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Magnetic ordering in UCu_2Si_2 at high pressure

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Abstract

We have performed resistivity measurements as a function of applied pressure to 2.0 GPa on a single crystal of UCu_2Si_2 . We find that the ambient pressure magnetic ordering temperature of 100 K, and the low-temperature ordering at 50 K, both decrease gradually in a manner consistent with itinerant magnetism. The results will be compared to previous measurements on UCu_2Ge_2 .

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Certain magnetic correlated-electron systems are known to superconduct under pressure at a quantum critical point (QCP) where the magnetic ordering temperature (T_c for ferromagnets or T_N for antiferromagnets) goes to 0 K. The way in which a system approaches this QCP, namely the shape of the magnetic ordering temperature as a function of pressure, is due to a competition between magnetic order and a non-magnetic Kondo singlet ground state. This characteristic shape is called the Doniach necklace where an asymmetric magnetic ordering temperature versus pressure is predicted [1]. Initially, pressure enhances the magnetic ordering temperature by increasing the RKKY interactions. Eventually, however, the more drastic dependence of the destabilizing Kondo effect forces the magnetic ordering temperature to go through a maximum and drop rapidly. As the system reaches the QCP, superconductivity is sometimes observed. While the Doniach behavior is observed in most magnetically ordered Ce and Yb compounds [2], it is often seen to not hold for U compounds. The more extended 5f wave functions in U compounds relative to 4f in Ce and Yb compounds can be seen in pressure-dependent magnetic ordering studies on the uranium chalcogenides as very

different behavior is found for localized (Doniach) and itinerant (band magnetism like Fe) electrons [3]. We have been searching for U compounds that approach a QCP with localized 5f electrons in the hope of observing pressure induced superconductivity similar to that seen in Ce and Yb compounds. To date, ALL of the U compounds that display a QCP as a function of pressure (UGe_2 [4], UPtGa_5 [5], UN [5], and UGa_3 [6], to name a few) do so in a manner consistent with itinerant magnetism. While UCu_2Ge_2 shows a behavior similar to the Doniach phase diagram, the value of T_c could not be determined above 10 GPa [7], and the volume of isostructural UCu_2Si_2 is very close to that of UCu_2Ge_2 at 10 GPa.

Single crystals of UCu_2Si_2 grown by the self-flux technique were used for the experiments. The tetragonal lattice constants a and c were measured by X-ray diffraction (XRD) as a function of pressure to 10 GPa at HP-CAT beamline ID-B at the Advanced Photon Source. High pressure four probe electrical resistivity measurements were performed at pressures up to 2.0 GPa in a piston-cylinder cell made of a Ni–Cr–Al alloy chamber inside a Cu–Be cylinder. A 1:1 mixture of Flourinert FC70 and FC77 was used as pressure transmitting medium. Ruby fluorescence was used to determine the pressure in all pressure experiments.

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The high-pressure XRD measurements on UCu_2Si_2 have found that the ThCr_2Si_2 tetragonal structure is stable up to around 10 GPa. This means that no phase transitions are observed in the pressure range of the electrical transport measurements we report below. Electrical resistance data normalized to the room temperature value are shown in Fig. 1 at 0.6 GPa. As temperature is lowered from room temperature, the resistance initially increases and goes through a broad maximum around 150 K. This behavior is seen in numerous correlated-electron systems and is usually attributed to the Kondo effect. As observed before [8], two magnetic transitions are seen as anomalies in the resistivity data in UCu_2Si_2 . The location of the magnetic transitions was determined by measuring shifts in the derivative of the electrical resistance versus temperature plots. These anomalies are observed due to changes in magnetic scattering of the conduction electrons when magnetic ordering occurs. At ambient pressure, the first (T_c) at around 105 K is due to ferromagnetic order; the second (T_N) at around 50 K is due to antiferromagnetic order. It should be noted that there has been some controversy about the 50 K transition (see comments to Ref. [8] in Ref. [9] and the response in Ref. [10]) [8–10].

