Pressure tuning of the non-Fermi liquid state in UCo$_{0.95}$Fe$_{0.05}$Al

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1. Introduction

The deviation of the low-temperature physical properties such as specific heat, electrical resistivity or magnetic susceptibility from the predictions of the Fermi-liquid (FL) theory has been reported already decades ago and observed in numerous materials (e.g. [1]). Particularly interesting is the universal non-FL (NFL) behavior occurring around the onset of magnetic order. Experimental findings amounted into a concept of quantum criticality, in which dynamical degrees of freedom related to quantum fluctuations (depending on the type of coupling and dimensionality) coexist with classical degrees of freedom if the magnetic phase transition approaches $T = 0$ under the influence of some control parameter (composition, pressure…) [2]. Such a quantum critical point (QCP) is then followed on the non-magnetic side by a “heavy” FL regime spreading gradually from the $T = 0$ limit, in which spin fluctuations are responsible for a strong renormalization of basic parameters of the electronic system (as enhancement of effective mass), similarly to the critical fluctuations at the temperature-driven phase transitions. As the NFL behavior can occur also due to disorder in different types of system (see [3, 4] for overview), it is essential to investigate materials without any intrinsic disorder. Application of external pressure on an undoped system is, therefore, a unique tool for tuning magnetic materials through a genuine quantum critical regime.

Extended investigation of U ternary compounds in the vicinity of the onset of magnetic transition led to the recognition that the band metamagnet UCoAl exhibits a NFL scaling of electrical resistivity $\rho =$
\( \rho_0 + bT^{6/3} \) (predicted by theories for ferromagnetic 3D spin fluctuators) in the low-field non-magnetic state [5-7], whereas the field-induced ferromagnetic state (first-order transition, critical field is 0.65 T in the low-\( T \) limit) displays a standard FL behavior.

Proceeding towards the non-magnetic side by applying pressure (which broadens the 5f band and generally suppresses the 5f magnetic moments), the NFL behavior vanishes only above 5 GPa [8], where the tendency to metamagnetism disappears. These findings indicate that some sort of a robust NFL phase exists in UCoAl, while the scenario of a single critical point is untenable in this case.

The proximity of UCoAl to the border of stable ferromagnetism is illustrated by the sensitivity to doping. Few percent doping by \( \text{e.g.} \) Fe can induce ferromagnetism stable even without the magnetic field [9, 10]. This property can be used for covering all the regimes of the phase diagram shown in Figure 1, since undoped UCoAl itself can be driven by the pressure only through the regions (2) and (3). The doping with iron makes the region (1) also accessible. Therefore, we have undertaken the high-pressure experiment on \( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) aiming at covering the whole pressure region of the QCP phase diagram.

\( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) has the same crystal structure as UCoAl, it orders ferromagnetically below 30 K, and its magnetic ordering is pressure sensitive [9, 10]. The latter is related to the fact that hydrostatic pressure induces predominantly compression in the basal plane of the hexagonal structure, and the U-U spacing within the basal plane is the crucial parameter controlling magnetic properties [11]. Also, the uncertainty of whether the NFL regime in \( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) would be due to QCP or to the disorder, is not the issue for \( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) because the role of the quantum critical regime was shown for the Fe-free UCoAl.

### 2. Experimental details

Electrical resistivity was studied on different pieces of a single crystal with RRR = 4 prepared in a tetra-arc furnace from pure metals of at least 3N purity.

High-pressure measurements have been performed at ITU Karlsruhe in zero magnetic field using a four-probe DC technique with the sample and a stripe of lead (manometer) pressed in a piston-cylinder device, with steatite as a pressure-transmitting medium. Typical sample thickness, which can be used in this cell, is 30-40 \( \mu \)m. Extreme brittleness of the \( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) crystal imposed the limitations on the size of the sample (up to 500 \( \mu \)m) and the maximal pressure applied (up to \( P = 3.6 \) GPa).

### 3. Results and discussion

The resistivity data obtained are presented in Figure 2. The measurements were performed with the electrical current along the \( \alpha \)-axis. In the experimentally accessible pressure range, the sample compression has primarily affected the low-temperature part of the \( \rho(T) \) curves, and, therefore, its details are shown in the figure.

Firstly, the increase of pressure leads to the disappearance of the ferromagnetic ordering in \( \text{UCo}_{0.95}\text{Fe}_{0.05}\text{Al} \) below 2 GPa. The magnetic phase transition can be well observed for the zero-pressure data. It is indicated by the kink on the resistivity curve at \( T_C = 29 \) K, in the good agreement with \( T_C \) determined from the measurements of the specific heat reported in literature [9, 10], which is followed by a broader feature at \( T = 32 \) K. As the pressure increases, the anomalies smear out. Already in \( P = 1.0 \) GPa the kink disappears and the broad feature transforms into a change of slope at \( T = 34 \) K.
The rapid smearing of the sharp anomaly could be partly related to a certain non-hydrostaticity in the present experimental setup. For the next data set, collected at $P = 1.8$ GPa, even the change of slope is hardly visible and its position, $T = 35$ K, can be determined only by the temperature derivative of $\rho(T)$. At $P = 2.3$ GPa and above, there are no clear traces of any resistivity anomalies below 50 K. Thus, the resistivity data do not provide the exact value of the critical pressure for the disappearance of the ferromagnetism in UCo$_{0.95}$Fe$_{0.05}$Al, but the 2 GPa limit seems quite sound because the character of the resistivity changes qualitatively between 1.8 GPa and 2.3 GPa. Another argument in favor of this assumption can be found in the high-pressure magnetization studies reported in Ref. [9]. These measurements indicate the rapid decrease of $T_C$ with pressure at the rate of about 15 K/GPa. The maximum pressure achieved in the study [9] was $P = 1.2$ GPa, so the complete suppression of the magnetic ordering was not observed, yet it is quite likely to occur even below the expected $P_c = 2$ GPa because the pressure dependence of the Curie temperature usually becomes non-linear as it approaches $T = 0$.

