Structure of the superconducting phases of UBe_{13} and $U_{1-x}Th_xBe_{13}$

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Within the framework of the Landau theory of phase transitions, we investigate the types of phase changes which can occur between superconducting phases of the heavy-fermion compounds $U_{1-x}Th_xBe_{13}$. We show that none of these phase changes agree with the experimentally-observed phase diagram (x,T_c), in which there is one second-order phase transition in the region $x<1.75\%$ from the normal to the superconducting state and two successive second-order phase transitions for $x>1.75\%$. The overall properties of $U_{1-x}Th_xBe_{13}$ argue in favor of the following two scenarios, each of which involves a sequence of phase transitions that comes closest to the experimentally established sequence. (1) For $x>1.75\%$ a second-order phase transition occurs from a normal to a superconducting phase with symmetry $O(T)\times\tilde{R}$; then, as the temperature decreases, a first-order phase transition occurs to a superconducting phase with symmetry $D_3(C_3)\times\tilde{R}$. For $x<1.75\%$, a phase transition of second order occurs from the normal metal to a superconductor with symmetry $D_3(C_3)\times\tilde{R}$. (2) In the range $x>1.75\%$ a second-order transition occurs from the normal metal to a superconductor with symmetry $O(D_2)$, followed by a first-order transition to a superconductor with symmetry $D_3(E)$; for $x<1.75\%$, we have a second-order phase transition from the normal metal to a superconductor with symmetry $D_3(E)$.

1. INTRODUCTION

Although about five years have passed since the discovery of superconductivity in uranium heavy-fermion compounds, the nature of the superconducting and normal states in these compounds remains unclear (see the review Ref. 1). What is lacking is an experimental method which would allow the superconducting phases to be identified unambiguously (i.e., the way unclear magnetic resonance methods were used to investigate the superfluid phases of He^3). Study of the (x,T_c) phase diagram of the superconducting compounds $U_{1-x}Th_xBe_{13}$, which are formed by replacing atoms of uranium in UBe_{13} by thorium, provides important information of this sort. These compounds exhibit two phase transitions in the range of concentrations $1.75\% < x < 6\%$[2–6] (the lines T_{ca} and T_{cb} in Fig. 1): a transition of T_{ca} to a superconducting state, and an additional second-order phase transition at a lower temperature T_{cb}.

Quite a few attempts have been made to explain the mechanism of these transitions.[5,7–13] In this paper we will adopt the most controversial point of view, according to which the transition of $T=T_{cb}$ is a transition between two superconducting phases. Our arguments are based on the following experimental facts: (1) Inversion of the magnetic field dependence of the lower transition temperature $T_{cb}(H)$ gives rise to a plot[3] of the function $H(T_{cb})$ which resembles the temperature dependence of the upper critical field $H_{c2b}(T)$. (2) There is a sudden change of slope in the dependence of the lower critical field on temperature at the transition point T_{cb} (Ref. 4). (3) The line T_{cb} in the region $x>1.75\%$ appears to be a continuation of the line T_c in the region $x<1.75\%$ (Ref. 11, 14). Anomalous ultrasonic absorption has been observed in the vicinity of this line (in contrast to the line T_{ca}).[5,6,15] (4) Under pressure, the minimum in the curve $T_c(x)$ becomes deeper and shifts to the region of larger concentrations, until at a pressure $P=10$ kbar superconductivity disappears entirely in the neighborhood of $x\approx 3\%$ even at $T=0$. At this pressure, the material is superconducting only for concentrations less than this.[14] The behavior of T_c as a function of pressure[14] and concentration of impurities of other elements[16] for $x<x_m$ differs qualitatively from its behavior for $x>x_m$, which leads us to believe that near T_c these composition regions correspond to different superconducting phases.