Both transitions were found to persist up to the highest pressure of the current study, 2.0 GPa. The changes in the transition temperatures are shown in Fig. 2. Both transitions decrease gradually as a function of pressure at the rate of approximately -0.9 K/GPa for T_c ($= 105 \text{ K}$ at 0 GPa) and -0.5 K/GPa for T_N ($= 50 \text{ K}$ at 0 GPa). This decrease as a function of pressure is contrary to what was expected from previous high-pressure results on UCu_2Ge_2 as discussed above. However, the values are consistent with what is expected for 5f itinerant magnetism [3]. These results show that UCu_2Si_2 is an

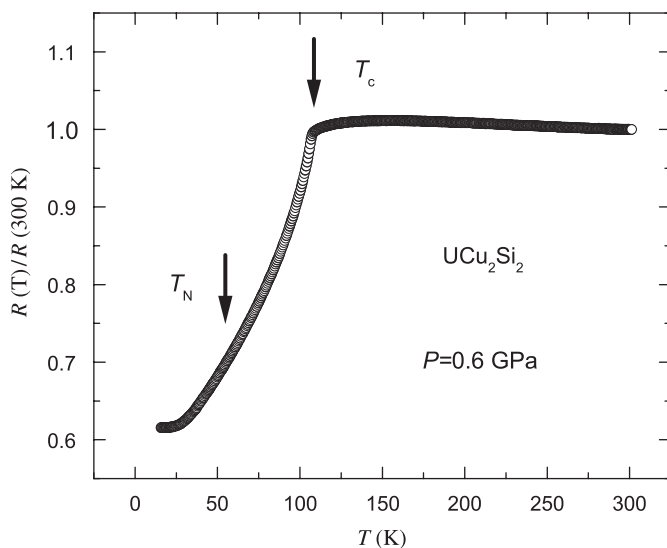


Fig. 1. Temperature-dependent electrical resistance normalized to the room temperature value at 0.6 GPa for UCu_2Si_2 . The location of two magnetic transitions, T_c and T_N , are shown.

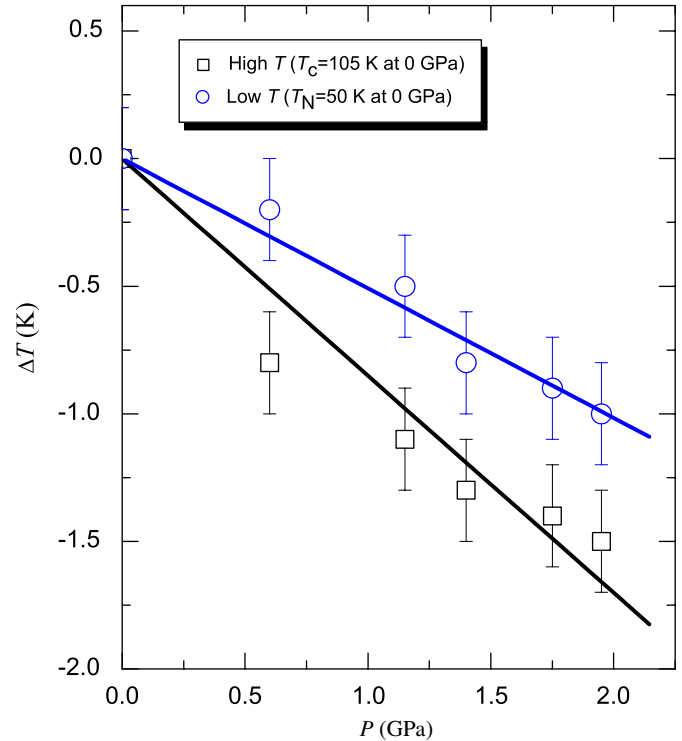


Fig. 2. Pressure induced changes in the two magnetic transitions in UCu_2Si_2 .

itinerant 5f system and significant pressure will be needed to reach a QCP.

This leads to the following question: Why did UCu_2Ge_2 behave in a manner consistent with the Doniach phase diagram? One hint may come from the fact that the magnetic signal in UCu_2Ge_2 rapidly vanishes above 10 GPa when the ordering temperature drops. Originally, this was taken as an evidence for Doniach-like behavior. However, another explanation might lie in a pressure induced phase transition in UCu_2Ge_2 . Recently, isostructural UMn_2Ge_2 was found to undergo a transition from the tetragonal structure to a complicated structure around 12 GPa [11]. High-pressure XRD on UCu_2Ge_2 as well as more detailed measurements on UCu_2Si_2 are planned.

In summary, we have performed high-pressure electrical transport measurements on single crystals of UCu_2Si_2 up to 2.5 GPa. The tetragonal ThCr_2Si_2 crystal structure is stable up to at least 10 GPa. Both magnetic transitions decrease gradually with pressure consistent with the magnetic order from the 5f electrons being itinerant in UCu_2Si_2 . Along with previous results, there might be a pressure induced phase transition in UCu_2Ge_2 around 10–12 GPa.

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