The second interesting subject is the pressure-related modification of the polynomial $\rho = \rho_0 + AT^n$, which describes the low-$T$ part of $\rho(T)$ in UCo$_{0.95}$Fe$_{0.05}$Al (Figure 3). The Figure 3 shows both the experimental data and the $T^2$ fits corresponding to the FL behavior. The fits are straight lines due to the $T^2$ abscissa scale. The region, across which the experimental data follow the fits, is the temperature range of the stability of FL state in UCo$_{0.95}$Fe$_{0.05}$Al. At $P = 0$ (in the ordered state) it extends up to approximately 16 K, but it shrinks rapidly with increasing pressure. Starting from $P = 2.7$ GPa, the $\rho(T)$ curve cannot be fitted to the $T^2$-dependence across the temperature range of more than 5 K (Figure 3), and, therefore, the FL regime at these pressures should not be considered any more.

A more detailed analysis reveals that the increase of pressure induces a non-Fermi liquid type of resistivity above the $T^2$-region, which shrinks progressively towards zero $T$. The exponent $n$ in the expression $\rho = \rho_0 + AT^n$ appears to be pressure dependent (see Figure 4), and the range of NFL resistivity extends up to $T = 15$ -17 K.

The observation of the two regimes, FL and NFL following each other, in UCo$_{0.95}$Fe$_{0.05}$Al places the compound on the left-hand side of the QCP diagram (Figure 1), where the FL state still survives at
the lowest temperatures due to the V shape, but the influence of the spin fluctuations determines the resistivity above that temperature range. The question remains open whether the FL state survives at the lowest temperatures in the pressures exceeding 2 GPa. For unambiguous conclusion one would need to cover temperature range extending to the mK range. In any case, we do not observe the re-emergence of the FL state on the right side of the phase diagram in Figure 1. This goes in line with the experimental findings for UCoAl where, in spite of the non-magnetic ground state, the pressure above 5 GPa is required for complete suppression of the NFL regime [8]. It is natural to assume that in the ferromagnet UCo$_{0.95}$Fe$_{0.05}$Al this value would be even higher, and the pressures of 10 - 20 GPa are necessary in order to cover the whole QCP phase diagram. The maximal pressure $P = 3.6$ GPa reached in the present work is sufficient only to drive UCo$_{0.95}$Fe$_{0.05}$Al from the region (1) of the magnetically ordered FL, into the region (2) close to the QCP (Figure 1). Most likely $P = 3.6$ GPa is sufficient complete suppression of the FL regime in UCo$_{0.95}$Fe$_{0.05}$Al.

4. Concluding remarks
The ferromagnetic order in UCo$_{0.95}$Fe$_{0.05}$Al is suppressed above $P = 2$ GPa. The range of the Fermi liquid regime in this compound, which is associated with the ordered state, shrinks with the increasing pressure, but is observed till at least to $P = 2.3$ GPa. The non-Fermi liquid resistivity emerges atop of the FL regime, and the exponent $n$ in the expression for $\rho(T)$ is the function of pressure. The robust character of the FL-NFL states in UCo$_{0.95}$Fe$_{0.05}$Al is similar to the properties of UCoAl. The NFL behavior can be attributed to the critical state with quantum fluctuations present between the phase with well defined ordered magnetic moments and the state in which the moments completely disappear. Unlike the two neighbor phases, in which electronic system exhibits FL properties, the FL scaling breaks down in the NFL regime, and basic properties as resistivity follow different scaling, depending on the type of magnetic coupling and dimensionality of the system. The combined high-pressure experiments on pure and Fe-doped UCoAl illustrate how the NFL state develops from the magnetic FL state and gives up again to the non-magnetic FL state. The novelty of the present data dwells in the fact that the NFL behavior exists over an extended pressure/concentration range, which contradicts a common belief that it should exist only in a close vicinity of a single critical point.

Acknowledgements
AVK acknowledges the European Commission for support in the frame of the “Training and Mobility of Researchers” program. This work is a part of the research plan MSM 0021620834 that is financed by the Ministry of Education of the Czech republic. Financial support of the Czech Science Foundation under the Grant no. 202/06/0178 also has been enjoyed. The work was also supported by the project D/B ‘Promin’.

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