Several possible explanations for this kind of behavior have been proposed in the literature,[7,8,12,13] The authors of Ref. 7 claimed that the transition of T_{cb} was a transition between an isotropic (due to impurities) and an anisotropic superconducting phase. However, these authors also predicted that the splitting of the lines T_{ca} and T_{cb} had to start $x=0$, which does not agree with experiment (Fig. 1). In Ref. 8 the phase transition of $T=T_{cb}$ was interpreted to be a transition between a superconducting phase with cubic symmetry and a superconducting phase with a tetragonal distortion, and the anomalous ultrasonic absorption near T_{cb} was associated with oscillations of domain walls separating superconducting domains with different orientations of the tetragonal anisotropy axes. However, one of the predictions of this reference was the absence of anomalous ultrasonic absorption near T_{cb} along the $\langle 111\rangle$ direction, which again is not confirmed by experiment.[6] In Refs. 12,13 a phenomenological description of the phase change was proposed in terms of a mixture of two superconducting phases possessing different symmetries. Some remarks on these last two references will be given in the text of this article.

Apparently, there is also experimental evidence for an alternative point of view, in which the curve T_{cb} corresponds to an antiferromagnetic transition (Ref. 5, 9).[1] For instance, the results of experiments on muon spin resonance[17] point to the presence to $T<T_{cb}$ of a very small magnetic moment of $10^{-3}\,\mu_B$ associated with the U atoms. However, the presence of this moment is not confirmed by neutron scattering and NMR measurements.[18]

In this article, we will thoroughly analyze the $U_{1-x}Th_xBe_{13}$ phase diagram in the spirit of the Landau theory of phase transitions, assuming that the curve T_{cb} is a line of transitions between two superconducting states. The

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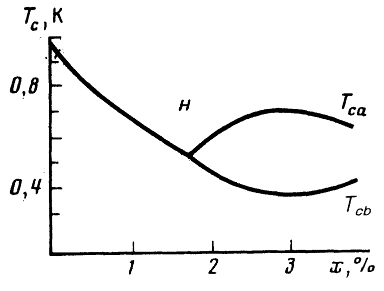

FIG. 1. Schematic (x, T_c) phase diagram for the phases of $U_{1-x}Th_xBe_{13}$ (n is the normal state).

results of our analysis show that none of the possible scenarios based on this picture—including the ones investigated in Refs. 12, 13—can lead to the phase diagram shown in Fig. 1. If the transition to the superconducting state proceeds from a nonmagnetic normal phase (Sec. 2 and the Appendix), then either the line T_{cb} must be a line of second-order transitions (see Sec. 3), or there is at least one other second-order phase transition line (or even one of first order) in addition to the second-order phase transition line T_{cb} emerging from the point where the curves T_{ca} and T_{cb} split off (see Ref. 4). The possibility of a transition to a superconducting state from a magnetic normal state (Sec. 3) also leads to phase diagrams which are more complex than the diagram in Fig. 1. The general conclusion of this paper is that it is not possible to explain the phase diagram of $U_{1-x}Th_xBe_{13}$ unambiguously at the present time. Any such explanation would require, e.g., additional measurements of the behavior of the transition lines for large concentrations of thorium, a test for the presence or absence of magnetism for $T < T_{cb}$, a search for possible additional phase transition lines, a more careful test to confirm that the line T_{cb} corresponds to a second-order phase transition, etc.

In this paper we predict and discuss the properties of certain superconducting phases and compare them with the experimental data (Sec. 4) to determine which is most preferable. When we do this, we eliminate all but two scenarios. Each of these corresponds to a sequence of phase transitions which occur successively as the temperature decreases, and each resembles closely what is shown in Fig. 1. According to the first of these scenarios, for $x > 1.75\%$ a second-order phase transition occurs from the normal phase to a superconducting phase with symmetry $O(T) \times R$, followed by a first-order phase transition to another superconducting phase with symmetry $D_3(C_3) \times \tilde{R}$; for $x < 1.75\%$ a second-order phase transition occurs from a normal metal to a superconductor with symmetry $D_3(C_3) \times \tilde{R}$. According to the second scenario, for $x > 1.75\%$ a second-order phase transition occurs from a normal metal to a superconductor with symmetry $O(D_2)$, followed by a first-order phase transition to another superconducting phase with symmetry $D_3(E)$; for $x < 1.75\%$ a second-order phase transition occurs from a normal metal to a superconductor with symmetry $D_3(E)$. The difference between the phase diagram predicted by these scenarios and the one shown in Fig. 1 is that in the former the line T_{cb} is a line of first-order transitions. Observation of a latent heat at this transition would put everything in its place, because the other properties of these superconducting phases do not contradict the experimentally-observed properties of the phases of $U_{1-x}Th_xBe_{13}$, and their phase diagram (see Fig. 3) do not contain any additional phase transition curves emerging from the point where the curves T_{ca} and T_{cb} split off.

