Specific heat of URu_2Si_2 in fields up to 42 T: Clues to the hidden order

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(Received 18 March 2002; revised manuscript received 17 September 2002; published 13 January 2003)

The large ΔC observed at 17.5 K in URu_2Si_2 is inconsistent with the small, $0.04\mu_B$ moment measured for the antiferromagnetism observed starting (perhaps coincidentally) at the same temperature. We report measurements of this specific-heat transition, thought to be due to some hidden order, in magnetic fields between 24 and 42 T, i.e., through the field-region where three metamagnetic transitions are known to occur at 35.8, 37.3, and 39.4 T. The response of ΔC in single crystal URu_2Si_2 to magnetic field, which includes a change to ΔC being possibly associated with a first-order phase transition for high fields, is analyzed to shed further light on the possible explanations of this unknown ordering process. At fields above 35 T, a new high-field phase comes into being; the connection between this high-field phase revealed by the specific heat and earlier magnetization data is discussed.

DOI: 10.1103/PhysRevB.67.014404 PACS number(s): 71.27.+a, 75.30.Kz, 75.30.Mb, 75.40.Cx

I. INTRODUCTION

The compound URu_2Si_2 was initially a focus of research due to the discovery[1] of the coexistence of antiferromagnetism ($T_N = 17.5$ K) and superconductivity. Later work[2,3] focused on the three steps in the magnetization (called "metamagnetic transitions") discovered[4] by magnetoresistance and magnetization measurements at 35.8, 37.3, and 39.4 T with the field parallel to the *c* axis in this medium heavy fermion compound. Recently, the explanation for the large (in proportion to the ordered moment[5] of only $0.04\mu_B$) size of the discontinuity in the specific heat at 17.5 K in URu_2Si_2 has been the subject of much theoretical as well as experimental effort,[6–22] with explanations as diverse as quadrupolar order[16] and various unusual kinds of magnetic order being proposed.[23] (Some of the latter have been found to be inconsistent with later neutron-diffraction experiments.[8]) The term hidden order has been coined to refer to the as-yet unknown order that leads to the large observed ΔC, i.e., "hidden" in the sense that the nature of the order has not yet revealed itself to a variety (e.g., neutron scattering,[8] resistivity,[9] and NMR[17]) of measurement techniques that have been used to probe the order responsible for the large ΔC.

It is apparent from various perspectives that the observed anomaly in the specific heat is *not* due to the observed[8] low moment magnetism. One argument (see Ref. 10) is that Landau theory predicts that $\Delta C/T \approx (k_B/T_N)$ times the square of the ratio of the observed moment to the paramagnetic moment observed (either from neutron scattering or from an analysis of the magnetic susceptibility fitted to the Curie-Weiss law) above T_N. For URu_2Si_2, this gives[10] a prediction for $\Delta C/T$ that is three orders-of-magnitude smaller than that observed. Recent neutron-diffraction[19] and NMR (Ref. 20) measurements under pressure have been interpreted to imply that the observed magnetic moment is in fact from a minority ($\sim$1%) second phase, although this is still under discussion.[22]

Hall-effect data to 40 T by Bakker *et al.*[3] show transitions in the Hall coefficient at 35.6, 36.2, and 39.2 T (i.e., the Hall-effect data appear linked to the three transitions at 35.8, 37.3, and 39.4 T in the magnetization data). The decrease of the Hall coefficient down to a value close to zero at 39.2 T was interpreted[3] as a closing of the gap in the Fermi surface which was opened at 17.4 K. This energy gap of about 110 K, measured below the ordering temperature in zero and applied field by various methods,[9] was seen by resistivity data[9] in fields to 25 T to be associated with order that had a critical field of 40 T, i.e., with the hidden order. Thus, since the Hall-effect data appear to show the closing of the gap at 40 T associated[9] with the hidden order and since the Hall effect appears also to have three transitions comparable to those shown in the magnetization, perhaps the "metamagnetic" transitions in *M* vs *H* between 35.8 and 39.4 T are also associated with the hidden order.

