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Shireen Adenwalla
University of Nebraska-Lincoln, sadenwalla1@unl.edu

J. B. Ketterson
Northwestern University

S. K. Yip
Northwestern University

S. W. Lin
University of Wisconsin - Milwaukee

M. Levy
University of Wisconsin - Milwaukee

See next page for additional authors

Authors
Shireen Adenwalla, J. B. Ketterson, S. K. Yip, S. W. Lin, M. Levy, and Bimal K. Sarma

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S. Adenwalla, J. B. Ketterson, and S. K. Yip
Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208

S. W. Lin, M. Levy, and Bimal K. Sarma
Department of Physics, University of Wisconsin–Milwaukee, Milwaukee, Wisconsin 53201
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In this paper we present a thermodynamic analysis of the UPt_3 superconducting phase diagram in the H-T plane. The analysis relates the specific-heat jumps to the slopes of the four phase-transition lines. The existing data are found to be in good mutual agreement, and consistent with the assumption that the inner phase-transition line at low field, T_c^*, is second order for both $\hat{\mathbf{H}}\perp\hat{\mathbf{c}}$ and $\hat{\mathbf{H}}\parallel\hat{\mathbf{c}}$. For $\hat{\mathbf{H}}\perp\hat{\mathbf{c}}$, the inner phase-transition line at higher field, $H_{\rm FL}$, is first order albeit with a small latent heat. For $\hat{\mathbf{H}}\parallel\hat{\mathbf{c}}$, it can be second order.

The heavy-fermion superconductor UPt_3 is a strong candidate for unconventional superconductivity. A number of experiments have shown the existence of multiple superconducting phases. Early measurements of the ultrasonic attenuation in the superconducting state revealed a peak in the attenuation as a function of field.[1–3]

This peak can be interpreted as a phase transition and the position of this line in the H-T plane at finite fields as a phase boundary. This transition line was named the $H_{\rm FL}$ transition as it was conjectured that it may be the signature of a transition in the flux lattice. Subsequent heat-capacity measurements showed two superconducting transitions, even at zero field, separated by about 50 mK.[4] These heat-capacity signatures moved to lower temperatures and closer together as the field was increased.[5,6]

The position at which the phase-transition lines met was still an open question as the measurements were made on different samples and the ultrasonic attenuation signature faded out as one approached H_{c2}. A number of phase diagrams for superconducting UPt_3 had been proposed.

Recent ultrasonic velocity measurements[7,8] on the heavy-fermion superconductor UPt_3 have confirmed the existence of three superconducting phases which appear to meet at a single point on the H_{c2} curve. These measurements are the first to reveal the entire superconducting phase diagram on a single sample using a single measurement technique. Our measurements give strong support for a single multicritical point at which three superconducting phases and the normal phase meet. Yip, Li, and Kumar[9] have derived thermodynamic constraints for such a multicritical point if at least three of the lines are second order. Since the upper critical field curve (above and below the multicritical point) is second order, if we assume that the transition T_c^* is also second order (no evidence of a latent heat has been seen at this transition), then we can apply this analysis to the phase diagram obtained from ultrasonic measurements.

In Fig. 1 we show a schematic phase diagram for UPt_3. We follow the notations of Ref. 9. The phases are designated by the letters A, B, C, and D. The multicritical point is denoted by P, and we shall use p_1, p_2, p_3, and p_4 as the slopes at P of the lines AN, BN, CA, and CB, respectively. AN and BN are the H_{c2} curves below and above the multicritical point, CA is the T_c^* line and CB is the $H_{\rm FL}$ line. We define r_1^2 and r_3^2 as the ratios of the heat-capacity jumps $\Delta C_{AN}/\Delta C_{BN}$ and $\Delta C_{CA}/\Delta C_{BN}$, respectively.