2. TRANSITION BETWEEN SUPERCONDUCTING PHASES ARISING FROM A NONMAGNETIC NORMAL STATE

In this section, we will investigate various scenarios in which the transition corresponding to the line T_{cb} in Fig. 1 is between superconducting phases which arise directly from a normal nonmagnetic state. It is well known[19] that in the case of a strong spin-orbit interaction the order parameter of the superconducting state can be expanded in terms of basis functions from one or several of the irreducible representations of the symmetry group of the crystal. For UBe_{13}, this means the following representations of the cubic group O: two one-dimensional representations A_1 and A_2, a two-dimensional representation E, and two three–dimensional representations F_1 and F_2. Each irreducible representation has its characteristic electron-electron pairing interaction constant, and consequently its own superconducting transition temperature. Therefore, the phase transition occurs from the normal state to a superconducting state with an order parameter Δ which transforms according to only one of the irreducible representations of the group O, i.e., the one characterized by the highest critical temperature. The expansion coefficients of the order parameter in terms of basis functions of this representation are determined by minimizing the corresponding Ginzburg-Landau functional (GLF). In this case the order parameter is found to be invariant relative to a certain subgroup H of the "symmetry group of physical laws" $G = O \times R \times U(1)$ (R is the time-reversal operator, while $U(1)$ is the gauge transformation group).

If, as happens in $U_{1-x}Th_xBe_{13}$ in the region $x > 1.75\%$, a second phase transition occurs to another superconducting phase as the temperature decreases, then three cases are possible:

A. The transition proceeds within the framework of a single representation from one minimum of the GLF to another, and is due to temperature variation of the coefficients in the GLF expanded to terms of fourth order. Because the symmetry groups of all the minima of the GLF which are related to any given representation are not subgroups of one another,[19] a phase transition between two superconducting states within the framework of a single representation is always a first-order transition. This case is precisely analogous to that of the phase transition between the A and B phases of He^3 (Ref. 20), and, just as for helium, the line of first-order phase transitions T_b must end on a smooth curve T_c (see Fig. 2). This is not in agreement with the experimental phase diagram, for which the curve T_c has a sharp kink at the point where the curve T_b ends (Fig. 1).

B. The phase transition occurs because of mixing of the principal phase $\hat{\Delta}_a$, which transforms according to the irreducible representation $\hat{\Gamma}_a$ of the group O, with a phase $\hat{\Delta}_b$ which transforms according to another irreducible representation $\hat{\Gamma}_b$ and which possesses a critical temperature $T_{cb}(x) < T_{ca}(x)$ for $x > 1.75\%$. The symmetry groups of these states are respectively H_a and H_b. The possibility that such a transition could explain the experimental phase diagram (Fig. 1) was analyzed in Ref. 13, in which it was point-

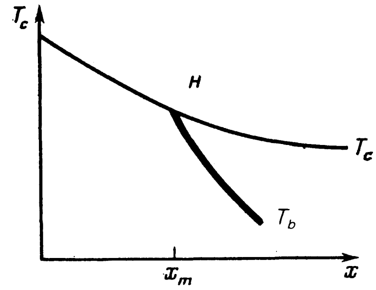

FIG. 2. Phase diagram of phase corresponding to a single representation. T_b is a line of first-order phase transitions.

ed out that a necessary condition for the occurrence of such a phase transition is that

$$H_b \subset H_a \subset G, \tag{1}$$

which assumes in particular that the states $\hat{\Delta}_a$ and $\hat{\Delta}_b$ have the same spatial parity. According to this subordination scheme, which follows from the theory of second-order phase transitions, two phase transitions will occur for $x > x_m = 1.75\%$ (in this composition range we have assumed that $T_{ca} > T_{cb}$): the first to the phase $\hat{\Delta}_a$ and the second to the phase $\hat{\Delta}_a + \hat{\Delta}_b$. For $x < x_m$, within which composition range $T_{ca} < T_{cb}$, there is one transition to the phase $\hat{\Delta}_a + \hat{\Delta}_b$; in the region $x < x_m$ near $T_{ca}(x)$ we should observe[12] the insignificant increase in specific heat $C(T)$ mentioned in Ref. 11. Sorting out all possible pairs of superconducting states arising directly through a transition from a normal nomagnetic state, and including the conditions $\hat{T}_a \neq \hat{T}_b$, $H_b \subset H_a$, we arrive at four possibilities in all, which are listed in Table I.

