HIGH MAGNETIC FIELD SPECIFIC HEAT AND MCE OF URu_2Si_2*

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We have measured the specific heat and magnetocaloric effect of URu_2Si_2 at magnetic fields up to 45 T. The large specific heat anomaly at $T_0(H = 0) = 17$ K shifts to lower temperatures when the magnetic field is increased and is suppressed at 36 T. Between 36 T and 38 T a new anomaly in the specific heat *vs* temperature indicates a magnetic phase never previously reported. At $H \geq 40$ T no transition is observed and crystal electric field effects dominate the specific heat. Measurements of the magnetocaloric effect are used to stablish a new phase diagram for URu_2Si_2.

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URu_2Si_2 is a heavy fermion superconductor with $T_c = 1.8$ K [1, 2] that displays an intriguing low temperature second-order phase transition at $T_0 \simeq 17$ K. In this work we measured for the first time the specific heat *vs*

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temperature at constant magnetic fields up to 45 T, and the magnetocaloric effect during field sweeps. The measurements were done in the hybrid magnet at the National High Magnetic Field Laboratory. The specific heat was measured using a calorimeter made of plastic materials and a standard relaxation method in both small and large delta T limits. The magnetocaloric effect was measured using the same setup, recording the sample temperature while the magnetic field was swept at a rate of ~ 12 T/min. Single crystals of URu_2Si_2 (Sample ♯1) were grown by tri-arc melting (Czochralski method) stoichiometric amounts of U, Ru and Si and then annealing the so obtained crystal for one week at 950° C. Sample ♯2 was grown by arc-melting followed by vertical float-zone refining [3].

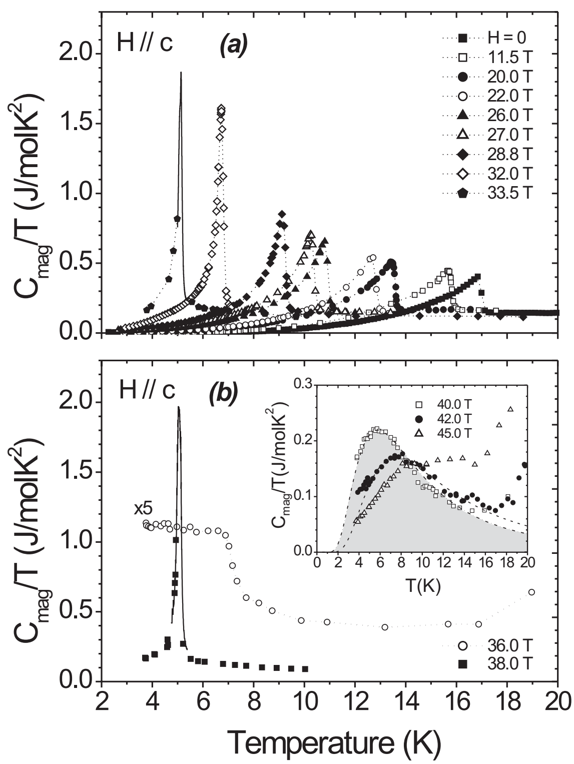

Fig. 1. (a) C_m/T *vs* T for magnetic fields from zero to 33.5 T in sample ♯1. Note how the anomaly associated to the hidden order phase becomes sharper and symmetric at low temperatures. The solid line was measured using large delta T method. Dotted lines are guides to the eye. (b) C_m/T *vs* T for $H = 36$ T and $H = 38$ T. The suppression of hidden order is followed by a small step-like anomaly which in turn is followed by a new peak. Inset: C_m/T *vs* T for $H = 40$, 42, 45 T shows only crystal electric field effect component. Dash-dotted and dashed lines indicate fits using the Schottky expression.

