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Low-temperature anomalies in the high-field specific heat of UCd_{11}

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The specific heat of UCd_{11} has been investigated in magnetic fields up to 16 T and in the temperature range 0.35–10 K. In addition to the well-known 5-K magnetic transition, another anomaly is observed below 2 K in fields larger than 10 T. This anomaly can be related to the previously reported signature in the temperature-dependent resistivity under pressure. Furthermore, the magnetic fields do not affect the high-temperature specific heat (above the 5-K transition). The latter observation is discussed in relation to the origin of the extremely large electronic effective mass of UCd_{11}.

INTRODUCTION

The origin of extremely large electronic effective masses (m^*) in materials commonly described as heavy-fermion systems continues to be a subject to intense investigations.[1] Although a few possible scenarios for the mass enhancement have been postulated, there is no general consensus as yet.[2,3] In fact, the possibility that not one, but several different mechanisms lead to the observed large m^* in this group of materials has recently received some experimental support.[4] UCd_{11} is one of the most interesting heavy-fermion compounds for a number of reasons. It undergoes a phase transition at temperature close to 5 K.[5] The exact nature of the transition is not well understood, although the magnetic-field dependence of the critical temperature (T_N) and results of the muon-spin-rotation experiment[6] are consistent with an antiferromagnetic ordering. However, the shape of the specific heat divided by the temperature versus temperature squared (C/T vs T^2; see Fig. 1) below and above the transition is rather uncommon for antiferromagnetic ordering. A broad tail, from 5 K (T_N) to about 8 K, is observed on the high-temperature side of the transition, while a well-pronounced shoulder appears below T_N (at 3–4 K). Several explanations have been put forward to explain this unusual temperature dependence of the specific heat below T_N.[7-9] Temperature derivatives of electrical resistivities obtained under sufficiently high hydrostatic pressure are indicative of two additional phase transitions.[8] However, there has not been found any direct evidence for these transitions from either magnetic-susceptibility or specific-heat measurements.[8] The question whether the anomalies in the resistivities under pressure are related in any way to the unusual specific-heat behavior at ambient pressure remains unanswered.

The temperature dependence of the specific heat of UCd_{11} well above T_N, i.e., above 8 K, is atypical of the known heavy-fermion systems [with the exception of U_2Zn_{17} (Ref. 10)]. C/T is proportional to T^2 up to at least 13 K with an extremely high value of the intercept of the ordinate axis, of about 840 mJ/(K^2 mol).[5] The value of the linear term above 8 K in UCd_{11} is the highest among all heavy-fermion systems (in this temperature range). We have used large magnetic fields up to 16 T in order to suppress the 5-K transition and to find out whether this simple $C/T=\gamma+\beta T^2$ relation between the specific heat and temperature persists down to lower temperatures or whether a Kondo-like increase in C/T vs T^2 takes place as for many other heavy-fermion systems.

RESULTS AND DISCUSSION

The specific heat was measured using a time-constant method. The temperature in magnetic fields was determined from a $SrTiO_3$ capacitance sensor (above 1 K) and 220-Ω Speer resistor (below 1 K), calibrated in zero field against a Ge resistance thermometer. Experiments were performed on two assemblies of single crystals, each of a total weight of about 3 mg of UCd_{11}, which has a cubic $BaMg_{11}$ crystal structure. Magnetic fields were applied in the (100) crystallographic direction.

The magnetic-field data have several interesting features, including an appearance of a second transition at lower temperatures for sufficiently high fields. Following the notation of Ref. 8, we denote the upper temperature transition as T_1 and the lower as T_2 and the critical temperatures, as inferred from the maxima in C/T, T_{c1} and T_{c2}, respectively.