The condition (1), while necessary, does not yet guarantee that the phase diagrams for the pairs of phases listed in the table will coincide with the experimentally-observed phase diagram shown in Fig. 1. In the Appendix we have carried out an investigation of the problem of minimizing the GLF to find the phase diagram corresponding to an order parameter which is in the form of a linear combination of basis functions from two irreducible representations, for the example of a mixture of the representations A_2 and F_1. Although such an analysis would differ in detail from the one given in the Appendix if carried out for the other pairs of representations given in Table I, it would lead to qualitatively similar results for all the pairs. We are also led to the same results from general symmetry considerations; let us now turn to a discussion of these.

Following Ref. 13 verbatim, let us attempt to construct a phase diagram in which a second-order phase transition occurs for $x < x_m$ from the normal state to the superconducting state with symmetry H_b, while for $x > x_m$ the transition is from the normal to a superconducting state with symmetry H_a (where $H_b \subset H_a$). We will assume that there is only one transition between these phases; this is possible, e.g., in the case of a mixture of A_2 and F_1 for $\gamma_3 < 0$ (see Appendix). In order to clarify what the order of this transition will be, we must investigate the factor group H_a/H_b, i.e., the broken-symmetry group associated with a transition from a state with symmetry H_a to a state with symmetry H_b. The factor groups H_a/H_b, together with their irreducible representations $\hat{T}(H_a/H_b)$, are listed in Table I. The phase transition under study here is a first-order transition if we can construct a third-order invariant out of the basis functions of all the nonunique irreducible representations $\hat{T}(H_a/H_b)$ (because they have identical transition temperatures). It is important that during this transition no breaking of gauge symmetry or time-reversal symmetry occur; this ensures that the third-order invariant will be real. The groups $D_2(C_2)$ and $D_2(E)$ are isomorphic to D_2; therefore the representations of all three groups coincide. The third-order invariant for the group D_2 is xyz, where x,y,z are basis functions for the one-dimensional representations B_1, B_2, B_3; for the group C_3 this invariant equals $e_+^3 + e_-^3$, where $e_\pm = x \pm iy$ are the basis functions for the one-dimensional representations Γ_+ and Γ_-. Thus, if only one transition can occur between phases with symmetries H_a and H_b, this transition is necessarily first-order.

It is interesting to note that the same conclusions were reached by the authors of Ref. 12, who investigated the case where a phase with s-pairing was chosen as the $\hat{\Delta}_a$ phase, i.e., $\hat{\Delta}_a = i\hat{\sigma}_y\Delta_a$, while a polar phase with d-pairing was chosen as the $\hat{\Delta}_b$ phase, i.e., $\hat{\Delta}_b = [(\hat{\mathbf{k}}\hat{\mathbf{l}})^2 - 1/3]i\hat{\sigma}_y\Delta_b$ (here, σ_y is a Pauli matrix). That is, if there is only one transition between these phases, then it surely is a first-order transition. Actually, the phase with s-pairing is invariant relative to three-dimensional rotations, so its symmetry group $H_a = SO_3$. The polar phase with d-pairing is symmetric relative to one-dimensional rotations around the mirror anisotropy axis $\hat{\mathbf{l}}$, and its symmetry group $H_b = S_\infty$. The result of the factorization H_a/H_b is not even a group, but is rather a factor space $SO_3/S_\infty = RP^2$, i.e., the real projective plane. Thus, the symmetry which is broken by this transition is determined by RP^2; this is like the transition from an isotropic liquid to a nematic liquid crystal, which, as is well known,[22] is a first-order transition thanks to the presence of a single third-order invariant.