The specific heat of sample ♯1 was measured on a bar-shaped 9 mg piece with the c-axis along the bar principal axis. From the total measured specific heat ($C_{\rm tot}$) we subtracted the phonon contribution ($C_{\rm ph}$) as measured in a sample of $ThRu_2Si_2$ [4], and from spin fluctuations ($C_{\rm sf}$) [5]. Fig. 1(a) displays $C_m/T = (C_{\rm tot} - C_{\rm ph} - C_{\rm sf})/T$ *vs* temperature for magnetic fields up to 33.5 T. We observe that the anomaly at $T_0(H)$ is shifted to lower temperatures by the external magnetic field and changes shape becoming sharper and more symmetric before becoming completely suppressed near 36 T. Fig. 1(b) displays C_m/T measured at 36 T and 38 T. We see here the complete suppression of the specific heat anomaly associated with the hidden order phase. Indeed, the data at $H = 36$ T show only a small step feature. Surprisingly, at $H = 38$ T yet another large anomaly develops in the specific heat which is then suppressed with a magnetic field of 40 T, and its origin is unknown at the present time. Fig. 1(b) Inset shows the specific heat measured at $H = 40$ T, 42 T, and 45 T. In this regime all that is left in C_m is a Schottky-like anomaly, indicating a specific heat dominated by crystal electric field (CEF) effects. We fit our data to an expression for a Schottky anomaly using $\Delta_{\rm 40T} = 1.58$ meV, $\Delta_{\rm 42T} = 2.03$ meV, and degeneracy factor equal to 0.75, obtaining an associated entropy $\sim 0.3 \times R$. The effect of the external field in Δ point to a possible singlet f-electron CEF level crossing at $H = 34$ T. Such a level crossing was proposed as a semiquantitative explanation for the observed phenomenology in magnetic fields [6]. The entropy recovered at the transition, computed from data in Fig. 1(a), remains close to 0.15 R for all magnetic fields, in good agreement with previous results [5].

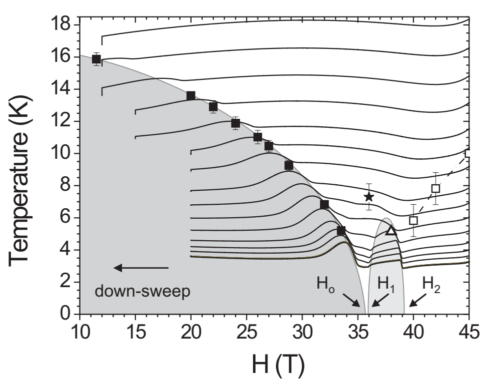

Fig. 2. Phase diagram constructed using transition temperatures from specific heat *vs* temperature measurements and MCE temperature traces. The *hidden order phase* is suppressed at $H_0 = 35.5$ T at $T = 3$ K. A new phase is observed below $T = 6$ K between $H_1 = 36$ and $H_2 = 39$ T.

Fig. 2 exhibits the transition temperatures obtained from specific heat *vs* temperature at constant field in sample ♯1 and temperature changes observed in sample ♯2 due to magnetocaloric effect when, during the magnetic field down-sweep, various initial temperatures were used. Our temperature traces show no significant anomalies when the initial temperature is higher than $T_0 = 17$ K. When the initial temperature is lower than 17 K we observe one broad anomaly that moves to higher fields and sharpens as the temperature is reduced. Below 6 K we can distinguish three anomalies at H_0, H_1, and H_2 that shift with temperature. H_0 and H_2 both increase with decreasing temperature, while H_1 decreases. In this way the MCE curves are used to map the low temperature/high field phase diagram of URu_2Si_2. Some characteristics of this phase diagram include *(i)* T_0 vanishes at about $H = 36$ T; *(ii)* $H_1(T)$ and $H_2(T)$ together define a new region in the phase diagram; *(iii)* $H_0(T)$ and $H_1(T)$ seem to converge at the same point when $T \to 0$.

In summary, from specific heat *vs* temperature at constant field in URu_2Si_2 we observe the complete suppression of the *hidden order phase* at magnetic fields close to 36 T. We also observe a new high field phase above 36 T that is suppressed with magnetic fields in excess of 39 T. Between 39 T and 45 T, no transition is observed in the specific heat *vs* temperature. In this range all that is left in the magnetic component of the specific heat is a Schottky-like anomaly. Fitting the experimental curves to a Schottky expression suggests singlet f-electron crystal electric field levels that cross near 35 T as the possible origin. Magnetocaloric effect data was used to obtain a detailed low temperature/high field phase diagram. The two observed phases in URu_2Si_2 seem to meet at a bicritical point close to $H_0 = 36$ T.

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