Since the deciding measurement that determines the existence of the hidden order is the specific heat, since the specific heat (resistivity) of URu_2Si_2 has been reported[12] only in fields up to 17.5 (25[9]) T, and since there is clearly something unusual occurring in the phase diagram as a function of magnetic field at 35.8, 37.3, and 39.4 T, we undertook to measure[24] specific heat in dc fields up to 42 T in the new dc hybrid magnet at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee on single crystals of URu_2Si_2. (Prior to the advent of this unique magnet, all measurements above 35 T had to be performed in pulsed field magnets.) These measurements should help to further elucidate the nature of the hidden order by determining the response of the anomaly over the entire field range expected[9] to suppress the anomaly and also by determining how the anomaly reacts (if at all) at the fields where the jumps in the magnetization occur.

II. EXPERIMENT

Although there are some minor differences[25] in measuring specific heat in fields above 24 T versus techniques used[24] for $H \leqslant 24$ T, in general the measurements are quite similar. In order to avoid eddy current heating (much more a problem at the old Francis Bitter National Magnet Laboratory than in the new National High Magnetic Field Laboratory due to the quieter, modern transistor-based power supply used at the

NHMFL), the reference block[24] used is made out of nonelectrically conducting sapphire. The transfer standard to calibrate the sample platform thermometer (a flash-evaporated film of Au-Ge with[26] a *positive* magnetoresistance—i.e., magnetic field enhances the thermometer's sensitivity—of about 20% at 1 K in 33 T) is a capacitance thermometer from Lakeshore Cryotronics that is essentially[27] field independent up to 45 T.

Flat platelet single crystals, with the *c* axis perpendicular to the platelet, were obtained by removing the crystals that form as surface facets on a large (∼2-g mass) arc-melted button of high purity URu_2Si_2, a technique that has also been used[28] to produce single crystals of $CeRu_2Si_2$. A collage of 16.18 mg of these crystals thermally bonded to a sapphire disk using GE7031 varnish was used for the specific-heat measurements. The Ru and Si used were 99.95% and 99.9999% pure, respectively, from Johnson Matthey; the U used was the best that is commercially available, an electrotransport refined material from Ames Lab. The zero-field specific heat of these crystals is comparable with the best samples reported, with $\Delta C/T_{order}=335$ mJ/mol K^2 at the high-temperature ordering transition in the present work vs 320 mJ/mol K^2 in Ref. 12 (ΔC is defined as C/T_{max}-C/T extrapolated from the higher-temperature, normal-state data down to T_{max}). Further, the transition width of the hidden order phase transition in zero field in the sample used in the present work is 0.30 vs 0.38 K in the sample measured in Ref. 12. The susceptibility with field parallel to both the *c* and *a* axes was also measured (where the two directions have a factor of ∼8 difference in magnitude as well as a much different temperature dependence), with excellent agreement with the literature[1] results.

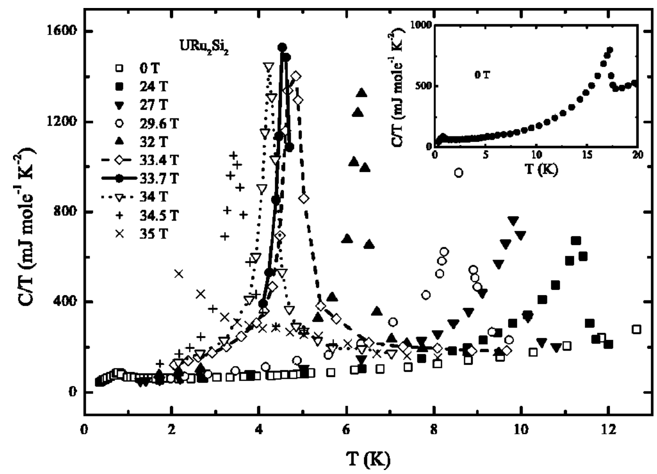

FIG. 1. Specific heat *C* divided by temperature *T* at low temperatures between 0 and 35 T for single crystal URu_2Si_2 with the field aligned along the *c* axis. Note the very rapid change of both the magnitude of $\Delta C/T_{order}$ and T_{order} for fields above 33.7 T: in only 1.3 T T_{order} decreases by more than 2.5 K as the hidden order phase transition is being finally suppressed to $T=0$.

III. RESULTS AND DISCUSSION

The specific heat divided by temperature, C/T, of single crystal URu_2Si_2 between 24 and 35 (where the first metamagnetic transition is reported at 35.8 T) is plotted in Fig. 1, between 35 and 37.5 in Fig. 2, and between 37.5 and 42 T in Fig. 3. The discussion of these data divides naturally into two field regions.