Let us discuss first the effects of magnetic fields on T_1 that can be partially inferred from Fig. 1 and Table I. T_{c1} decreases faster than linearly with a magnetic field,

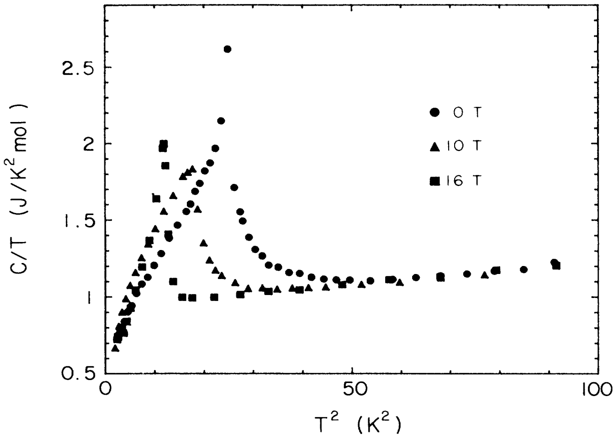

FIG. 1. C/T vs T^2 of UCd$_{11}$ in $H=0$, 10, and 16 T.

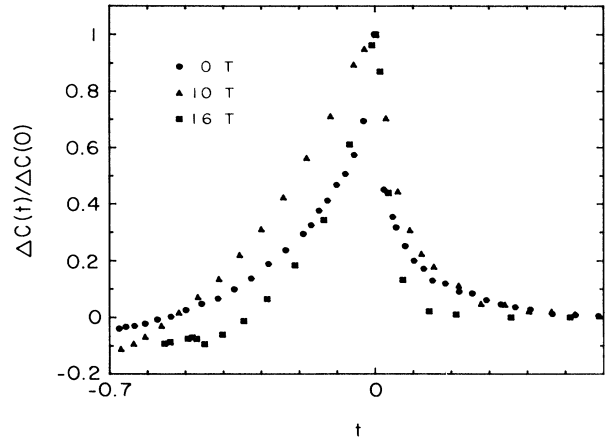

FIG. 2. $\Delta C(t)/\Delta C(0)$ vs t for T_1 transition in $H=0$, 10, and 16 T. ΔC is a difference between C and C_N (see text), and t is a reduced temperature $t=(T-T_{c1})/T_{c1}$.

but sublinearly with the square of the magnetic field; the last dependence is observed for a number of antiferromagnets. The value of C/T at T_{c1} changes nonmonotonically with the magnetic field, as shown in Fig. 1. First, it decreases between zero field and 10 T, then increases to 14 T (not shown for clarity) value, and again decreases between 14 and 16 T. This peculiar behavior can be further correlated with an appearance of the second transition somewhere between 10 and 14 T and with a broadness of the T_1 transition in a particular field as illustrated in Fig. 2. In Fig. 2, ΔC, a difference between the total specific heat and extrapolated "normal-state" specific heat, normalized by its maximum value is shown versus the reduced temperature $t=(T-T_{c1})/T_{c1}$. The normal-state specific heat has the temperature dependence $C_N=\gamma T+\beta T^3$, where γ and β coefficients are obtained from the data at sufficiently high temperatures where C/T is proportional to T^2. The T_2 transition, which is rather broad in the zero field, becomes even broader in the 10-T field, but its width is substantially reduced in 14 T (not shown in Fig. 2) and 16 T. The nonmonotonic dependence of the width of the T_1 transition and the narrowing of the transition in high fields is very uncharacteristic of an antiferromagnetic phase transition. In order to explore further the nature of the T_1 anomaly, we investigated the temperature dependence of the specific heat in the vicinity of T_{c1}. The zero-field specific heat associated with the T_1 transition (after subtracting C_N) is proportional to $\ln|T-T_{c1}|$ for $0.1<|T-T_{c1}|<0.7$ K (inset of Fig. 3). The last observation has been deduced from a small number of data points and therefore cannot be completely conclusive. Such a temperature dependence has been derived for the two-dimensional Ising model[11] and has been observed for some antiferromagnets.[12] It is interesting to note that the proportionality constants between ΔC and $\ln|T-T_{c1}|$ below and above T_{c1} are close to each other, are in the range of values found for other antiferromagnets,[12] and are consistent with the Ising model. The character of the specific-heat–temperature relation around T_{c1} changes upon application of the magnetic field (Fig. 3). The highest-field data (14 and 16 T) in the temperature range between T_{c1} and T_{c2} can be described by the relation $C=350T+115T^3$, where T is expressed in K and C in mJ/(K mol). An extremely large T^3 coefficient (β) points to the importance of the antiferromagnetic spin-density-wave contribution. [The Debye temperature, extrapolated from the specific-heat data above 8 K,[8] is 152 K, which corresponds to β of only 5.75 mJ/(K^{4}mol).]

