# Thermodynamic considerations and the phase diagram of superconducting $UPt_3$

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In this paper we study the thermodynamics of bicritical and polycritical points, where at least two or three second-order phase-transition lines intersect, respectively. The results are applied to the superconductivity phase diagram of $UPt_3$.

## I. INTRODUCTION

It has long been suspected that $UPt_3$ is an unconventional superconductor. Various experiments have explored different regions of the magnetic-field–temperature ($H$-$T$) plane and suggest a variety of different phases. Specific-heat measurements in zero field[1] have indicated two distinct phase transitions. Ultrasonic attenuation[2] and torsional oscillator[3] experiments have shown various peaks, often identified with phase transitions involving the superconducting order parameter(s). $H_{c_2}$ measurement for $\mathbf{H}\perp\hat{\mathbf{c}}$ (Refs. 4 and 5) shows a kink at $(T^*,H^*)\approx(0.37\text{ K}, 0.5\text{ T})$, identified as a bicritical or polycritical point where several phase-transition lines meet. Recently, the specific-heat jumps along three of these lines have been measured.[6] This kink seems to be absent for $\mathbf{H}\|\hat{\mathbf{c}}$. $H_{c_1}$ curves,[4,7] however, seem to have kinks for $\mathbf{H}\perp\hat{\mathbf{c}}$ but not $\mathbf{H}\|\hat{\mathbf{c}}$. Recently, neutron scattering[8] experiments further indicated the seemingly different effect of superconductivity on the antiferromagnetic order parameter in different regions of the $H$-$T$ plane.

None of the above experiments have been able to detect all of the phase transitions on the entire $H$-$T$ plane. Moreover, the ultrasonic attenuation and torsional oscillator damping peaks tend to become somewhat ambiguous when the "transition lines" meet. Thus, often phase diagrams are constructed via extrapolations based on collections of data from two or more experiments. Theoretical efforts are then hindered by the lack of a precisely determined phase diagram. There is, however, a consensus that the relevant superconducting order parameter is a vectorial one: $\boldsymbol{\eta}=(\eta_1,\eta_2)$ (where $\boldsymbol{\eta}$ transforms like a vector in the $a$-$b$ plane under the symmetry group elements of the crystal). Ginszburg-Landau (GL) free energies have been written for this order parameter, following symmetry requirements, and also for the antiferromagnetic order parameter $\mathbf{m}$ which exists below $\sim 5$ K.[9–14] (Details of the models differ as to the assumptions on the behavior of $\mathbf{m}$.) The phase diagram is then determined by minimizing this free energy. It is, however, a nontrivial task to solve the resulting nonlinear differential equations, and one is often left to resort to educated guesses about the solution and the corresponding phase diagram. The task of reconciling theoretical expectations about the phase diagram is made enormously difficult by this ephemeral character of the available information. In fact, so far phase diagrams involving three[12–14] or four[6,7] transition lines meeting at one or more points have been proposed.

In this paper, we point out a different approach that is enormously helpful in settling some of these questions, namely, analyzing *thermodynamical constraints* on a phase diagram. We shall find that some phase diagrams proposed thus far are either forbidden outright or at least quantitatively in error. We also discuss the implications on those which are thermodynamically possible. Our work parallels an earlier approach by Leggett[15] to study the phase diagram of superfluid $^3$He. The assumptions needed are simply the continuity of the free energy and its derivatives (for second-order phase transitions) which lead to consistency requirements about the slopes of the phase boundaries and changes in thermodynamical variables across them. Thus the arguments are model independent and lead to general requirements irrespective of the details of the Ginzburg-Landau free energy.

The outline of this paper is as follows: In Sec. II, we analyze a point where three transition lines meet. We analyze the constraints if two of these lines are second order. In Sec. III we consider a polycritical (tetracritical) point. We provide the constraints on the slopes and specific-heat jumps if there are at least three second-order transition lines. The implications on the superconductivity phase diagram of $UPt_3$ are discussed. In Sec. IV we summarize our results.