A. $H\leqslant 35$ T—suppression of the hidden order anomaly

The trend of the ordering temperature decreasing and $\Delta C/T_{order}$ increasing with increasing field up to 33.7 T apparent in Fig. 1 was also qualitatively observed by van Dijk *et al.*[12] in their specific-heat data in fields up to 17.5 T. As may be seen, the anomaly in C/T sharpens up remarkably with increasing field above 27 T, becoming in appearance (see discussion below) like a first-order phase transition. This sharpness of the anomaly makes the accurate determination of T_{order} for each field straightforward.

One result of these measurements is the critical field of the specific-heat anomaly as a function of temperature, shown in Fig. 4. The temperature dependence of the critical field of this hidden order anomaly up to 35 T is not unusual at all, and follows at low field (as was known from e.g., the specific-heat data up to 17.5 T from Ref. 12) $H_c=H_0[1-(T/T_{order})]^{0.5}$ rather well, where $H_0\sim 35.3$ T and T_{order} is the zero-field ordering temperature (defined as the temperature of the peak in C/T, rather than the onset of the transition) of 17.4 K in URu_2Si_2. At high field, i.e., near H_0

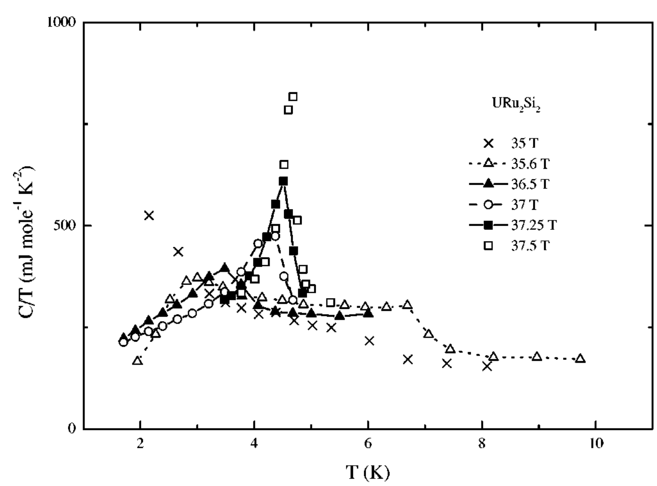

FIG. 2. Specific heat *C* divided by temperature *T* at low temperatures between 35 and 37.5 T for single crystal URu_2Si_2 with the field aligned along the *c* axis. Although any peak in 35 T occurs below 2 K, already by 35.6 K a small anomaly is visible in C/T near 3 K, which, as discussed in the text, is the appearance of a high-field phase. T_{order} and $\Delta C/T_{order}$ for this anomaly then continue to increase with increasing field in the field range shown here. A second anomaly at higher temperatures (∼6 K) may be apparent in the 35-T data but is clearly visible in the 35.6-T data. Data in this temperature range were not taken again until 38.1 T (at which point the small anomaly appears to be absent, see Fig. 3). This anomaly may correspond to a second high-field anomaly bounded by the two lower dashed lines shown in Fig. 4 corresponding to the two lower metamagnetic transitions at 35.8 and 37.3 T, as discussed in the text.

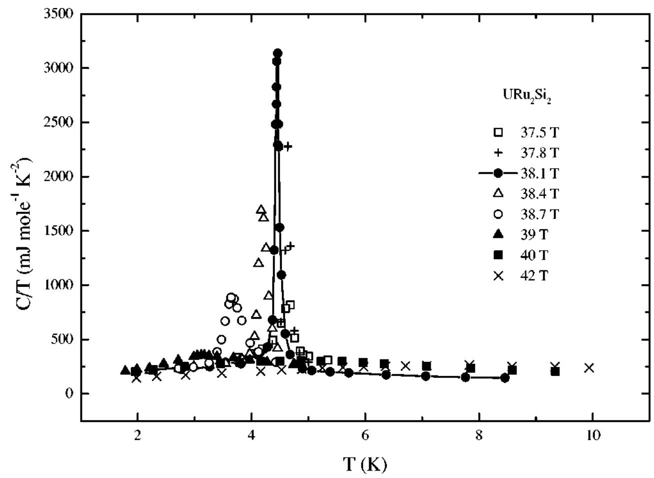

FIG. 3. Specific heat C divided by temperature T at low temperatures between 37.5 and 42 T for single crystal URu_2Si_2 with the field aligned along the c axis. T_{order} decreases monotonically for fields starting at 37.5 T, and $\Delta C/T_{order}$ has a maximum at 38.1 T.