Thus, in the case under study here, the phase diagram will have the form shown in Fig. 3 (see also the Appendix). For various reasons, a first-order phase transition may differ only weakly from a second-order transition, in which case

TABLE I.

№	H_a	H_b	$\hat{T}_a$	$\hat{T}_b$	H_a/H_b	$\hat{T}(H_a/H_b)$
1	$O(T)\times\tilde{R}$	$D_3(C_3)\times\tilde{R}$	A_2	F_1	$D_2(C_2)$	A, B_1, B_2, B_3
2	$O(D_2)$	$D_3(E)$	E	F_1, F_2	$D_2(E)$	A, B_1, B_2, B_3
3	$O\times\tilde{R}$	$D_3\times\tilde{R}$	A_1	F_2	D_2	A, B_1, B_2, B_3
4	$O\times\tilde{R}$	$D_4\times\tilde{R}$	A_1	E	C_3	A, Γ_+, Γ_-

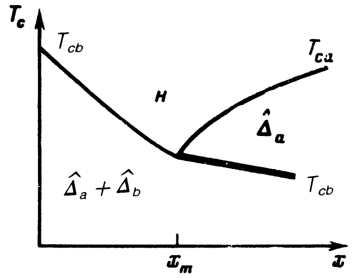

FIG. 3. Phase diagram for the phases $\hat{\Delta}_a$ and $\hat{\Delta}_b$ corresponding to different representations. T_{cb} in the region $x > x_m$ is a line of first-order phase transitions between superconducting phases.

the phase diagram shown in Fig. 3 corresponds to the phase diagram shown in Fig. 1.

Let us now drop the requirement that only one transition occurs between the phases with symmetries H_a and H_b. Now both transitions can be second-order. Calculations which apply to this case can be found in the Appendix for the example of a mixture of the representations A_2 and F_1. Here we will describe qualitatively the successive transitions between phases together with their symmetries. The pairs of phases 1,3,4 presented in Table I, and also the pair of phases investigated in Ref. 12, admit the following general description of the phase transition sequence.

For $x < x_m$ a second-order phase transition occurs to a phase $\hat{\Delta}_a + \hat{\Delta}_b$ with symmetry H_b (line T_{cb} in Fig. 4). In its turn, this phase converts by means of a second-order phase transition to a phase $\hat{\Delta}_a + \hat{\Delta}_b e^{i\psi}$ with symmetry H_a/R (line T_{cR} in Fig. 4). For $x > x_m$ two second-order phase transitons occur in succession to a phase $\hat{\Delta}_a$ with symmetry H_a (line T_{ca} in Fig. 4), and then to the same phase as in the case $x < x_m$, i.e., $\hat{\Delta}_a + \hat{\Delta}_b e^{i\psi}$, with symmetry $H_b/\tilde{R}$ (line T_{cb} in Fig. 4). For phases No. 2 in the table the phase transition picture looks the same. Namely, for $x < x_m$ a phase transition (line T_{cb}) occurs to the phase $\hat{\Delta}_a + \hat{\Delta}_b$ with symmetry $H_b = D_3(E)$, followed by a second-order phase transition (line T_{cR}) to the phase $\hat{\Delta}_a + \hat{\Delta}_b e^{i\psi}$ with symmetry $C_3(E)$, accompanied by a breaking of the symmetry relative to elements of the group $D_3(E)$ which contains the time-reversal operator R. For $x > x_m$ consecutive second-order transitions occur, first to a phase Δ_a with symmetry $H_a = O(D_2)$ (line T_{ca}), and then to a phase with symmetry $C_3(E)$ (line T_{cb}).

Turning now to case C, we note that the curve T_{cR} is not

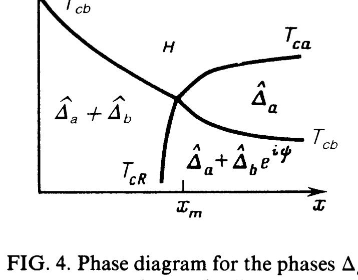

FIG. 4. Phase diagram for the phases Δ_a and Δ_b corresponding to different representations. The lines T_{cb} in the region $x > x_m$ and T_{cR} in the region $x < x_m$ are lines of second-order phase transitions between superconducting phases.

observed in experiment. However, this is not a problem if we assume a strong dependence of T_{cR} on concentration. Finally, there is also a scenario covered under case B in which there are two transitions with symmetries H_a and H_b, one of which is first-order and the order second-order (see the Appendix).

