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Quenching of ferromagnetism in β–UB$_2$C and UNiSi$_2$ at high pressure.

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Abstract. β-UB$_2$C exhibits itinerant ferromagnetism below $T_c = 75$ K; whereas, UNiSi$_2$ exhibits ferromagnetism of localized uranium moments below $T_c = 95$ K and Kondo lattice behaviour at higher temperatures. We have found that ferromagnetism in both compounds is quenched at high pressure. In β-UB$_2$C the Curie temperature continuously approaches zero at 2.95 GPa, where resistivity and specific heat reveal the behaviour similar to that observed in Ce-based antiferromagnets near quantum critical points. In UNiSi$_2$ the Curie temperature decreases gradually to 25 K at 5.34 GPa. Above this pressure hysteretic phenomena appear in the temperature dependences of resistivity between 5 and 17 K, signalling a change in the magnetic order. No signature of magnetism was found above 5.5 GPa down to 1.15 K. Kondo lattice behaviour of resistivity with the enhanced residual resistivity is observed in UNiSi$_2$ above 5.5 GPa.

1. Introduction
Strongly correlated f-electron systems exhibit a variety of interesting properties at low temperatures. Magnetic order, which is the ground state of many lanthanides and actinides, can be destroyed simply by raising temperature, but quenching of magnetism also may be accomplished at zero temperature by adjusting a non-thermal control parameter. For example, pressure can suppress a magnetic ordering temperature to zero and produce a change of the ground state at a quantum-critical point (QCP), where magnetic order is destroyed by quantum and not thermal fluctuations. Ferromagnetic QCP’s are rare because ferromagnetic order is relatively uncommon for cerium and uranium compounds. Nevertheless, superconductivity has been found near zero-temperature ferromagnetic instabilities in UGe$_2$ [1] and in UR$_2$ [2] under high pressure. Recently, it was suggested [3] that the itinerant ferromagnet β-UB$_2$C, by analogy with UGe$_2$ and UR$_2$, might exhibit a pressure-induced QCP and superconductivity. UNiSi$_2$ on the other hand is a nearly localized ferromagnet [4] and the trends in UFeSi$_2$-UCoSi$_2$-UNiSi$_2$ series allow one to expect the suppression of magnetism in this compound at high pressure. The aim of our experiments has been to establish the evolution of transport and
thermodynamic properties of these two uranium compounds in a wide range of pressures and temperatures and to clarify the effect of pressure on magnetism.

2. Experimental details

We produced $\beta$-UB$_2$C by argon arc melting a stoichiometric mixture of constituents on a water-cooled copper hearth in a Zr-gettered high-purity argon atmosphere, a method similar to that used by Tran et al. [3]. Powder x-ray diffraction of the product revealed the main phase to be rhombohedral $\beta$-UB$_2$C ($a = 0.6537(2)$ nm, $c = 1.0784(2)$ nm) which is formed during rapid cooling of the arc-melted boule. Orthorhombic UBC also was found as an $\sim$10% impurity phase in the powder pattern. Single crystals of UNiSi$_2$ with orthorhombic CeNiSi$_2$-type structure were grown in gallium flux. The lattice parameters ($a = 0.4016(3)$ nm, $b = 1.6014(1)$ nm, $c = 0.4016(3)$ nm) are close to those reported for polycrystalline samples [4]. Magnetic properties and the specific heat of both materials were characterized with Quantum Design MPMS and PPMS instruments at atmospheric pressure. Electrical resistivity, ac-susceptibility and ac-specific heat measurements at high hydrostatic pressure to 6 GPa were performed with a clamped toroidal anvil device. Samples and Pb pressure sensor were placed in a Teflon ampoule 2 mm in diameter and height filled with a glycerol-water (3:2) liquid.

3. Results and discussion

3.1. $\beta$-UB$_2$C

As shown in the left panel of Figure 1, a kink in $\rho(T)$ at ambient pressure signals the establishment of magnetic order at the Curie temperature $T_c \approx 75$ K. For comparison, the temperature-dependent resistivity of single-crystalline UBC at ambient pressure also is plotted. Pressures to 6 GPa do not produce substantial changes in the overall shape or magnitude of $\rho(T)$ of $\beta$-UB$_2$C above 75 K; however, the resistive anomaly at $T_c(P)$ shifts to lower temperatures and broadens with increasing pressure. Differentiation of $\rho(T)$ reveals a hump at a characteristic temperature $T^*$ in addition to a sharp increase and a peak around $T_c$. Representative curves of $\frac{d\rho(T)}{dT}$ are depicted in the right panel of Figure 1. At ambient pressure, the hump, located at $T^* \approx 40$ K, reflects excitations of the ordered state [3]. Application of pressure also shifts $T^*$ to lower temperatures. It is still visible at 3.6 K and 2.78 GPa, but both $T^*(P)$ and $T_c(P)$ tend to zero at a critical pressure $P_c$, just below 3 GPa. At higher pressures no signature of magnetic ordering is found in $\rho(T)$, and there is only a broad maximum in the $\frac{d\rho(T)}{dT}$ at $T_{\text{max}}$, which increases gradually between 3 and 6 GPa and is distinct from $T^*$.