$=35.3$ T, our data shown in Fig. 4 allow the determination that H_c follows $H_0[1-(T/T_{order})^2]$. These two temperature dependencies in the two limits $T\rightarrow T_{order}$ and $T\rightarrow 0$ provide useful information for theories (e.g., Ref. 7) which examine the nature of the hidden order.

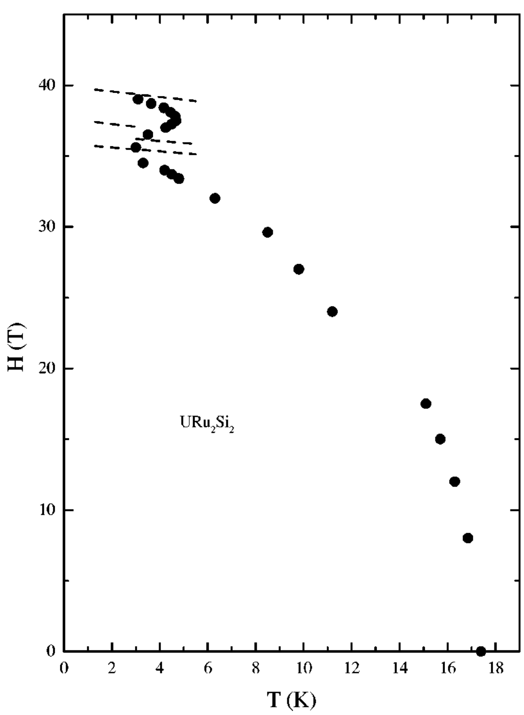

FIG. 4. The critical field is plotted versus T_{order} for single crystal URu_2Si_2 with the field aligned along the c axis using the temperature of the maximum in C/T for T_{order}. The data for $H\leqslant 17.5$ T are from Ref. 12, while the dashed lines at high fields—as discussed in the text—represent the temperature dependence of the fields where the three jumps in the magnetization occur from Ref. 29. Note the rapid approach to a zero slope for dH/dT as $H\rightarrow 35$ T, as well as the high-field phase that is induced by field between 35.6 and 39 T (data from Figs. 2 and 3.)

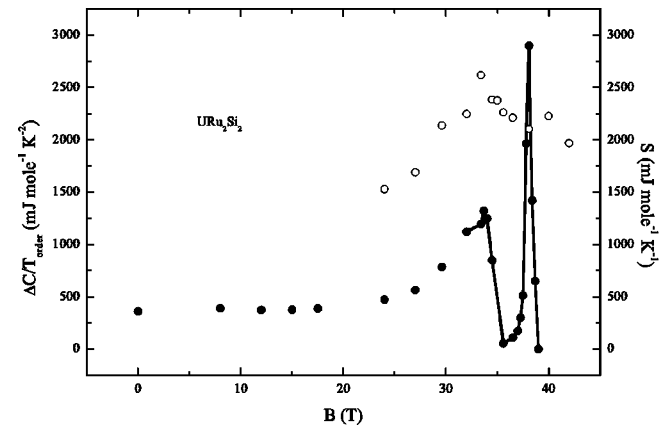

FIG. 5. $\Delta C/T_{order}$ for the anomalies in the specific heat (shown in Figs. 1–3) plotted as a function of field (data for $H\leqslant 17.5$ T from Ref. 12). These values have not been corrected for the jump due to the first-order phase transition and thus overestimate the quantity $\Delta C/T$ in the theoretical treatment in Ref. 7. Note the two peaks in $\Delta C/T_{order}$ at 33.7 and 38.1 T, with the higher-field peak occurring over a very narrow range of field. Also shown is the entropy up to 10 K as a function of field, showing [although with a less fine resolution than for $\Delta C/T_{order}$, since data up to 10 K necessary to calculate S (10 K) were not taken for every field due to time constraints] that also the peak in the entropy clearly occurs at a field below that of the jump in the magnetization. In relating these entropy results via $dS/dH=dM/dT$ to what is known about M as a function of field and temperature, the relation $d^2S/dH^2=d/dT$ (dM/dH) implies—since we know that the jump in the magnetization with increasing field at 35.8 T broadens with increasing temperature—that d^2S/dH^2 should be negative around 35.8 T. With the accuracy of the entropy data and the spacing of the data as a function of field, this relation between second derivatives is difficult to confirm.