## II. THERMODYNAMICAL CONSIDERATIONS

### A. Bicritical points

In this subsection we shall study some thermodynamical aspects of phase diagrams involving three transition lines meeting at a point. Recently, Blount, Varma, and Appeli[14] constructed a phase diagram as shown in Fig. 1 based on their analysis of a GL theory.

We would like to point out that Fig. 1 is thermodynamically forbidden. In particular, we shall show the

 

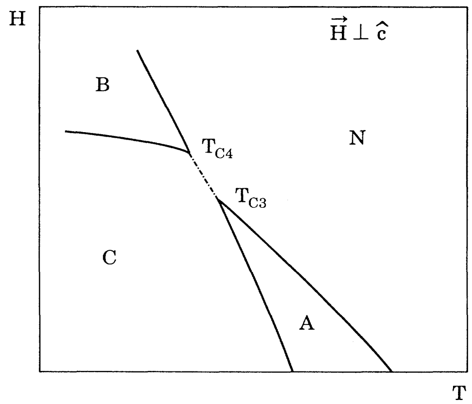


FIG. 1. The phase diagram proposed by Blount, Varma, and Aeppli (Ref. 14). Second-order lines are represented by full lines (as in all figures). The order of the line between $T_{c_3}$ and $T_{c_4}$ is not specified (these lines will be represented by dashed-dotted lines).

following (see Fig. 2)

*Theorem.* Suppose the phase $N$ has a second-order instability, into the phase $A$, as we lower the temperature at fixed field, and which in turn has a second-order instability towards the phase $B$ at a still lower temperature. Then it is impossible that $AN$ and $BA$ meet at a finite angle so that there is a phase-transition line (irrespective of its order) between $N$ and $B$, i.e., Fig. 2 is impossible.

The method used in the proof of this statement is closely parallel to that of a related theorem of Leggett.[15,16] Consider the behavior of the free energy $G(T,H)$ near the point $P$. Since $N \rightarrow A$, $A \rightarrow B$ are both second order, the first derivatives of $G$ are continuous around $P$ (i.e., the same in all phases near $P$). Denote the second derivatives of $G$ at $P$ by

$$\left[\frac{\partial S}{\partial T}\right]=\alpha \ , \tag{2.1a}$$

$$\left[\frac{\partial b}{\partial T}\right]=-\frac{\partial S}{\partial H}=\beta \ , \tag{2.1b}$$

$$\frac{\partial b}{\partial H}=-\gamma \ , \tag{2.1c}$$

in each of the three phases, where $b \equiv \partial G/\partial H$. Define the differences

$$\Delta\alpha_{AN}=\alpha_A-\alpha_N \ . \tag{2.2}$$

The slopes of $NA$, $AB$ are determined by the condition of continuity of $G$ and $S$, i.e.,

$$p_1 \equiv \left[\frac{dH}{dT}\right]_{AN}=\frac{\Delta\alpha_{AN}}{\Delta\beta_{AN}}=\frac{\Delta\beta_{AN}}{\Delta\gamma_{AN}} \ , \tag{2.3}$$

$$p_2 \equiv \left[\frac{dH}{dT}\right]_{BA}=\frac{\Delta\alpha_{BA}}{\Delta\beta_{BA}}=\frac{\Delta\beta_{BA}}{\Delta\gamma_{BA}} \ , \tag{2.4}$$

respectively.

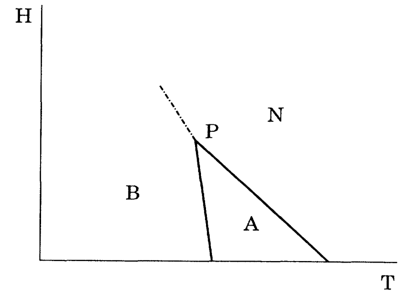


FIG. 2. This phase diagram is thermodynamically forbidden, irrespective of the nature of the line $BN$, if $NA$ and $AB$ are second order.

