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Sidorov, VA
Tobash, PH
Wang, C
et al.

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Quenching of ferromagnetism in β–UB₂C and UNiSi₂ at high pressure.

V A Sidorov[1, 2], P H Tobash[1], C Wang[1], B L Scott[1], Tuson Park[1, 3], E D Bauer[1], F Ronning[1], J D Thompson[1] and Z Fisk[4]

[1] Los Alamos National Laboratory, Los Alamos, New Mexico, 87545, USA

[2] Institute for High Pressure Physics, Russian Academy of Sciences, 142190 Troitsk, Russia

[3] Department of Physics, Sungkyunkwan University, Suwon 440-746, Korea

[4] Department of Physics and Astronomy, University of California, Irvine, California, 92697, USA

E-mail: vs_hppi@mail.ru

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 UIr [2] under high pressure. Recently, it was suggested [3] that the itinerant ferromagnet β-UB$_2$C, by analogy with UGe$_2$ and UIr, 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 β-UB_2C 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 β-UB_2C (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 ~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. β-UB_2C

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 ~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 β-UB_2C 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 $d\rho(T)/dT$ are depicted in the right panel of Figure 1. At ambient pressure, the hump, located at T* ≈ 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 $d\rho(T)/dT$ at T_{max}, which increases gradually between 3 and 6 GPa and is distinct from T*.
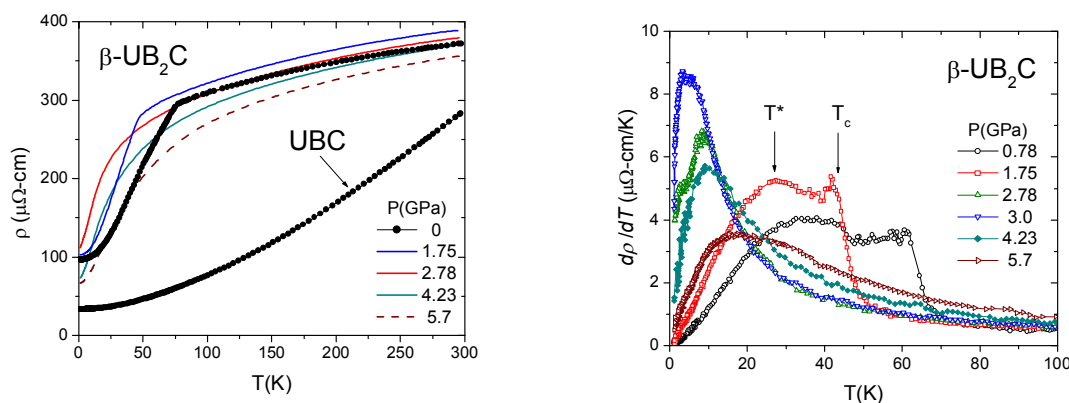

Figure 1. Temperature dependences of resistivity $\rho(T)$ of β-UB_2C and $d\rho(T)/dT$ at some representative pressures.

From fits of the low-temperature resistivity to $\rho(T) = \rho_0 + AT^n$, we extract the pressure dependence of ρ_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 ~2.5 in the magnetically ordered state at low and modest

pressures, above which it decreases sharply to $n \sim 1.2$ near P_c. For $P > P_c$, n gradually approaches the Fermi-liquid value $n = 2$ at the highest pressures. The residual resistivity ρ_0, which already is large at ambient pressure, increases by ~ 20 $\mu\Omega$-cm as P approaches P_c from below and then decreases by over 40 $\mu\Omega$-cm for pressures well above P_c.
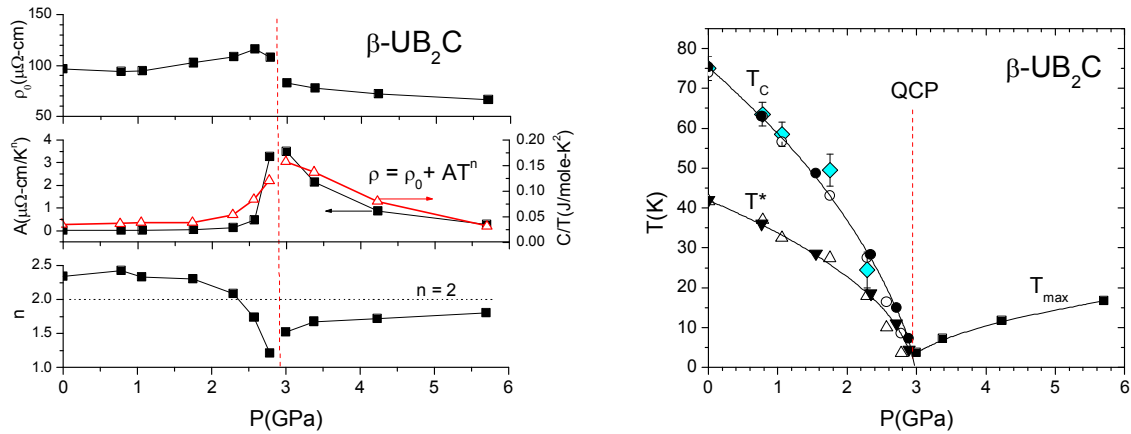

Figure 2. Pressure dependences of ρ_0, A, and n extracted from fits of low temperature resistivity and pressure dependence of C/T at 1.12 K (left panel). P-T diagram of β-UB$_2$C (right panel) .