The field behavior of $\Delta C/T_{order}$ is plotted in Fig. 5. Special care was taken while measuring the data to accurately determine the field where the maximum in C/T occurs. As clear from Figs. 1 and 5, this field is 33.7 T to an accuracy of better than 0.3 T. This does not agree with the field determined in magnetization measurements for the first jump in M as a function of field, which is at 35.8 T. This disagreement is also apparent when considering the entropy as a function of field (also shown in Fig. 5). These facts rule out explanations for the jump in magnetization (related to the entropy through the Clausius-Clapeyron equation) which involve large changes in the entropy (e.g., level crossing transitions). It is interesting to note that the Hall-effect data[3] taken up to 40 T show an anomaly (unremarked upon in Ref. 3) at 0.6 K between 33.8 and 34.5 T (i.e., similar to the field of 33.7 T where we see the maximum in C/T) that is actually slightly *larger* than the anomaly between 35.1 and 36.1 T that was identified[3] as corresponding to the magnetization anomaly at 35.8 T.

The sharpness of the anomalies shown in Fig. 1 for $H\geqslant 29.6$ T raises the questions of whether the hidden order anomaly in $\Delta C/T$ becomes first order in high field, and if so, at what field this first occurs. If the hidden order phase transition becomes first order with increasing magnetic field, this

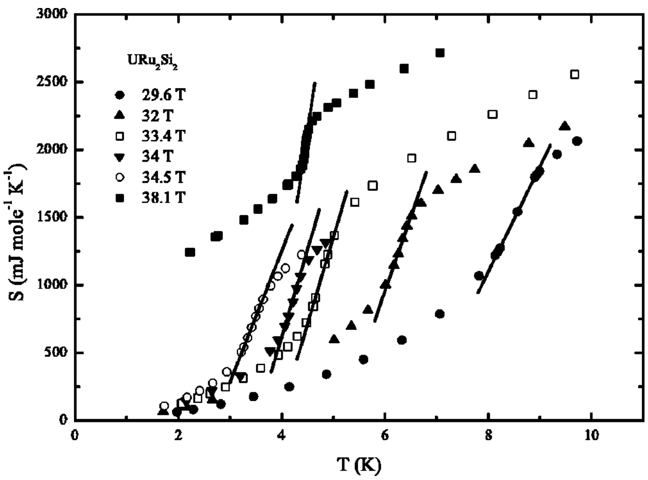

FIG. 6. Entropy S vs temperature for single crystal URu_2Si_2 with the field aligned along the c axis for fields between 29.6 and 34.5 T, as well as for 38.1 T (plotted with the same vertical and horizontal scales but shifted upwards by 1500 mJ/mol K to make the data more visible). The entropy ($\equiv\int_0^T C/T' dT'$) is calculated numerically from the C/T data shown in Fig. 1 and, for 38.1 T, in Fig. 3. For a second-order phase transition, or for a first-order phase transition rounded by local impurities or defects (Ref. 30), only one inflection point is expected in the S vs T curve. Note the tendency for the slope of S vs T to become steeper around 34 T. Note also that the slope of S vs T for the 38.1-T data is much higher (a factor of 2) than for any other field, i.e., the field-induced transition at 38.1 T appears to be the most likely to be first order in nature.