Stability of $A(B)$ over $N(A)$ at lower temperatures (for $H<H^*$) demands the positivity of the specific-heat jumps and, hence,

$$\Delta\alpha_{AN}>0, \quad \Delta\alpha_{BA}>0 \ . \tag{2.5}$$

These inequalities still hold as one approaches $P$, except under the special circumstances where the specific-heat jumps vanish. Let $(dT,dH)$ be the differences in coordinates of a point from $P$. For a line separating $N$ and $B$ to be possible as in Fig. 1, it must be possible to have $G_B=G_N$ on this line; thus

$$\Delta\alpha_{BN}(dT)^2-2\Delta\beta_{BN}dT\ dH+\Delta\gamma_{BN}(dH)^2=0 \ . \tag{2.6}$$

This has a solution only if

$$(\Delta\beta_{BN})^2-(\Delta\alpha_{BN})(\Delta\gamma_{BN})\geq 0 \ . \tag{2.7}$$

The left-hand side of (2.7) can be rewritten entirely in terms of $\Delta\alpha_{BA}$, $\Delta\alpha_{AN}$, $p_1$, and $p_2$. To do this we notice that $\Delta\alpha_{BN}=\Delta\alpha_{BA}+\Delta\alpha_{AN}$ and similarly with $\alpha$ replaced by $\beta$ or $\gamma$. Equations (2.3) and (2.4) then allow us to eliminate $\Delta\beta_{AN}$, $\Delta\gamma_{AN}$, $\Delta\beta_{BA}$, and $\Delta\gamma_{BA}$ in favor of $\Delta\alpha_{AN}$, $\Delta\alpha_{BA}$, $p_1$, and $p_2$. We get

$$\left[\frac{\Delta\alpha_{BA}}{p_2}+\frac{\Delta\alpha_{AN}}{p_1}\right]^2 -(\Delta\alpha_{BA}+\Delta\alpha_{AN})\left[\frac{\Delta\alpha_{BA}}{p_2^2}+\frac{\Delta\alpha_{AN}}{p_1^2}\right] ,$$

which can be simplified to

$$-(\Delta\alpha_{BA})(\Delta\alpha_{AN})\left[\frac{1}{p_1}-\frac{1}{p_2}\right]^2 . \tag{2.8}$$

Equations (2.7) and (2.8) are consistent only if $p_1=p_2$, [recall (2.5)], in which case there is simply no phase $A$. Hence, Fig. 1 is impossible, irrespective of the order of transition of the line $BN$; i.e., the intersection $T_{c_3}$ in Fig. 2 is forbidden. The above argument can be trivially generalized to the case where there is more than one intervening phase $A_1, A_2 \ldots$, so long as all transitions $N \rightarrow A_1 \rightarrow A_2 \cdots \rightarrow B$ (in order of decreasing temperature) are second order. It can also be generalized to the

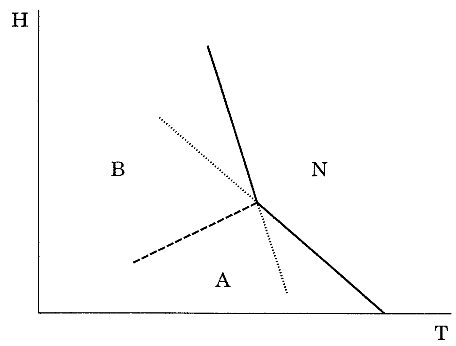


FIG. 3. An example of a bicritical point which is allowed. Line $AB$ is first order (represented by dashed lines) and must lie within the dotted lines (which are the extrapolations of $AN$ and $BN$).

point $T_{c_4}$, which is, therefore, also forbidden. As a trivial corollary, three second-order transition lines cannot meet at a point unless some other line(s) emerges from it.