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). The absolute value of C/T was normalized to the ambient pressure value 0.035 J/mole-K^2 determined in a PPMS. All these measurements suggest a ferromagnetic QCP at $P_c \sim 3$ 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 (Figure3, left panel).
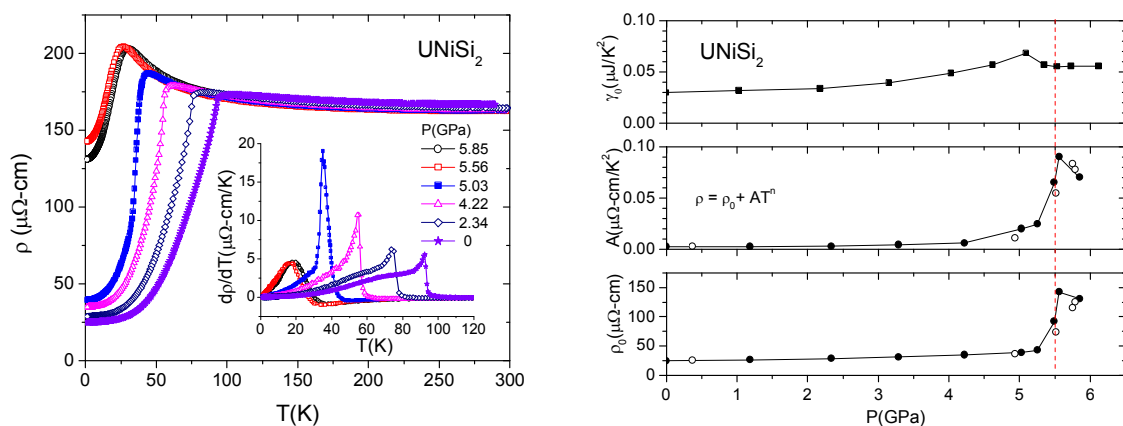

Figure 3. Temperature dependences of $\rho(T)$ at different pressures (left panel) and pressure dependences of ρ_0 and A extracted from fits of low temperature resistivity in the form $\rho(T) = \rho_0 + AT^n$ (right panel). n was found to be 2 ± 0.05 at all pressures. C/T from ac-calorimetry (T=0 extrapolation) 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).
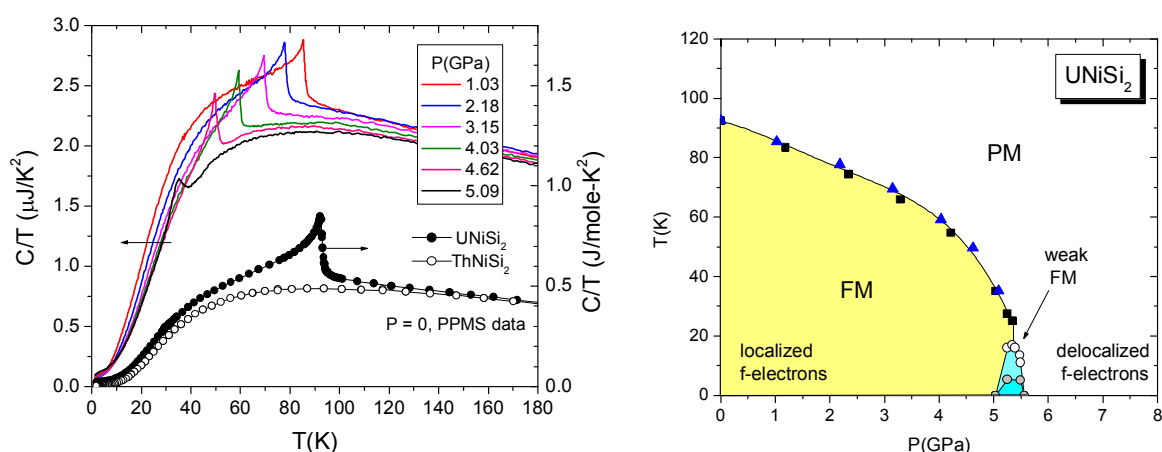

Figure 4. Ac-specific heat of $UNiSi_2$ at high pressures and the specific heat of $UNiSi_2$ and $ThNiSi_2$ measured at P = 0 in PPMS (left panel). Right panel shows the P-T diagram of $UNiSi_2$.

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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