It has been shown in Ref. 9 that, under the above assumptions, the slope of line BC is given by

$$p_{41}=\frac{p_{13}r_3^2+r_1^2-p_{12}-E^{1/2}}{r_3^2p_{13}^2+r_1^2-p_{12}^2}\ , \tag{1}$$

where

$$E\equiv d-cr_3^2\ , \tag{2}$$

$$c\equiv r_1^2(1-p_{13})^2-(p_{13}-p_{12})^2\ , \tag{3}$$

$$d\equiv r_1^2(1-p_{12})^2\ , \tag{4}$$

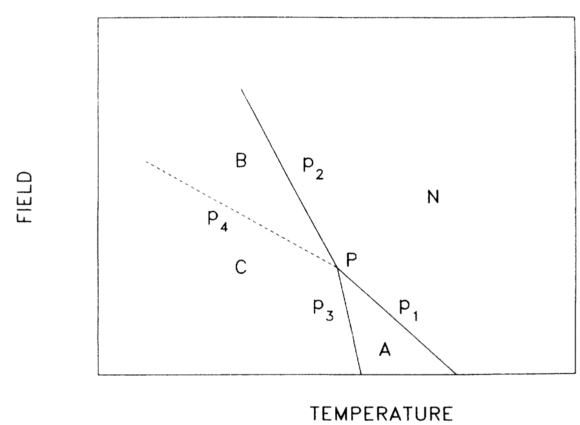

FIG. 1. Schematic phase diagram and notations.

and $p_{ij} \equiv p_i / p_j$.

Since the slopes in our sound velocity measurement are more accurately measured than the specific-heat jumps in Refs. 5 and 6, we invert Eq. (1) to give r_3 in terms of the slopes and r_1. To do this we rewrite Eq. (1) as

$$(r_3^2 a + b) = -[d - c r_3^2]^{1/2} \tag{5}$$

with

$$a \equiv p_{13}(p_{13}p_{41} - 1) , \tag{6}$$

$$b \equiv r_1^2(p_{41} - 1) + p_{12}(1 - p_{12}p_{41}) . \tag{7}$$

Upon squaring both sides, we obtain a quadratic equation in r_3^2 which we can solve readily. The result is

$$r_3^2 = -\frac{b}{a} + \frac{-c \pm f^{1/2}}{2a^2} , \tag{8}$$

where

$$f \equiv (2ab + c)^2 - 4a^2(b^2 - d) . \tag{9}$$

There are two possible signs for Eq. (8). By examining Eq. (5) it is clear that the criterion is such that the combination occurring on the left-hand side of Eq. (5) must be nonpositive. We see that, since all the slopes are negative and $|p_4| < |p_3|$, a is always negative for our present situation. The condition thus requires the sign be chosen so that

$$\pm f^{1/2} - c > 0 . \tag{10}$$

In Table I, we list the slopes at the multicritical point for $\mathbf{H} \perp \hat{\mathbf{c}}$. Using these values and Eq. (8), we calculate r_3^2 as a function of r_1^2. The results are shown in Fig. 2. The heat-capacity data of Hasselbach, Taillefer, and Flouquet[5] show that, as the multicritical point is approached, the size of the lower heat-capacity jump decreases, approaching a very small number at the multicritical point; i.e., $r_3^2 \approx 0$. There appears to be no discontinuity in the size of the upper heat-capacity jump at the multicritical point so that one would expect $r_1^2 \approx 1$. Figure 2 shows that r_3^2 decreases from 0.116 at $r_1^2 = 1.0$ to 0 at $r_1^2 = 1.14$. [It is useful to note that, near this region $a, b < 0$ and $c, d > 0$. Thus, by Eq. (10), the + sign in Eq. (8) has to be taken.] Thus, the experimental facts that the value of r_1 is close to 1 and the specific-heat jump on line AC approaches a very small value as one approaches the multicritical point are consistent (c.f., Ref. 10). This can be regarded as supporting evidence for the assumption that line AC is second order.