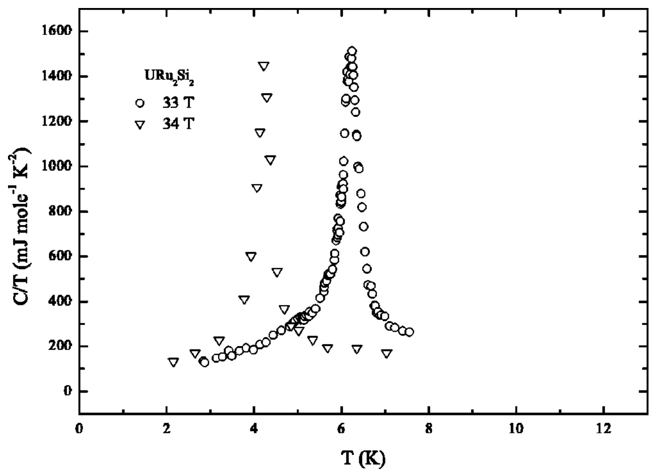

FIG. 7. Specific heat divided by temperature vs temperature for a 5.03-mg single crystal of URu_2Si_2 with the field aligned along the c axis in 33 T (high-density data as discussed in the text to address the order of the transition) and, for comparison, data in 34 T from Fig. 1. These 33-T data were taken with much smaller (0.01 vs 0.05 K) temperature excursions (Ref. 24) than for the other data in this work in order to smear out the transition as little as possible.

would certainly be an important constraint for any theory explaining this as-yet not understood order. To address this question, plots of the entropy S for fields $\geqslant$29.6 T are presented in Fig. 6. Leaving the discussion of the 38.1-T entropy results also shown in Fig. 6 for the next section, it is clear from Fig. 6 that, although the slope of S vs T is a maximum around 34 T and despite the quite large and sharp specific-heat anomalies in Fig. 1, the entropies never have the truly vertical jump characteristic of a first-order phase transition. In order to address the question of the possibility that inhomogeneities in the collage of single crystals created a broadening of the jump in the entropy, and to improve the data density vs temperature (see, e.g., Fig. 1) achieved within the (severe) time constraints of measuring in the 45-T hybrid magnet, we have measured during a separate week at NHMFL the specific heat of the largest URu_2Si_2 crystal (5.03 mg) from the collage in 33 T (see Fig. 7). These measurements were performed using the most painstaking data density possible with our measurement technique, with very small temperature excursions, and a very high density of measurement temperatures around the peak in C/T. (Measurement time in a magnet that reaches 33 T is easier to obtain than time in the unique, 45-T hybrid magnet at NHMFL.) The entropy calculated from these high-density data in 33 T is shown in Fig. 8. The slope of the steepest part of the entropy vs temperature data of this single-crystal data with much higher data density, Fig. 8, is only about 9% higher than that of the data for the most comparable field, 33.4 T, data shown in Fig. 6. Thus, unless there is a high level (significantly above what is expected) of either (i) internal (e.g., variation in stoichiometry) inhomogeneities in the individual single crystal, (ii) inhomogeneities in the field,[31] or (iii) impurities or defects[30] in the single crystal, the hidden order transition for fields $\leqslant$34.5 T appears to be second order.

Above 33.7 T (see Figs. 1 and 5), the size of the anomaly in C/T falls precipitously, well before the first anomaly in the magnetization at 35.8 T. At 35 T, the peak in the C/T anomaly is below our lowest temperature of measurement (2 K) and is (based on an extrapolation of the data at 34 and 34.5 T where the peak is still visible) smaller by about a factor of 3 from the maximum (see Fig. 5) in $\Delta C/T_{\mathrm{order}}$ at 33.7 T. One possible explanation for the rather precipitous fall in T_{order} and the size of the transition between 34.5 and 35 T shown in Fig. 1 is best seen by considering the phase

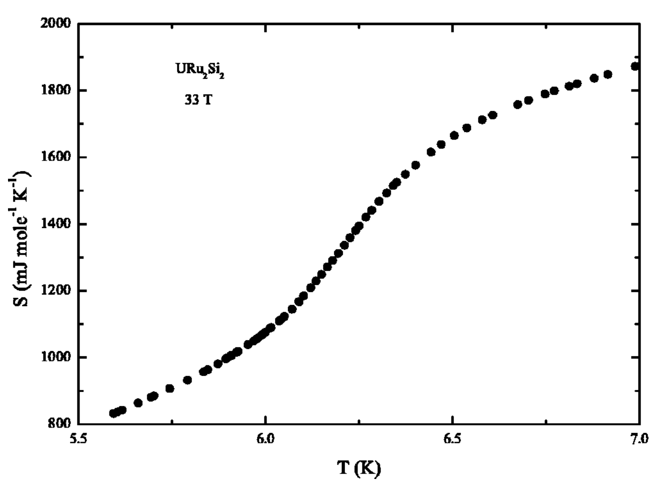

FIG. 8. Entropy S vs temperature for single crystal URu_2Si_2 with the field aligned along the c axis for 33 T using the data from Fig. 7.

diagram shown in Fig. 4. As the hidden order transition temperature is suppressed with increasing field towards lower temperatures, Fig. 4 shows that this phase line becomes more and more horizontal as $H \rightarrow 35$ T (or slightly greater). At this point in a phase diagram, measurements in a given field as a function of temperature will show a broadening of any field-dependent phase transition. Measurements to lower temperatures would help further investigate this possibility.