C. The phase transformation picture shown in Fig. 4, which contains four second-order phase transition curves, is of course possible even when the subordination condition (1) is not fulfilled. It can also be realized if the phases Δ_a and Δ_b belong to different irreducible representations $\hat{T}_a$ and $\hat{T}_b$ and plots of their critical temperatures intersect at $x = x_m$ so that $T_{ca}(x) > T_{cb}(x)$ for $x > x_m$ and $T_{cb}(x) > T_{ca}(x)$ for $x < x_m$, and if their symmetry groups $H_a \subset G$ do not satisfy condition (1). The symmetry group of the phase $\hat{\Delta}_c$ which forms as the temperature falls from phase $\hat{\Delta}_a$ for $x > x_m$ and from phase $\hat{\Delta}_b$ for $x < x_m$ is such that $H_c \subset (H_a \cap H_b)$. The number of scenarios of this kind is rather large; however, so long as none of them are observed experimentally, there is no sense in concerning ourselves with a detailed discussion of them.

3. TRANSITION BETWEEN SUPERCONDUCTING PHASES, ONE OF WHICH ARISES FROM A MAGNETIC NORMAL STATE

As we have already pointed out in the Introduction, the possibility that the transitions on the line T_{cb} are transitions to an antiferromagnetic state has been discussed in the literature.[5,9] If we adopt this point of view, then the line of antiferromagnetic transitions T_{cb}, which runs below the line of the superconducting transitions T_{ca} for $x > x_m$, must also be present in the region $x < x_m$. Although experiments do not reveal any phase transitions in this region besides the transition to the superconducting state (the curve T_c), it is noteworthy that the curve of the dependence of the resistivity $\rho(T)$ on temperature in $U_{1-x}Th_xBe_{13}$ has a maximum ($T_M = 2.5$ K for $x = 0$), and that the temperature which corresponds to this maximum decreases with increasing concentration, becoming less than T_c for x slightly less than x_m.[23] If we assume that T_M is the transition temperature to an ordered magnetic state, then we are led to the phase diagram shown in Fig. 5, where the magnetic transition curve (T_M,T_{cb}) intersects the superconducting transition curve (T_c, T_{ca}). An analogous situation is observed in UPt_3 ($T = 0.5$ K), where according to neutron scattering a transition to an antiferromagnetic state[24] is observed at $T_N = 5$ K, which correlates with a maximum in the function $\partial\rho/\partial T$ on T at the same temperature. However, no anomaly in the specific heat near this temperature is observed.

From the point of view of symmetry of the phases formed, there are many scenarios involving breaking of the symmetry $O \times R \times U(1)$ of the original normal state which correspond to this picture of the phase transitions. Let us describe, e.g., the simplest of these scenarios: for $x > x_m$ there is a transition (the line T_{ca}) to the usual superconducting phase with symmetry $O \times R$, and then to some sort of superconducting magnetic phase, e.g., with symmetry $O_R(T) = (E, 8C_3, 3C_2, 6U_2R, 6C_4R)$. For $x < x_m$ the first transition to occur is to a trivial antiferromagnetic phase with the symmetry of one of the colorless groups[(2)] $(O \times R) \times U(1)$ (we also admit transitions to a phase with the symmetry of the color group $O_R(T) \times U(1)$), followed by another phase transition to some sort of superconducting magnetic phase, e.g., to the phase mentioned above with symmetry $O_R(T)$. We will not immerse ourselves in any deeper discussion of the possibilities which arise here, in view of their large number and the absence of definite experimental indications pointing to a picture of the phase transitions resembling what is shown in Fig. 5. We note only that the phase diagram must be more complex than what is known at this time about the diagram shown in Fig. 1.