In fact, the only bicritical point that is possible is the one where $N$ has second-order instabilities into the $A$ or $B$ phase, and a first-order transition line between them (see Fig. 3). Even in this case further restrictions apply: Following the reasoning in Ref. 15, but specializing to $p_1, p_2 < 0$ instead, where

$$p_1 \equiv \left[\frac{dH}{dT}\right]_{AN}, \quad p_2 \equiv \left[\frac{dH}{dT}\right]_{BN}$$

(this is more relevant to our case of superconductivity phase diagram) one can show that the phase diagram is impossible unless $|p_2| \geq |p_1|$. Furthermore, the slope of the first-order transition line satisfies[17]

$$p_3 \equiv \left[\frac{dH}{dT}\right]_{AB} = p_1 \frac{r-1}{r-y} , \tag{2.9}$$

where

$$r \equiv \left[\frac{\Delta\alpha_{AN}}{\Delta\alpha_{BN}}\right]^{1/2} , \tag{2.10}$$

$$y \equiv |p_1|/|p_2| < 1 . \tag{2.11}$$

It can be easily verified that none of the three phases can occupy more than 180° of the phase diagram.

In Tokuyasu, Hess, and Sauls[13] a tentative phase diagram for $UPt_3$ ($\mathbf{H}\|\hat{\mathbf{c}}$) was constructed based on experimental data. It should be noted that many features of their phase diagram are possible only if some suitable lines there are first-order phase-transition lines. These authors would like to explain some of the phase transitions by vortex transitions. If that is the case, then the thermodynamical results here have important implications on the nature of the vortex transitions.

We do not believe that the GL theory of Ref. 14 itself can violate thermodynamics. The problem simply lies on the incorrect guess of the solution. It is, however, out of the scope of the present paper to discuss this question.

### B. Polycritical points

Since the specific-heat experiment for $\mathbf{H}\perp\hat{\mathbf{c}}$ suggests that the two second-order transition lines emerging from the two $H=0$ transitions meet at $(H^*, T^*)$,[6] and we have shown that we cannot have only one phase-transition line emerging from this point, we now turn to the simplest possibilities, i.e., four phase-transition lines meeting at $(H^*, T^*)$ (Figs. 4 and 5). Notice that experiments suggest that the lines $AN$, $CA$, and $BN$ are probably second order. [No latent heat,[6] hystersis, nor jumps in frequencies[4] at these transitions have ever been reported.] Thus the remaining unanswered question is whether $CB$ is first or second order [provided, of course, it is not the case that yet some other phase-transition line(s) meets at $(H^*, T^*)$].

We shall first consider the case where CB is first order (Fig. 4). The thermodynamics can be studied by a simple extension of the paper by Leggett.[15] Again using similar definitions as in Sec. II A, we have, at $P \equiv (H^*, T^*)$,

$$p_1 \equiv \left[\frac{dH}{dT}\right]_{AN} = \frac{\Delta\alpha_{AN}}{\Delta\beta_{AN}} = \frac{\Delta\beta_{AN}}{\Delta\gamma_{AN}} , \tag{2.12}$$

$$p_2 \equiv \left[\frac{dH}{dT}\right]_{BN} = \frac{\Delta\alpha_{BN}}{\Delta\beta_{BN}} = \frac{\Delta\beta_{BN}}{\Delta\gamma_{BN}} , \tag{2.13}$$

$$p_3 \equiv \left[\frac{dH}{dT}\right]_{CA} = \frac{\Delta\alpha_{CA}}{\Delta\beta_{CA}} = \frac{\Delta\beta_{CA}}{\Delta\gamma_{CA}} . \tag{2.14}$$

The specific-heat jumps are positive; thus

$$\Delta\alpha_{AN}, \Delta\alpha_{BN}, \Delta\alpha_{CA} > 0 . \tag{2.15}$$

The line $CB$ is determine by the condition of equality of $G$, i.e.,

$$\Delta\alpha_{CB}(dT)^2 - 2\Delta\beta_{CB} dT\, dH + \Delta\gamma_{CB}(dH)^2 = 0 . \tag{2.16}$$

There are two roots to this quadratic equation. However, the latent heat of a first-order phase transition cannot be negative, i.e., $S_c < (>) S_B$ if $C(B)$ is the lower temperature phase [$p_4 < (>) 0$]. One can easily verify that in either case the condition can be written as, with

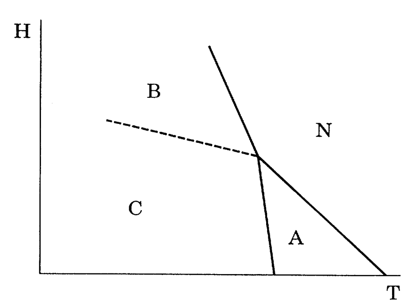


FIG. 4. An example of a polycritical point which is allowed. Here $CB$ is first order.