B. $35 \leqslant H \leqslant 42$ T—a new high-field phase

The specific-heat data between 35 and 37.5 and between 37.5 and 42 T are shown in Figs. 2 and 3, respectively. A specific-heat anomaly at $\sim$3 K is seen to be moving up in temperature with increasing field already in 35.6 T after the hidden order anomaly was suppressed below 2 K in 35 T. While the second anomaly in the magnetization data taken at 1.3 K is at[4] 37.3 T (or 37.4 T in the more recent work of Sugiyama *et al.*[29]), the data in Fig. 2 show a monotonic increase in the size of both $\Delta C/T_{\mathrm{order}}$ as well as T_{order} up to 37.5 T, after which (see Fig. 3) the anomaly continues to increase in size up to 38.1 T, but T_{order} begins to decrease for $H > 37.5$ T. The fields and T_{order} are shown in Fig. 4; clearly the high-field phase diagram shows another phase transition that appears after the hidden order anomaly is suppressed around 35 T. Considering now the entropy, S (10 K) shown in Fig. 5 trends slightly downwards in this field range after its peak around 33 T.

A second, small anomaly at $\sim$6.5 K is visible in the 35.6-T data, with indications of a related anomaly in the trend of the 36.5-T data up to 6 K. This smaller anomaly is absent by 38.1 T, and—as discussed below—may correspond to a small second field-induced transition in addition to the very large anomaly in C/T with a maximum as a function of field at 38.1 T. Further data in the field and temperature ranges 35–38 T and 4–10 K are needed to resolve this. Another possibility is based on the discussion of the phase diagram and the 35-T data at the end of the section above: this broadened anomaly in 35.6 T is again where a phase boundary line is nearly horizontal. Thus, the broadened anomaly with its beginning at $\sim$6.5 K may in fact just be due to the spreading out of a phase transition as the phase boundary for the new, higher-field phase joins the field axis horizontally. Clearly from Fig. 2, the anomaly above 36.5 T begins to sharpen and increase in temperature, corresponding to a finite slope for the phase boundary at increasing fields as shown in Fig. 4.

Sugiyama *et al.*[29] consider the temperature dependence of the three magnetization anomalies (shown as dashed lines in Fig. 4), which addresses the intercomparability of the anomalies in magnetization measured—in most cases—at different points (1.5 K and 35.8, 37.3, and 39.4 T in the early work of de Visser *et al.*[4] and at 1.3 K and 35.4, 37.4, and 39.2 T in the more recent work of Sugiyama *et al.*[29]) in the H,T phase diagram than the anomalies in C/T. They find that the anomaly in the magnetization at 1.3 K and 37.4 T disappears by around 3 K and a new anomaly appears at 36 T faintly in their 3-K data but grows in size and is quite visible in their 3.6 and 4.2 K data. This anomaly is comparable to the part of the H,T phase space sampled by our specific-heat data in 36.5 T, where an anomaly in C/T is peaked at 3.5 K.

