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RESPONSE OF URANIUM-BASED HEAVY-FERMION MAGNETS TO HYDROSTATIC PRESSURE

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We have studied the response of heavy-fermion magnets UCd_{11}, U_2Zn_{17} and $UAgCu_4$ to hydrostatic pressures exceeding 17 kbar through temperature-dependent electrical resistance measurements. Results for U_2Zn_{17} and $UAgCu_4$ can be understood in terms of competing Kondo and RKKY interactions. UCd_{11} exhibits two new phase transitions induced by pressure.

Four uranium-based compounds UCd_{11} [1], U_2Zn_{17} [2], $UAgCu_4$ [3] and UCu_5 [4] are known that exhibit features consistent with magnetic ordering arising out of a strongly correlated f-electron system. Specific heat measurements on these materials reveal electronic contributions (extrapolated to $T = 0$ from above the ordering temperature) ranging from over 250 to ≈ 840 mJ/(mol f K^2). Assuming a Fermi liquid description, we infer from the γ values effective masses $m^* \gg m_e$, where m_e is the free electron mass, hence the label heavy-fermion magnets. Certain features are common to all four: high temperature Curie-Weiss susceptibilities with negative paramagnetic Curie temperatures; entropy associated with the magnetic transition considerably less than $R \ln 2$; and large ($\gtrsim 100$ $\mu\Omega$ cm) resistivities typical of Kondo-like scattering. Of the four U_2Zn_{17} and UCu_5 have been studied by neutron scattering. U_2Zn_{17} was found [5] to be a commensurate antiferromagnet with an anamolously small moment below T_N; while uranium moments in UCu_5 coupled ferromagnetically within (111) planes but parallel planes coupled antiferromagnetically [6]. In view of their interesting behavior at ambient pressure and the well-known correlation between actinide–actinide separation (d_{U-U}) and magnetism, we have studied the response of UCd_{11}, U_2Zn_{17} and $UAgCu_4$ to hydrostatic pressures exceeding 17 kbar.

Four lead ac resistance measurements were made in the temperature interval 1 to 290 K on single-crystal UCd_{11} and U_2Zn_{17} and polycrystalline $UAgCu_4$. All samples were single phase within the resolution of X-ray diffraction techniques. Details of the sample preparation and characterization have been given elsewhere [1–3]. Preliminary X-ray intensity analysis on $UAgCu_4$ suggests that the Ag atoms reside on specific Cu sites in the UCu_5 crystal structure. Pressures were produced in a Be–Cu self-clamping cell whose design and operation have been described thoroughly [7].

The resistive response to pressure for the three heavy-fermion magnets is summarized in fig. 1. Consistent with progressively larger electronic specific heats (see table 1), we observe a systematic increase in changes in the electrical resistance with pressure for $T \gtrsim T_N$ in the sequence $UAgCu_4$–U_2Zn_{17}–UCd_{11}. Unlike $UAgCu_4$ and U_2Zn_{17}, UCd_{11} shows a relatively large pressure-induced increase in its room temperature resistance [R(16.9 kbar)-$R(0)/R(0) \approx 17\%$] that appears to be intrinsic since only insignificant hysteresis was observed upon repeated pressure cycling. UCd_{11} is also unique in that at the highest pressure shown an additional phase transition appears at temperatures below $T_N(P)$. Not shown in fig. 1 are results obtained at intermediate pressures where yet another phase transition manifests itself initially near 3 K.

From these measurements we arrive at a phase diagram shown in fig. 2 for each of the three systems. All show the ordering temperature increasing initially with pressure at a rate $\partial T_N/\partial P$ given in table 1. These values may be compared with those calculated from Ehrenfest's relation

$$\partial T_N/\partial P = 3VT_N\Delta\alpha/\Delta C_p, \tag{1}$$

where $\Delta\alpha$ and ΔC_p are the thermal expansion and specific heat changes respectively at T_N and V is the

Table 1
Relevant parameters of heavy-fermion magnets

Compound	T_N (K)	γ (mJ/(mol K^2))	d_{U-U} (Å)	$\partial T_N/\partial P^{(a)}$ (K/kbar)	$\partial T_N/\partial P^{(b)}$ (K/kbar)
UCd_{11}	5.05	840	6.56	0.070	0.060
U_2Zn_{17}	9.70	480	4.39	0.017	0.032
$UAgCu_4$	18.15	310	5.03	0.032	–
UCu_5	15.2	> 250	4.97	–	–

(a) measured near $P = 0$; (b) from eq. (1).

