Magnetic field study of the “hidden transition” in UCD$_{11}$

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The specific heat of UCD$_{11}$ was measured in magnetic fields to 27 T. Besides the antiferromagnetic transition, there is a second transition that can be clearly resolved in fields between 14 and 20 T. This second transition (at $T_m$) extrapolates to a broad shoulder in $C/T$ in zero field. The two lines of transitions cross somewhere between 20 and 23 T. $T_m$ displays unusually weak dependence on the strength of the magnetic field. Our results argue for the intrinsic origin of this “hidden” transition. © 2005 American Institute of Physics. [DOI: 10.1063/1.1850394]

I. INTRODUCTION

UCD$_{11}$ is one of the first discovered uranium-based heavy fermion compounds with puzzling low-temperature properties. As most of other U heavy fermion materials, it belongs to a cubic crystal structure, BaHg$_{11}$ type. It orders antiferromagnetically at about 5 K. The specific heat ($C$) above the antiferromagnetic anomaly has an unusual temperature dependence. $C/T$ is proportional to the square of temperature ($T^2$) with a positive slope and an enormous intercept of $C/T$ axis of nearly 900 mJ/K$^2$ mol. This temperature variation was observed up to at least 14 K, resulting in a very large measured entropy at 14 K, more than 12 J/K$^2$ mol. This very large entropy persisting to low temperatures suggests that excited crystalline electric fields (CEFs) of U contribute to the formation of the heavy fermion state, as opposed to better-understood Ce-based systems, for which only the ground CEF state is relevant. Similarly interesting is the specific heat of UCD$_{11}$ in the ordered state. A broad shoulder in $C/T$ near 2–3 K suggested a possibility of a second transition (we denote the temperature of this lower-temperature anomaly by $T_m$), hindered by the pronounced peak at 5 K ($T_n$). Our previous investigation$^2$ in magnetic fields to 16 T, produced a clear evidence of this lower-temperature transition. Magnetic fields 14 T and larger reduce the size of the anomaly at $T_n$, uncovering a lower-temperature peak at $T_m$. There was a small decrease of $T_m$ between 14 and 16 T, suggesting the antiferromagnetic nature of the transition at $T_m$. In the present investigation we have focused on magnetic fields greater than 16 T, up to the highest field applied of 27 T. Our study was performed on the same sample used in Ref. 2, allowing for a direct comparison of these two sets of results. Some sample dependence was recently reported for UCD$_{11}$.

II. RESULTS AND DISCUSSION

Our specific heat measurements of UCD$_{11}$ were performed in zero field, and in fields of 18, 20, 23, 25, and 27 T, using a resistive magnet in the National High Magnetic Field Laboratory. The temperature range for each field value was chosen to map the magnetic field phase diagram. Figure 1 shows $C/T$ versus $T$ for $H=0$, 18, 20, and 27 T. Note that the range of $C/T$ values shown in the two lower panels is reduced by a factor of 1.5 with respect to the upper two panels. The aforementioned shoulder corresponding to the lower-temperature transition in magnetic fields is marked by an arrow in the upper panel of Fig. 1. Two specific-heat anomalies can be clearly resolved in 18 and 20 T. The temperatures of these anomalies in 18 T are only slightly reduced with respect to those found previously in 16 T. There is a further decrease of both $T_X$ and $T_m$ for $H=20$ T. Another interesting aspect of these data is a continuous reduction of the maximal value of $C/T$ at $T_X$ as a function of the field’s
strength versus the very weak if any dependence of the $C/T$ value at $T_m$, suggesting somewhat different character of these two transitions. On the other hand, we were not able to resolve two transitions in the highest applied fields of 23, 25 (not shown), and 27 T (the lowest panel of Fig. 1).

The magnetic field phase diagram of UCd$_{11}$ obtained from the specific heat is shown in Fig. 2. In this figure we have included also data points from our previous study$^3$ for fields lower than 18 T. Points corresponding to the higher-temperature transition ($T_{3N}$) are connected by a solid line. The highest field in which we have clearly observed this higher-temperature anomaly is 20 T. Peaks in $C/T$ found in 23, 25, and 27 T seem to fall on the line representing $T_m$. Our phase diagram exhibits close similarities to that obtained by the high-field magnetization investigation on a different UCd$_{11}$ sample.$^3$ This high-field magnetization study implies that the $T_N$ line becomes approximately parallel to the temperature axis at temperatures lower than 2 K and intercepts the field axis just above 20 T. The magnetization versus field at these lowest temperatures changes the slope near 20 T.$^4$ The corresponding moment in this field is about 1 $\mu_B$/U. Above 20 T, the magnetization still increases with the field, most probably due to low-lying excited CEF levels.

The extrapolation of the line corresponding to $T_m$ (Fig. 2) intercepts the temperature axis somewhere between 2.5 and 3 K. This latter temperature seems to coincide with the position of the shoulder in $C/T$ in the ambient field, shown in the upper panel of Fig. 1. This lower-temperature transition shows up more clearly in the $H=0$ specific-heat investigation of Aoki et al.$^3$ although at a somewhat lower temperature of 1.7 K. According to the capacitance study,$^3$ $T_m$ is almost field independent to 7 T, but decreases slightly for fields larger than 7 T, up the highest field used of 13 T. Our $T_m$ line seems to imply an inflection point around 20 T, thus for the field at which both lines of transitions most probably intercept.

If the antiferromagnetic character of the 5 K transition in UCd$_{11}$ is now quite unambiguously determined by a variety of experimental techniques,$^1$ the origin of the transition at $T_m$ is not established. $T_N$ and $T_m$ have quite different field dependences, excluding the possibility of two different parts of a sample undergoing a similar antiferromagnetic transition at two different temperatures. Based on investigations of two different samples one can conclude that $T_N$ does not show important sample dependence while $T_m$ is sample dependent. Very small entropy associated with the anomaly at $T_m$ (Ref. 3) in zero field can be used to argue for an extrinsic origin of this transition. However, these entropies become comparable in fields of 18–20 T. There also seems to be some interaction between the two anomalies displayed by the change of the slope $dT_m/dH$ in the field corresponding to the crossing of the two lines of transitions. Thus, our investigation favors the intrinsic character of the transition at $T_m$.

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1Z. Fisk et al., Phys. Rev. B 30, 6360 (1984).
2B. Andraka, G. R. Stewart, and Z. Fisk, Phys. Rev. B 44, 10346 (1991).
3D. Aoki et al., J. Phys. Soc. Jpn. 68, 3117 (1999).
4T. Inoue et al., Physica B 281–282, 204 (2000).
5A. L. Cornelius et al., Phys. Rev. B 59, 13542 (1999).