Considering now the specific-heat data in the field range around the third anomaly in the magnetization, Fig. 3 (see Fig. 4 for the resultant phase diagram), we see that the growing anomaly in C/T evident in 37.5 T in Fig. 2 rises up very sharply as a function of field (see also Fig. 5), peaking in magnitude at 38.1 T and 4.44 K in a transition that—even more than the transitions shown in Fig. 1—is reminiscent of a first-order phase transition. One important comparison (see Fig. 6) is the steepness of the S vs T curve at the anomaly. As shown in Fig. 6, the anomaly at 38.1 T is clearly, based on the criterion of how steeply S rises as a function of temperature, more first order in appearance. (Quantitatively, the slopes of the entropies at 38.1 and 33.4 T at their steepest sections differ by a factor of 2). The question then arises as to whether the transition evinced by the (rather steep) entropy anomaly in 38.1 T shown in Fig. 6 is a broadened first-order phase transition, or simply a rather sharp second-order phase transition. Thus, just as a second-order phase transition should theoretically have a discontinuous change in the specific heat at the transition, but in fact there is always a finite and sometimes a significant transition width ΔT in the specific-heat jump, a first-order phase transition may also have a finite transition width ΔT in the entropy jump. In zero field at 17.5 K, our sample of single crystal URu_2Si_2 has sufficient inhomogeneity to have a ΔT of 0.30 K in its specific-heat jump. The width of the steep increase in the entropy at the transition in 38.1 T is less than 0.20 K. High data density, low-temperature excursion data will be taken on the 5.03-mg single crystal of URu_2Si_2 just as was done for the 33-T data discussed above as soon as hybrid magnet time is available in order to further investigate the possible first-order nature of the 38.1-T transition.

In the phase diagram constructed by Sugiyama *et al.*,[29] the highest-field anomaly occurs at 39.1 T for 4.4 K. This disagreement, plus the complete lack of any hint in the magnetization data of a rapid variation with field of the magnitude of this third anomaly, imply once again that the specific-heat results are presenting additional information about the phase diagram of URu_2Si_2 in high magnetic field. In the specific heat measured at various fields as a function of temperature, an anomaly appears in C/T at 35.6 T and grows smoothly in magnitude (see Fig. 5) with increasing field up to 38.1 T, with a peak in T_{order} of 4.68 K at the slightly lower field of 37.5 T. This is incorporated in the high-field portion of the phase diagram shown in Fig. 4.

The magnetization, rather than showing a region of existence in the phase diagram of the high-field phase, shows[29] a smooth, gradual decrease in the fields of the upper and lower anomalies with increasing temperature, and a decrease in the field of the middle anomaly with increasing temperature that shows a sudden decrease at around 3 K. Thus, the magnetization-data-generated phase diagram shows the three separate, apparently independent dotted lines drawn in Fig. 4. Comparing the magnetization and the specific-heat data graphically in the phase diagram in Fig. 4 suggests that the two higher-field magnetization anomalies may mark the *boundaries* of the large $\Delta C/T$ high-field phase transition observed in the specific heat. Whether or not the slight anomaly in the 35.6-T data in Fig. 2 corresponds to a second high-field phase that is bounded by the lower two of the dashed lines in Fig. 4 representing the *H*,*T* behavior of the two magnetization anomalies at 35.8 and 37.3 T depends on the outcome of further work. It is interesting to note the possible connection in the jump in the magnetization phase line for the middle magnetization anomaly and the swerving away from joining the ordinate for the lower boundary of the specific-heat phase boundary shown in Fig. 4.

IV. CONCLUSIONS

Specific heat indicates that the hidden order anomaly is suppressed at ~35 T, and that the hidden order anomaly in high field before its suppression appears to remain second order. The order of this transition is important for understanding the nature of the hidden order in URu_2Si_2 and needs to be pursued in further work since whether the anomaly in the specific heat for $H \cong 33.7$ T becomes first order is only approximately determined by the data in the present work. In addition, the specific-heat data reported here show the existence of a high-field phase in URu_2Si_2, which is apparently linked to the two higher-field magnetization anomalies first seen in 1987.[4] This high-field phase may be first order in nature. Indications of a second high-field phase observed in the narrow field range between 35.6 and 38.1 T may also be explained by the rather flat *H* vs *T* phase boundary in this part of the phase diagram.

Recently, M. Jaime *et al.*[32] measured the high-field specific heat of a single-crystal sample of URu_2Si_2 in dc fields up to 45 T, with a spacing of approximately 2 T between measurement fields in the field range between 30 and 40 T. They also find the high-field phase reported herein and, using magnetocaloric measurements in pulsed fields, also find evidence that suggests that the transition at ~39 T is "a first order-like transition in field."

ACKNOWLEDGMENTS

The authors gratefully acknowledge helpful discussions with Professor M. Wortis. Work at the University of Florida was performed under the auspices of the U.S. Department of Energy, Contract No. DE-FG05-86ER45268. Data were taken at the NHMFL, Tallahassee, which is operated under the auspices of the U.S. National Science Foundation.

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