The thermodynamic analysis also allows us to determine the order of the transition BC. In Ref. 9 it has been shown that the condition for it to be second order is that

TABLE I. Slopes for $\mathbf{H} \perp \hat{\mathbf{c}}$ (T/K).

p_1	p_2	p_3	p_4	r_1^2	r_3^2
-3.7	-5.85	-10.2	-0.6	0.91	0.18
				1.0	0.11
				1.14	0.0

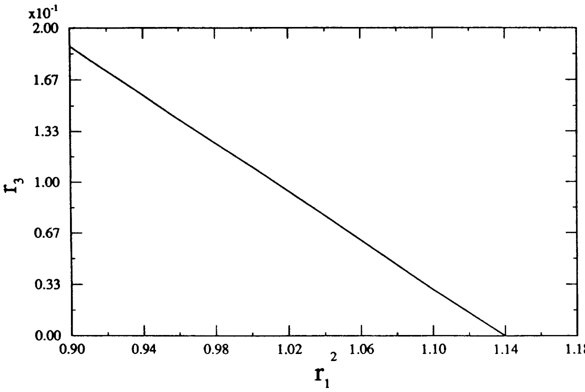

FIG. 2. Plot of r_3^2 vs r_1^2 for $\mathbf{H} \perp \hat{\mathbf{c}}$.

$E = 0$. At $r_1^2 = 1.0$, $r_3^2 = 0.116$, we find that $E = 0.0974$, and when $r_1^2 = 1.14$, $r_3^2 = 0$, we find that $E = 0.154$ (see Fig. 2). If one is willing to accept the hypothesis that the ratios of the specific-heat jumps and the slopes of the transition lines are determined to an accuracy of $\sim 10\%$ or better, then it is extremely unlikely that BC is a second-order transition, i.e., it is impossible to have $E = 0$ satisfied by varying the values of p's and r_1 within their error bars. This point can be stated in an alternative way. $E = 0$ implies, by Eqs. (2) and (5),

$$r_3^2 = \frac{d}{c} = -\frac{b}{a} , \tag{11}$$

which requires $r_1^2 = 0.45$, $r_3^2 = 0.55$ or $r_1^2 = 0.27$, $r_3^2 = 0.98$; i.e., the specific-heat jump on the H_{c2} line just below the multicritical point (i.e., AN) has to be only about $\frac{1}{2}$ of its value above it (i.e., BN) or less, while that on line AC should be comparable to that on BN. These values strongly disagree with those from the experiments.[5]

Granted that BC is first order, we can then determine the latent heat associated with this phase transition. Following the reasoning in Ref. 9, it is easy to derive the formula for the latent heat by subtracting the first derivatives of the free energy on either side of the transition

$$L = (\delta T)(\Delta C_{BN})[r_3^2(1 - p_{43}) + r_1^2(1 - p_{41}) - (1 - p_{42})] , \tag{12}$$

where δT is the deviation of the temperature at the point of interest from that of the multicritical point (hence, in our present situation it is always negative), ΔC_{BN} is the jump in the specific-heat across line BN, the sign of which is such that it is positive for a (physical) positive specific-heat jump when one lowers the temperature through the $B \rightarrow N$ transition. At $r_1^2 = 1.0$, $r_3^2 = 0.116$, we find that the value of the quantity inside the square bracket is -0.046, and when $r_1^2 = 1.14$, $r_3^2 = 0$, we get -0.06. (Notice that $L > 0$ as it should be.) As a result, the predicted value of the latent heat is small near the multicritical point (note that it is linear in the distance away from this point and vanishes exactly at P; however, the analysis only holds close to the point P). Using the heat-capacity data of Hasselbach, Taillefer, and Flouquet[5] and a value of -0.05 for the quantity inside the square brackets, we obtain a latent heat of 0.14 mJ/mol at a temperature 50 mK below the multicritical point. The heat capacity of UPt_3 is about 225 mJ/mol K, implying that, in order to see the latent heat signature, one would require a temperature resolution better than 0.6 mK at 400 mK. This may explain the fact that the latent heat across the BC transition has not been observed.[5]

For $\mathbf{H}\|\hat{c}$ (Table II), p_1 and p_2 are nearly equal. The small difference between them (0.06 T/K) is close to the limits of our resolution. If we take them to be exactly equal at the point P (i.e., there is no kink in H_{c2} for $\mathbf{H}\|\hat{c}$), then, using Eq. (8), we see that r_3 vanishes at $r_1=1$. At this r_1, E goes exactly to zero, implying that BC is a second-order transition. Specific-heat experiments[6] have indicated that r_1 is very close to 1, and r_3^2 very small.