TABLE I. Critical temperatures for various values of magnetic field. The absolute error of C is about the same for all measurements (5%, mainly instrumental error) and therefore does not effect an analysis of the field dependence of C.

H (T)	T_{c1} (K)	C/T_{c1} [mJ/(K^{2}mol)]	T_{c2} (K)
0	5±0.01	2610	
10	4.21±0.02	1830	
14	3.77±0.02	2190	1.93±0.03
16	3.45±0.02	1995	1.80±0.03

From Fig. 3 one can also see a gradual development of the T_2 transition. The 10-T data do not exhibit the second peak at low temperatures, but a broad shoulder that may be a precursor of the transition. The transition is clearly observable in higher fields of 14 and 16 T. Obviously, at this point, we are unable to speculate on the origin and character of the T_2 anomaly. The entropy associated with T_2 is at the most 2–3 % of R ln2 and thus ordering of local moments at T_{c2} is rather unlikely. On the other hand, a small decrease of T_{c2} between 14 and 16 T (see Fig. 3 and Table I) is compatible with some kind of antiferromagnetic ordering among itinerant electrons. For this reason, high-field, low-temperature magnetic susceptibility measurements of UCd$_{11}$ are in order. The small size of available crystals and rather poor sensitivity of our ac susceptibility apparatus did not allow us to per-

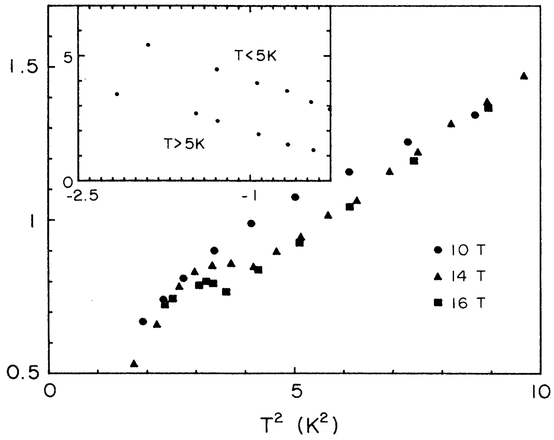

FIG. 3. Low-temperature C/T vs T^2 in $H=10$, 14, and 16 T. The inset shows ΔC in J/(K mol) vs $\ln(|T-T_{c1}|)$ for $H=0$ T.

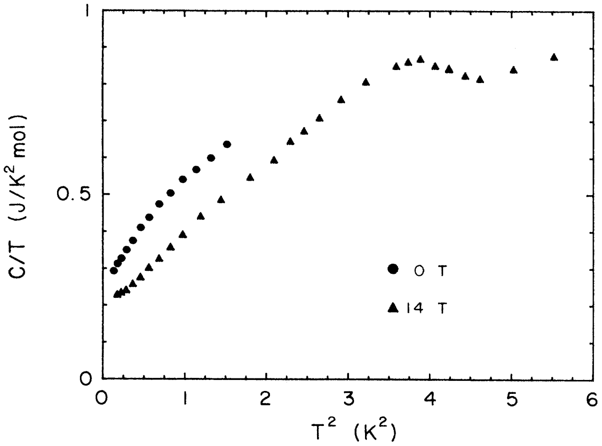

FIG. 4. Low-temperature C/T vs T^2 in $H=0$ and 14 T. These data were obtained on different assembly of UCd_{11} crystals than the data presented in Figs. 1–3.

form a reliable measurement. On the other hand, magnetic susceptibility under pressure measurements[8] were probably done at too high temperatures (for applied fields and pressures) to observe a distinct T_2 anomaly.