4. DISCUSSION

As we have already noted, out of all the scenarios described in this paper for the phase transition in $U_{1-x}Th_xBe_{13}$, the most plausible one is shown in Fig. 3. The only difference between this scenario and the experimental one show in in Fig. 1 is the line of transitions T_{cb}, which is found to be a line of first-order transitions. All the other scenarios include additional phase transitions; therefore, in our view, although no latent heat is observed along the transition line T_{cb}, it should be looked for, especially since the remaining properties of the phases implied by the picture in Fig. 3 do not contradict observation.

Out of the four scenarios corresponding to Fig. 3 and listed in Table I, only the first two are acceptable, since only these imply a power-law dependence of the specific heat at $T\to 0$. As pointed out in Ref. 19, the phases which are listed in Table I under Nos. 3 and 4 can have zeroes in the gaps of their excitation spectra only by virtue of some accidental circumstance, such as zeroes of some specific functions of the irreducible representations; zeroes cannot occur by reason of symmetry.

The phases listed under Nos. 1 and 2 can correspond to either singlet ($S=0$) or triplet ($S=1$) pairing. The latter case is more probable, because measurements show that the upper critical field at $T=0$ in $U_{1-x}Th_xBe_{13}$ (Ref. 26) significantly exceeds (almost by an order of magnitude) the paramagnetic limit. Among the phases which transform according to one of the representations F_1 or F_2, there is one phase (see Ref. 27) whose upper critical field is not at all subject to suppression by paramagnetism. This phase has the planar-phase structure (according to the terminology borrowed from the theory of superfluid He[3]):

$$\hat{\Delta}\sim\begin{pmatrix} k_x-ik_y & 0 \\ 0 & k_x+ik_y\end{pmatrix} f_0(\mathbf{R}),$$

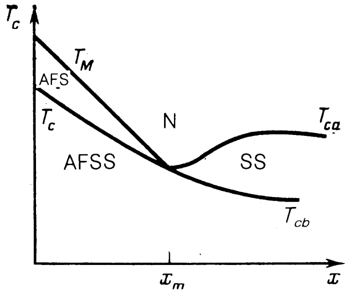

FIG. 5. Phase diagram including transitions for $x<x_m$ from the normal state to a normal antiferromagnetic state (AFS) and then to an antiferromagnetic superconducting state (AFSS). For $x>x_m$ there are transitions from the normal state to a superconducting state (SS) and then to an AFSS.

the upper critical field for this structure is a solution to the equation $\alpha^{\uparrow\uparrow}+\alpha^{\downarrow\downarrow}=2$ (see Appendix to Ref. 27). It is significant that for $T\to T_c$ the phase with the maximum field $H_{c2}(T)$ is not this phase, but the so-called Scharnberg-Klemm phase. As the temperature decreases the solution for $H_{c2}(T)$ corresponding to the latter phase is suppressed by the magnetic field because of paramagnetism, and in the high-field region is replaced by the solution corresponding to the planar phase. This behavior is not sensitive to admixture of the one-dimensional representation A_2 (No. 1) into the representation F_1, nor to admixtures of the two-dimensional representation E (No. 2) into the representations F_1 and F_2. Thus, it is not necessary to assume the presence of mixing of representations in order for the upper critical field to exceed the paramagnetic limit in triplet-pairing superconductors with strong spin-orbit interaction, as was assumed in Ref. 27; it is therefore sufficient to have phases belonging either to F_1 or F_2.

Near T_c, the upper critical fields of superconducting phases with representations F_1 and F_2 must be anisotropic. The absence of such anisotropy[26], and also the anomalously large slope of the function $H_{c2}(T)$ as $T\to T_c$ (Refs. 16, 26), apparently indicate that in the neighborhood of T_c there are strong scattering processes which suppress the superconducting correlations. Questions such as this still remain unresolved.

In discussing the properties of superconducting phases of $U_{1-x}Th_xBe_{13}$ it is pertinent to recall that according to Ref. 19, the phases $\hat{\Delta}_a$ and $\hat{\Delta}_b$ coresponding to No. 1 in Table I are nonmagnetic, while for No. 2, $\hat{\Delta}_a$ is a superconducting antiferromagnetic phase, while $\hat{\Delta}_b$ is a superconducting ferromagnetic phase. The latter scenario may possibly be relevant in light of experimental indications[17] that the phase below the curve T_{cb} possesses a small magnetic moment in the region $x>1.75\%$.