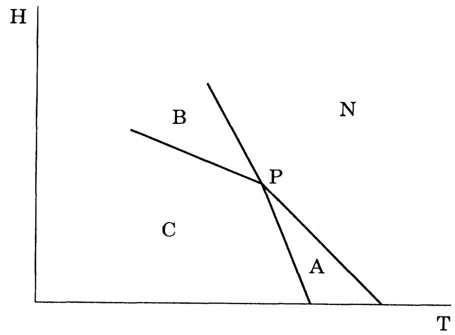


FIG. 5. An example of a polycritical point which is allowed. Here $CB$ is second order.

the help of (2.16),

$$\Delta\gamma_{CB}\left[\frac{dH}{dT}\right]_{CB} > \Delta\beta_{CB}$$

and thus

$$p_4 \equiv \left[\frac{dH}{dT}\right]_{CB} = \frac{\Delta\beta_{CB} + [(\Delta\beta_{CB})^2 - (\Delta\alpha_{CB})(\Delta\gamma_{CB})]^{1/2}}{\Delta\gamma_{CB}} . \tag{2.17}$$

The right-hand side of (2.17) can be rewritten entirely in terms of $p_1$, $p_2$, $p_3$, and specific-heat jump ratios ($\Delta\alpha$'s). To do this we use, in (2.17),

$$\Delta\alpha_{CB} = \Delta\alpha_{CA} + \Delta\alpha_{AN} - \Delta\alpha_{BN}$$

and similar relations with $\alpha$ replaced by $\beta$ or $\gamma$. We then eliminate $\Delta\beta_{CA}$, $\Delta\beta_{AN}$, $\Delta\beta_{BN}$, $\Delta\gamma_{CA}$, $\Delta\gamma_{AN}$, and $\Delta\gamma_{BN}$ in favor of $\Delta\alpha_{CA},\Delta\alpha_{AN},\Delta\alpha_{BN}$, and $p_1,p_2,p_3$ with the help of (2.12)–(2.14) [cf. the derivation of (2.8)]. We confine our case to $p_1 < 0$, which has been established experimentally. With

$$r_3 \equiv \left[\frac{\Delta\alpha_{CA}}{\Delta\alpha_{BN}}\right]^{1/2} , \tag{2.18a}$$

$$r_1 \equiv \left[\frac{\Delta\alpha_{AN}}{\Delta\alpha_{BN}}\right]^{1/2} , \tag{2.18b}$$

we obtain

$$p_4 = p_1 \frac{\left[\frac{p_1}{p_3}r_3^2 + r_1^2 - \frac{p_1}{p_2}\right] - \left[r_1^2\left[1-\frac{p_1}{p_2}\right]^2 + r_3^2\left[\frac{p_1}{p_3}-\frac{p_1}{p_2}\right]^2 - r_1^2 r_3^2\left[1-\frac{p_1}{p_3}\right]^2\right]^{1/2}}{r_3^2\frac{p_1^2}{p_3^2} + r_1^2 - \frac{p_1^2}{p_2^2}} , \tag{2.19}$$

which determines the slope of $CB$ in terms of the other slopes and specific-heat jumps.