We have confirmed the T_2 transition by a separate specific-heat measurement performed with a different appartus, involving different addenda (to avoid any possibility of a systematic error), on another assembly of single crystals of UCd_{11}. This second measurements was conducted in zero and 14-T fields, in the temperature range from 0.35 to 1.1 K (0 T) and from 0.35 to 2.3 K (14 T) (see Fig. 4). An ordering at T_{c2} leads to a further decrease of the electronic density of states as evidenced by the lowest-temperature values of C/T in 14 T versus the corresponding value for 0 T.

Our findings agree with the pressure–magnetic-field–temperature phase diagram of UCd_{11} inferred from the electrical resistivity[8] in at least two points. Both the specific-heat measurements and electrical resistivity studies do not find direct evidence for two separate phase transitions in zero field at ambient pressure, and second, both investigations indicate that T_{c2} decreases with H at constant pressure (ambient pressure in the case of the specific heat and 3.8 kbar in the case of the magnetoresistance measurements). Whether UCd_{11} undergoes only one low-temperature–zero-field–ambient-pressure phase transition or two transitions in a very narrow temperature range [similarly to another heavy-fermion compound $CeAl_2$ (Ref. 13)] has to be investigated thoroughly by other experimental techniques. Certainly, the dramatic changes of some characteristics of the T_1 anomaly, such as its width and the temperature dependence of the specific heat in the vicinity of T_{c1}, strongly favor the second eventuality. Also, additional microscopic probes in high fields are required to establish the nature of the T_2 transition.

Finally, we discuss effects of large magnetic fields on the normal-state specific heat of UCd_{11}. From Fig. 1, displaying C/T vs T^2, we can draw two important conclusions: (1) The temperature range where C/T is proportional to T^2 extends to lower temperatures as the magnetic field is increased, and (2) the values of C/T above the T_1 anomaly do not depend on H within the resolution of our measurement. [The least-squares analysis yields values of 290±50 mJ/(K^{2}mol) and 200±15 K for the linear coefficient of the specific heat, γ, and the Debye temperature, respectively. Our best value of γ is about 10% higher than the one previously reported.[4] The discrepancy may be due to sample dependence or different temperature ranges used in the fitting procedure.] These above-mentioned properties put UCd_{11} in a sharp contrast with a "canonical" heavy-fermion system,[1] which displays an increase of C/T upon lowering of T and a substantial field dependence of the low-temperature specific heat, especially when the Sommerfeld coefficient is as high as 900 mJ/(K^{2}mol).[14] (The notable exception is UBe_{13} for which the normal-state specific heat shows only moderate sensitivity to high fields.) Moreover, gross properties of the canonical heavy fermion can be accounted for by the Kondo model.[2] In view of our results, it is doubtful that the electron-mass enhancement in UCd_{11} originates from the Kondo effect and some other explanation is needed. It has been pointed out,[15] in reference to U_2Zn_{17}, that crystal-field contributions can result in large values of the specific heat at low temperatures for systems with sufficiently small separation of crystal-field levels. U_2Zn_{12} displays remarkable similarity of zero-field properties, including linearity of C/T with T^2 above the Néel temperature at 9.7 K,[10] with those of UCd_{11}. However, a replacement of only 2% of Zn by Cu destroys the antiferromagnetic state in U_2Zn_{17}, while C/T stays almost constant at about 500 mJ/(K^{2}mol), down to at least 1.5 K.[16] Such a temperature dependence over a decade of temperature is inconsistent with the behavior expected for the low-temperature side of a Schottky anomaly even if some reasonable broadening of the crystal-field levels is assumed. In view of the analogy between UCd_{11} and U_2Zn_{17} and because of the above-described magnetic-field dependence of the normal-state specific heat of UCd_{11} [conclusion (1)], we feel that the crystal-field explanation is not correct for either of the compounds.

In summary, we have demonstrated that the specific heat in high magnetic fields is a promising technique in exploring ordered as well as normal-state properties of heavy fermions, and in particular, it can be very discriminating with respect to the choice of a relevant theoretical model. The normal-state specific-heat results of UCd_{11} are incompatible with predictions of the Kondo model. Similar studies of U_2Zn_{17} are planned.

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