![Figure 1. Temperature dependences of resistivity $\rho(T)$ of $\beta$-UB$_2$C and $\frac{d\rho(T)}{dT}$ at some representative pressures.](image)

From fits of the low-temperature resistivity to $\rho(T) = \rho_0 + AT^n$, we extract the pressure dependence of $\rho_0$, $A$, and $n$, which is displayed in the left panel of Figure 2. The coefficient $A$ exhibits a pronounced peak at $P_c$. The exponent $n$ is $\sim 2.5$ in the magnetically ordered state at low and modest
pressures, above which it decreases sharply to $n \approx 1.2$ near $P_c$. For $P > P_c$, $n$ gradually approaches the Fermi-liquid value $n = 2$ at the highest pressures. The residual resistivity $\rho_0$, which already is large at ambient pressure, increases by $\approx 20 \, \mu\Omega\cdot\text{cm}$ as $P$ approaches $P_c$ from below and then decreases by over 40 $\mu\Omega\cdot\text{cm}$ for pressures well above $P_c$.

In ac-calorimetry measurements, a peak in specific heat due to magnetic order is well visible below 2 GPa (diamonds in the P-T diagram). A broad anomaly is still visible at 2.29 GPa, but close to $P_c$ a calorimetric signature of $T_c$ vanishes. The value of $C(P)/T$ at 1.12 K exhibits a peak at $P_c$ (Figure 2).

All these measurements suggest a ferromagnetic QCP at $P_c \approx 3$ GPa.

3.2. UNiSi$_2$

On cooling the electrical resistivity $\rho(T)$ of UNiSi$_2$ increases in a Kondo-like ($-\ln T$) manner, as was found earlier for polycrystalline samples [4], passes through a maximum and drops at the ferromagnetic ordering temperature $T_c$. At high pressure $T_c$ decreases and a qualitative change in $\rho(T)$ takes place between 5 and 5.6 GPa (Figure 3, left panel).

$n$ was found to be $2 \pm 0.05$ at all pressures. $C/T$ from ac-calorimetry ($T=0$ extrapolation) also is shown in the upper right panel. Open circles – measurements made on downloading.
In this pressure range the peak of magnetic ac-susceptibility accompanying the transition to the ferromagnetic state decreases dramatically and vanishes. The specific heat C(T) anomaly observed at T_c decreases and disappears above 5.1 GPa (Figure 4, left panel). All these experiments show that ferromagnetism of nearly localized U f-electrons is quenched above 5.5 GPa. In the pressure region around this critical pressure the appearance of another – weak ferromagnetism – was found. It is visible in an upturn in ac-susceptibility below ~15 K where a very weak peak of susceptibility due to the main transition is still present at 26 K. Signatures of this weak ferromagnetism also are visible in ρ(T) and C(T) behavior in the region centered near 5.2 GPa at the P-T diagram (Figure 4, right panel).

Uranium compounds are of special interest due to a dual nature of their f-electrons: localized and itinerant. For ferromagnetism of localized electrons the magnetic entropy is equal to Rln(2J+1), where J is the total moment. In the case of fully localized U^{3+} moments one should expect magnetic entropy of the transition ~Rln10 = 19.1 J/mole-K. For UNiSi_2 the magnetic entropy is 11.3 J/mole-K as estimated from Figure 4. It means that f-electrons in this material are mostly localized as concluded earlier [4]. On applying pressure the magnetic entropy is strongly reduced and vanishes above 5.5 GPa. At this pressure the ground state of UNiSi_2 changes to nonmagnetic with enhanced values of γ_0 and ρ_0 (Figure 3). We suggest the delocalization of f-electrons of UNiSi_2 at a critical pressure 5.5 GPa with the formation of a hybridized f-d band of heavy electrons. The magnetic entropy of localized U moments is now transferred to these heavy charge carriers. The enhanced value of ρ_0 may be explained by a combined effect of lower mobility of carriers in this band and strong inter-band scattering. We stress that the high value of ρ_0 in the high-pressure state of UNiSi_2 is not due to formation of defects in the lattice. Upon releasing pressure, the sample recovers its ambient pressure RRR within a few percent of the initial value. In contrast to β-UB_2C no non-Fermi-Liquid behaviour of resistivity was found around the critical pressure in UNiSi_2 and we can not regard this localized-to-itinerant transition as a QCP.

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References
[1] Saxena S S et al. 2004 Nature 406 587
[2] Akazawa T et al. 2004 J.Phys. Soc. Japan 73 3129
[3] Tran V H, Rogl P, André G, Bourée F 2006 J. Phys.: Condens. Matter 18 703
[4] Kaczorowski D 1996 Solid State Commun. 99 949

Figure 4. Ac-specific heat of UNiSi_2 at high pressures and the specific heat of UNiSi_2 and ThNiSi_2 measured at P = 0 in PPMS (left panel). Right panel shows the P-T diagram of UNiSi_2.