If, further more, $CB$ is a second-order transition (Fig. 5), $p_4$ has to satisfy

$$p_4 = \frac{\Delta\alpha_{CB}}{\Delta\beta_{CB}} = \frac{\Delta\beta_{CB}}{\Delta\gamma_{CB}} , \tag{2.20}$$

that is, the expression inside the square root in (2.19) has to be zero. This can be viewed as a condition on the slope $p_3$. Assuming that $p_1,p_2 < 0$ and $|p_2| > |p_1|$, so that we have an "up-kink" at $P = (H^*, T^*)$, we find

$$1 - \frac{p_2}{p_3} = \left[\frac{p_2}{p_1} - 1\right] r_1 \frac{r_1 r_3 \pm (r_1^2 + r_3^2 - 1)^{1/2}}{(1 - r_1^2) r_3} . \tag{2.21}$$

This is possible only if $r_1^2 + r_3^2 - 1 > 0$, or, since $\Delta\alpha_{CB} = \Delta\alpha_{BN}(r_1^2 + r_3^2 - 1)$,

$$\Delta\alpha_{CB} > 0 \tag{2.22}$$

and thus $C$ must be a lower temperature phase than $B$. From the figure it is then obvious that it requires $p_4 < 0$. [Recall the remark below Eq. (2.8).]

The relation (2.21) can be rewritten as, with the shorthand

$$r_4 \equiv \left[\frac{\Delta\alpha_{CB}}{\Delta\alpha_{BN}}\right]^{1/2} = (r_1^2 + r_3^2 - 1)^{1/2} , \tag{2.23}$$

$$1 - \frac{p_1}{p_3} = \left[1 - \frac{p_1}{p_2}\right] \frac{r_3 \pm r_1 r_4}{(1 - r_1^2) r_3} . \tag{2.24}$$

Equation (2.20) [or (2.19)] then determines $p_4$. With the help of (2.24), we find

$$\frac{p_2}{p_4} - 1 = \left[\frac{p_2}{p_1} - 1\right] \frac{\mp r_1 r_3 - r_1^2 r_4}{(1 - r_1^2) r_4} . \tag{2.25}$$

Thus

$$\frac{\frac{p_2}{p_4} - 1}{1 - \frac{p_1}{p_3}} = \mp \frac{p_2}{p_1} \frac{r_1 r_3}{r_4} . \tag{2.26}$$

Recall that we assumed $p_1,p_2 < 0$ and $|p_2| > |p_1|$. From Fig. 5, it is clear that, for either sign of $p_3$, we must have $1\text{-}p_1/p_3 > 0$. Since we have seen that $p_4 < 0$, from the figure it is obvious that $|p_2| > |p_4|$ and, hence, $p_2/p_4 - 1 > 0$. Thus the lower sign of Eq. (2.26) and, hence, also of (2.24) and (2.25) must be chosen. Notice that

$$(r_3 - r_1 r_4)(r_3 + r_1 r_4) = (1 - r_1^2)(r_1^2 + r_3^2)$$

and therefore, $(r_3 - r_1 r_4)/(1 - r_1^2)$ is always positive definite, and thus the inequalities listed above are always satisfied irrespective of whether $r_1^2 \gtrless 1$. Any two of the three relations (2.24)–(2.26) will then be the two (independent) conditions relating the slopes and the specific-heat jumps [in addition to the conditions (2.22) and $p_4 < 0$]. Notice that all conditions for the existence of Fig. 5 have been included. Thus, Fig. 5 is thermodynamically *possible*, though it is clear that, if we use a Ginzburg-Landau theory, particular conditions have to be met.[18]

In the remainder of this section we would like to apply the above relations to the phase diagram of $UPt_3$ for $\mathbf{H} \perp \hat{\mathbf{c}}$. A phase diagram involving four transition lines meeting at a point has been constructed by Hasselbach *et al.*[6,7] (see Fig. 6). As noted, *AN*, *BN*, and *CA* are all second-order transitions lines, and the specific-heat jumps along them have been measured. The corresponding quantity, however, was not measured on *CB*. [In fact they saw no specific-heat jumps, nor latent heat, anywhere else in the $(H,T)$ plane they have investigated; we shall come back to this point later.] They, in fact, completed their phase diagram near $(H^*, T^*)$ by taking a "transition line" measured by ultrasonic attenuation[2] (the position of which is represented by the dotted line in Fig. 6). Thus, it is open whether this transition is first or second order. We shall discuss both of these two possibilities below.