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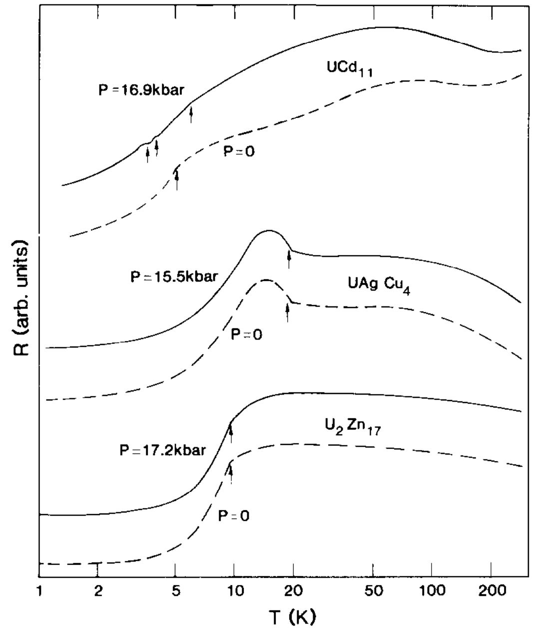

Fig. 1. Electrical resistance as a function of temperature for UCd_{11}, $UAgCu_4$ and U_2Zn_{17} at ambient and elevated pressures. Curves are displaced vertically for clarity. Arrows denote phase transition temperatures. Room temperature resistivities are estimated to be on the order of 100 $\mu\Omega$ cm for each material. $R(T)$ for $UAgCu_4$ is very similar to that of UCu_5 (see ref. [4]).

molar volume. In the two cases where this comparison can be made, we see that eq. (1) predicts the correct sign for $\partial T_N/\partial P$, with semiquantitative agreement in magnitude. Considering these materials as Kondo lattices, we can understand the variation of T_N and $R(T)$ with pressure in $UAgCu_4$ and U_2Zn_{17} in terms of a competition between Kondo and RKKY interactions [8,9] where in the case of $UAgCu_4$ the Kondo temperature T_K is much less than T_N but in U_2Zn_{17} T_K and T_N are comparable. At pressures not significantly greater than 17 kbar, we would expect $\partial T_N/\partial P < 0$ as T_K is pushed far above T_N in U_2Zn_{17}.

The most interesting behavior is exhibited by UCd_{11} in which two phase transitions (labeled T_2 and T_3 in fig. 2) are induced by pressure. The nature of these transitions is unknown; however, both transitions do respond non-linearly to an applied magnetic field with $\partial T_2/\partial H < 0$ and $\partial T_3/\partial H > 0$. The phase diagram for UCd_{11} suggests a possible relationship between T_1 and T_2 as well as between T_1 and T_3. Interestingly, ambient pressure specific heat measurements [1] on UCd_{11} show a shoulder in C/T versus T^2 near 3.5 K which agrees well with the temperature obtained by extrapolating the phase boundary $T_2(T, P)$ to $P = 0$, indicating that perhaps T_2 is beginning to form already at $P = 0$. Despite attempts to observe an unambiguous signature for T_2 below 3 kbar or for T_3 below 16 kbar, no resistive evidence for their presence could be detected in these regimes.

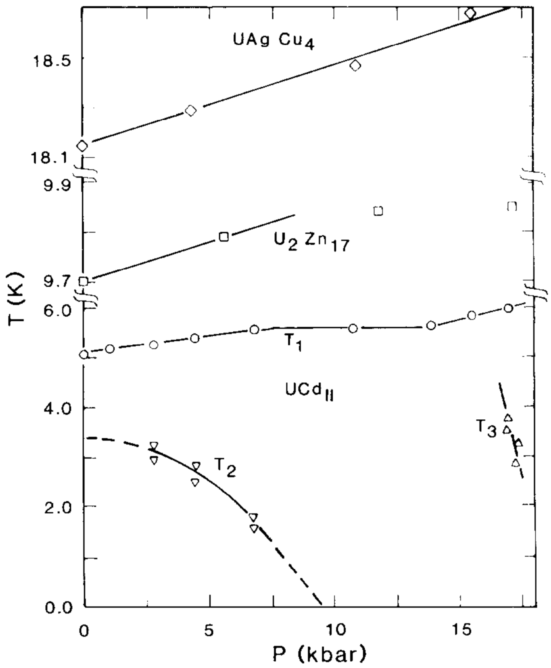

Fig. 2. Phase transition temperature versus pressure for $UAgCu_4$, U_2Zn_{17} and UCd_{11}. Note scale changes on vertical axis. T_1, T_2 and T_3 denote three phase transitions in UCd_{11}. Double symbols for T_2 and T_3 signify different criteria for determining the transition temperature. Pressure measurements are accurate to ±0.5 kbar.

In summary, we have studied the pressure response of heavy-fermion magnets UCd_{11}, U_2Zn_{17} and $UAgCu_4$. The effect of pressure on magnetic ordering in the latter two materials can be understood qualitatively in terms of competing Kondo and RKKY interactions, with $\partial T_N/\partial P$ in semiquantitative agreement with that calculated from Ehrenfest's relation. UCd_{11} shows unusual pressure behavior that requires additional experiments, e.g., magnetic susceptibility and neutron scattering under pressure, before its origin is understood.

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