If we assume the small difference between p_1 and p_2 is real (i.e., there is, in fact, a small kink in H_{c2}), then we find that r_3^2 vanishes when $r_1^2=0.98$. Thus, the experimental evidence that r_3^2 is indeed small and that r_1^2 is close to one are consistent with each other and can be taken as evidence that the AC transition is second order. Moreover, at almost the same value of r_1^2 (to within 1%), E vanishes, implying that the BC transition can be second order for $\mathbf{H}\|\hat{c}$. One can also easily check that E is indeed very small for these specific-heat jumps, and even if it is not, an examination of Eq. (12) shows that the latent heat would be extremely small. However, we cannot rule out the possibility that it is first order.

We now comment on the data of Bruls *et al.*[8] A feature that distinguishes their data from ours is that the slopes of the transition lines are practically continuous across the point P. Indeed, they draw their phase diagrams with precisely this feature. An analysis of this phase diagram, however, leads to significantly different thermodynamics from ours. With the assumptions $p_1=p_4$, $p_2=p_3$, one easily sees from Eq. (8) that

$$r_3^2=\frac{2a^2-c\pm|c|}{2a^2} .$$

TABLE II. Slopes for $\mathbf{H}\|\hat{c}$ (T/K).

p_1	p_2	p_3	p_4	r_1^2	r_3^2
-5.33	-5.39	-9.81	-1.68	0.98	0.0

Note that now $c=r_1^2(1-p_{12})^2>0$, a is still negative, and, hence, as before, the $+$ sign has to be chosen. This requires $r_3^2=1$ and, by examining the expression for E [Eq. (2)] or Eq. (11), that the BC transition is necessarily second order, i.e., P is necessarily a tetracritical point. Using the result $r_3^2=1$ and the fact that the total specific-heat jump from the normal phase to phase C should have identical values independently of whether one goes above or below the tetracritical point P, one finds that the specific-heat jumps are continuous along the phase-transition lines (i.e., $\Delta C_{AN}=\Delta C_{CB}$, $\Delta C_{BN}=\Delta C_{CA}$). This is in strong contrast to the fact that the specific jump on AC is much smaller than that on AN or BN at P.[6] In fact, the Frankfort phase diagrams are as if one just superimposes two completely noninteracting order parameters.

A recent experiment by Trappmann, Löhneysen, and Taillefer[11] measured the changes in T_c and T_c^* with pressure. The pressure-temperature phase diagram obtained appears to show a point at which three phases meet. A similar analysis of that phase diagram can be made (with pressure replacing magnetic field as the thermodynamic variable). If the transitions are all second order, then an additional phase-transition line should emerge from this point, unless all the lines are tangential with each other[9] (see Refs. 12 and 13). An experiment to distinguish between these two possibilities is highly desirable.

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[10] The specific heat reported in Fig. 4 of Ref. 5 at lower transition is the sum of the contribution from both transitions, which Ref. 9 has erroneously taken to be the entire contribution from the lower transition only. Hence, Ref. 9 has investigated a parameter region irrelevent for the present phase diagram, and their conclusion that the measured specific-heat jumps are inconsistent with the slopes should be ignored.

[11] T. Trappmann, H. V. Löhneysen, and L. Taillefer, Phys. Rev. B **43**, 13 714 (1991).

[12] One can see this by a microscopic picture as in that of Ref. 13.

There is one transition above the critical pressure P^* only if the splitting field (the square of the magnetization in Ref. 13) vanishes above this pressure. For this to be possible this splitting field has to vanish as $(P-P^*)^2$ (not linearly). This pressure dependence will produce a phase diagram with three second-order transition lines meeting at a point at a common slope. If the splitting field is linear in $P-P^*$, then above P^* there should still be two phase transitions.

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