The specific-heat jumps for $H < H^*$ reported in Ref. 6 have rather large nonmonotonic variations as a function of field, perhaps due to difficulties in resolving the contributions from the two transitions. Thus, it is difficult to obtain reliable extrapolated values for $r_1$ and $r_3$. (In fact, it seems that the data of Ref. 6 lie in a thermodynamically forbidden region; see Fig. 7.) Therefore, we assume that the slopes *AN*, *CA*, *BN* are correctly given in Ref. 6 (i.e., $p_1/p_3 = 0.533$, $p_1/p_2 = 0.625$; also notice the difference in nomenclature for *B* and *C* phases) and we determine the specific-heat jumps that will account for the phase diagram. If we assume that *CB* is first order, then Eq. (2.19) gives its slope as a function of the ratios of the specific-heat jumps $r_1, r_3$. The case where *CB* is second order corresponds to the special case where the expression $(E)$ under the square root vanishes. In Fig. 7 we plot the contours of the ratio $|p_4|/|p_1|$ as well as the locus (dashed line) where $E$ vanishes, for a particular portion of the $(r_1, r_3)$ plane. Above the dash-dotted line the phase diagram as in Figs. 4 or 5 will be forbidden.

If we choose the line *CB* as in Ref. 6, then the value $|p_4|/|p_1| \approx 0.15$. As is clear from the figure, we need a value of $r_1$ and $r_3$ smaller than 0.7 and 0.82 (specific-heat jump ratio = 0.49 and 0.67) to explain the observed slope. These values of $r_1$ and $r_3$ are *much* smaller than the ones reported in Ref. 6 [where they are close to 1, notice that the 2 (1) there is the upper (lower) temperature transition, and thus corresponds to 1 (3) here].

Thus, the assumption that a line *CB* of slope as low as $|p_4|/|p_1| \sim 0.15$ connects to the point $(H^*, T^*)$ is incompatible with the measured specific heat. Notice that the ultrasonic attenuation experiment[2b] seems to indicate a line which is almost flat only at low temperatures, but with increasing $|p_4|$ as temperature increases. Thus, we have seen that this increase in $|p_4|$ is necessary for compatibility with the assumption that the three other second-order transition lines are meeting at a point. Since Ref. 6 was unable to detect the line *CB*, the latent heat or specific-heat jump along *CB* must be small; it must be weakly first- or second-order transition with a small specific-heat jump. In the former case no sudden sharp changes in specific heat is reported either. Thus

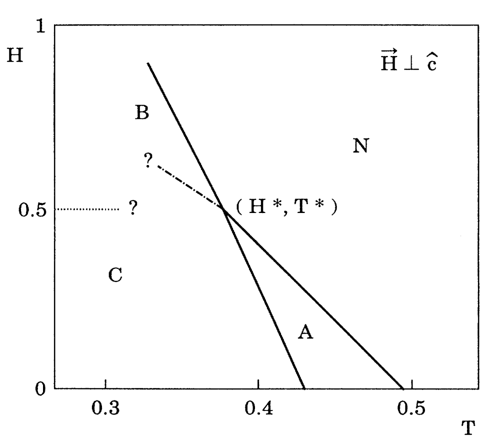


FIG. 6. A probable phase diagram of $UPt_3$ for $\mathbf{H} \perp \hat{\mathbf{c}}$. The nature of the dotted line is uncertain. It may or may not join the dash-dotted line.

