curie-Weiss behaviour. The $M(H)$ curves indicate the saturation of a minority ferromagnetic phase at low fields, although the structural analysis of the single-crystals did not reveal the presence of an impurity phase. The impurity that is likely to be present in small quantities in the samples at e.g. the grain boundaries, may be $\text{Ulr}$, a ferromagnet with $T_C = 46$ K and $\mu_1 \approx 0.5 \mu_B$ [22]. Considering that the slope of the $M(H)$ curves at high fields is due to the $\text{U}_2\text{Ir}_2\text{Sn}$ matrix, the saturation magnetization of the ferromagnetic impurity can be extracted. At 5 K one obtains $\mu_{\text{Ulr}} \approx 0.003 \mu_B$. Comparing this value with the moment of pure $\text{Ulr}$ results in the presence of about 0.6% of Ulr impurity phase in the single-crystalline samples.
Probing $U_2Pt_2In$ in the Doniach phase diagram

The high temperature ($T > 50$ K) susceptibility follows a modified Curie-Weiss behaviour with a high Curie-Weiss temperature and a very much reduced effective moment: $\chi_0 = 1.4 \times 10^{-8}$ m$^3$/mol, $\theta = -106$ K and $\mu_{\text{eff}} = 1.0 \mu_B$/U for $B \parallel a$. A strong anisotropy is present in the system which persists up to 300 K, typical of systems with pronounced spin fluctuations. The $c$-axis is the easy direction of magnetization.

Due to the presence of the ferromagnetic impurity phase, a detailed study of the magnetization of $U_2Ir_2Sn$ was not carried out.

6.4.5. $U_2Ni_2In$

The magnetization of $U_2Ni_2In$ is linear in applied fields up to 5.5 T. The ZFC $M(T)$ curves for $U_2Ni_2In$ in a field of 0.05 T are shown in Figure 6.16. The measurements show a typical behaviour of an antiferromagnet with $T_N = 15.0(5)$ K. In contrast with what is expected for antiferromagnets with collinear structures, an extremely small magnetic anisotropy is observed in the ordered state, as evidenced by the similarity of $M_s(T)$ and $M_c(T)$. 

Figure 6.15 - Magnetization of $U_2Ir_2Sn$ (solid symbols: $B \parallel c$, open symbols: $B \parallel a$). a) Temperature dependence for 1 T applied along the $a$-axis. The line is a high-temperature Curie-Weiss fit. b) Field dependence at $T = 5$ and 40 K.
Neutron-diffraction experiments on a U$_2$Ni$_2$In single-crystal [23] indicate a non-collinear antiferromagnetic structure with the moments aligned along the [110] and [110] directions and a magnetic propagation vector $k = (0,0,1/2)$. The size of the ordered U moments amounts to 0.92(2) $\mu_B$ and no moments were found on the Ni sites, in contrast with what was reported in studies on polycrystalline samples [24].

Magnetization measurements were also performed with magnetic fields applied along the [110] direction. Also in this direction, $M(T)$ follows the same behaviour as $M_a$ and $M_c$. High-temperature fits of the susceptibility to a modified Curie-Weiss law yield the results shown in Table 6.1. The parameters obtained are similar in all field directions.

The reason for the similarity of the $M(T)$ curves for all studied field directions remains unclear.
6.5. Discussion

The measured resistivity of U$_2$Pt$_2$In under pressure is consistent with an increase of the control parameter $J$ upon pressure, bringing this compound into the Fermi-liquid regime. The pressure dependence of the temperature $T_{H}$, below which $\rho_a \sim T^2$, is also consistent with the location of U$_2$Pt$_2$In at or close to a QCP. A highly anisotropic resistivity component mainly present for $I || c$ is strongly enhanced upon pressure. A minimum in $\rho_x(T)$ develops as a consequence of this enhancement. The origin of this extra component remains unclear.

The effect of Th doping can be considered, in a first approach, as being equivalent to negative hydrostatic pressure. A comparison between the unit-cell contraction upon application of pressure and the unit-cell expansion upon Th doping, yields a value of -20.4 GPa/x. This implies that $\Delta V/V_0$ for $x = 0.03, 0.05, 0.08$ and 0.1 corresponds to - $\Delta V/V_0$ for $p = 0.6, 1.0, 1.6$ and 2.0 GPa, respectively.

Therefore, magnetic order could be expected to occur for the Th-doped compounds. The ordering temperature should increase with increasing Th content. Although the preliminary $\mu$SR experiments described are consistent with this hypothesis, the specific-heat results seem to contradict it. The presence of impurity phases in the samples unfortunately hampers the proper determination of the magnetic phase diagram of (U$_{1-x}$Th$_x$)$_2$Pt$_2$In. Therefore, the results regarding Th doping presented in this chapter, should be considered as preliminary and taken with caution.

Assuming that the atomic coordinates do not change with pressure, the isotropic compressibility indicates that the primary effect of pressure is a uniform reduction of the interatomic distances. Therefore, the hybridization matrices can be calculated for U$_2$Pt$_2$In under pressure. The total hybridization $V_{ij}$ increases about 2.3% from $p=0$ to $p=1.8$ GPa. The
temperature $T_F$ below which a $\rho \sim T^2$ behaviour is observed is plotted in Figure 6.17b as a function of $V_{dd}^2$, where $V_{dd}^2 \propto J$.

The antiferromagnetic ordering temperature of $U_2Pd_3In$ decreases slightly with pressure, as evidenced by the resistivity measurements. Taking for $U_2Pd_3In$ the same compressibility value as for $U_2Pt_3In$, the hybridization can be calculated in the same way as before. From $p=0$ to $p=1.8$ GPa, $V_{dd}$ increases 2.4%. The values of $T_S$ are also shown in Figure 6.17b.

![Figure 6.17](image)

**Figure 6.17** - (a) Doniach phase diagram for the $U_2T_2X$ ($X=In, Sn$) family of compounds. (b) Possible phase diagram around $U_2Pt_3In$. The lines are guides to the eye.

Recently, magnetization, resistivity and neutron-diffraction studies on the system $U_2(Pt_{1-x}Ni_x)_3In$ have been reported [25]. No magnetic order was found down to 1.7 K for the polycrystalline samples with $x \leq 0.4$. For $x \geq 0.5$, antiferromagnetism is present with $T_S$ increasing from $\sim 11$ K for $x = 0.5$ to $\sim 14$ K for $x = 1$. The evolution of $T_S$ upon Ni doping is not consistent with the Doniach phase diagram for the $U_2T_2In$ compounds (Figure 6.17a). However, there is a strong suppression of the heavy-fermion character of $U_2Pt_3In$ upon Ni doping which might reflect a strong modification of the density of states at the Fermi surface [25].

The tentative phase diagram presented in Figure 6.17b is consistent with the location of $U_2Pt_3In$ at or close to a quantum critical point (QCP). Whether the non-Fermi liquid behaviour is
Probing $U_2$Pt$_2$In in the Doniach phase diagram

present only at the QCP or for a finite region of $J$ values around the QCP is a question that can only be answered by a more thorough study of the phase diagram. This could be done by investigating single-phase samples of $(U_{1-x}Th_x)_2$Pt$_2$In or preferably $U_2(Pt_{1-x}Pd_x)_2$In.

References