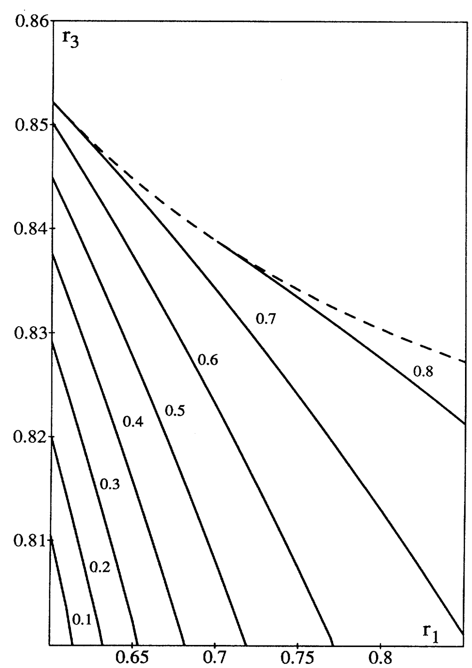


FIG. 7. The ratio of $|p_4|/|p_1|$ for Figs. 4 and 5 as a function of the specific-heat jumps ratios $r_1$ and $r_3$, when the other slopes have values as in the text.

$\alpha_C \approx \alpha_B$. One thus must lie near the dash-dotted line, as well as near $r_1^2+r_3^2 \approx 1$ in Fig. 7. In the latter case one must lie on the dash-dotted line, and $r_1^2+r_3^2 \gtrsim 1$. Reference 6 seems to indicate $r_1, r_3$ that are too large to be acceptable. Anyway, more careful measurement of $|p_4|$ as one approaches $H_{c_2}$, and the value of the specific-heat jumps near the "kink" of the $H_{c_2}$ curve is highly desirable to determine whether one really has four transition lines meeting at a point as in Fig. 4 or 5. This information will be useful in distinguishing between candidates of Ginzburg-Landau theories.

We would like to end this section by remarking that since our results are entirely based on thermodynamics, they are completely general and thus applicable to a variety of other systems. We shall, however, defer these discussions to a later publication.

## III. CONCLUSIONS

We summarize as follows. We have considered the implications of general thermodynamics on bicritical and polycritical points, and applied the relations to the phase diagram of $UPt_3$. Assuming that, for $\mathbf{H} \perp \hat{\mathbf{c}}$, the two $H=0$ transitions remain second order and meet at a point at finite $H$, as suggested by the specific-heat measurement,[6] we find that at least two other phase-transition lines must emerge from that point. Assuming that one of these lines is also second order, we have considered the thermodynamic implications on the slopes and specific-heat jumps near the intersection point.

Given the extrapolation of experimental data needed to deduce the phase diagram, and also the potential quantitative difficulty in the measured specific heat mentioned above, one cannot exclude the possibility that, for $\mathbf{H} \perp \hat{\mathbf{c}}$, the suggestion that the lines $AN$ and $CA$ meet is actually in error. In that case the phase diagram may simply be as in Fig. 8, where there is only a smooth crossover from the low-$H$–high-$T$ phase to the high-$H$–low-$T$ phase.[10]

The specific-heat experiment, in particular the extrapolated values of the ratios of the specific-heat jumps at the hypothetical intersection point of the phase-transition lines, will be crucial in distinguishing the above two possibilities.

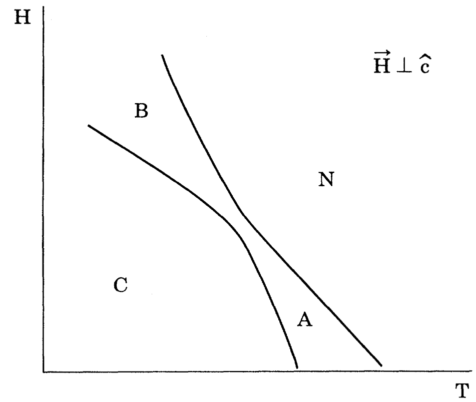


FIG. 8. Another possible phase diagram for $H$ in a general direction in the $a$-$b$ plane. There is no real phase transition between $A$ and $B$. Possible extra transition lines within the $C$ phase are not indicated.

*Note added in proof.* Very recently two experiments have been reported [G. Bruls *et al.*, Phys. Rev. Lett. **65**, 2294 (1990); S. Adenwalla *et al.*, *ibid.* **65**, 2298 (1990)] showing that a polycritical point as in Fig. 6 is indeed involved for all orientations of the magnetic field.

## ACKNOWLEDGMENTS

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