In addition to searching for a latent heat of the transition along the line T_{cb} ($x>1.75\%$) or looking with x-ray scattering for a jump in the lattice constant, an important test of the scenario proposed in this paper for the phase transitions in $U_{1-x}Th_xBe_{13}$ would be an investigation of the phase diagram in the high-concentration region. For example, the behavior illustrated in Fig. 6 would definitely argue in favor of the phase change scheme described in this paper.

In examining the experimental material, we find there are two facts which can be explained if the line T_{cb} is a line of first-order phase transitions in the region $x>1.75\%$. First of all, the coefficient of linear expansion for $T<T_{cb}$ has a deep minimum[3] in the region $x>1.75\%$, which may indicate a discontinuous change in the volume occupied by the material in the course of a first-order phase transition which is smeared out over the corresponding temperature interval. Secondly, there is the giant ultrasonic absorption near T_{cb} ($x>1.75\%$) (Refs. 5, 6), which we will pause to discuss in somewhat more detail.

According to Ref. 13, the anomalous ultrasonic absorption observed in the vicinity of the lines T_c and T_{cb} (Fig. 1) indicates a transition to a superconducting state with a multicomponent order parameter. However, the theory of ultrasonic absorption based on this point of view[29] contains an incorrect assumption about the Goldstone character of the order parameter oscillations near the transition tempera-

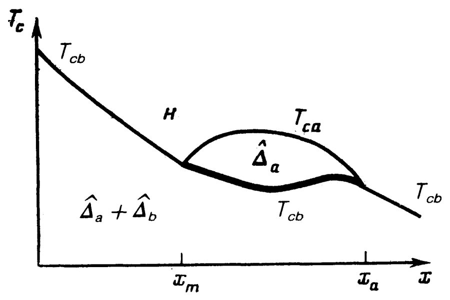

FIG. 6. A possible phase diagram for $U_{1-x}Th_xBe_{13}$ including the region of high thorium concentrations. The region of existence of the superconducting phase Δ_a ($x_m < x < x_a$) is bounded from below by the first-order transition line T_{cb}. For concentrations $x < x_m$ and $x > x_m$ the line T_{cb} is a line of second-order transitions.

ture. Actually, all collective modes in superconductors with strong spin-orbit interaction have a gap for any temperature on the order of the gap in the spectrum of elementary excitations. Apparently, the anomalous ultrasonic absorption near T_c in pure UBe_{13} is correctly described in terms of the Landau-Khalatnikov relaxation mechanism, as proposed in Ref. 30. The ultrasonic absorption near the line T_{cb} for concentrations in the range $x > 1.75\%$ differs significantly from the absorption in pure UBe_{13} (Ref. 5, 6). First of all, the intensity of the absorption peak exceeds by two orders of magnitude its value in the pure material; secondly, the dependence of the absorption coefficient on the sonic frequency is proportional to $\omega^{1-\gamma}(\gamma \ll 1)$, in contrast to the hydrodynamic dependence ($\propto \omega^2$) for pure UBe_{13}. This giant ultrasonic absorption can occur at a first-order phase transition when droplets of one superconducting phase form in the other superconducting phase; the attenuation of sound is then described within the theoretical framework developed by Isakovich[13] (see also Ref. 32) for polycrystals and emulsions. We present here some estimates corresponding to this viewpoint.

It is reasonable to assume that under these experimental conditions[5,6] the wavelength of sound $\lambda = 10^{-2}$ to 10^{-3} cm at the frequencies $\omega/2\pi = 50$–250 MHz is large compared to the size of a domain a of the droplet phase. When pinning of domain boundaries at microscopic inhomogeneities is possible, the size of a domain can be larger than the coherence length $\xi(T)$. We can estimate the penetration depth of a thermal wave $\delta \approx (\chi/\omega)^{1/2}$ [$\chi = \varkappa/C$ is the ratio of the thermal conductivity to the specific heat: $\chi(T_c) = 6\cdot 10^{-2}$ cm^2/sec—see Ref. 33] gives $\delta \sim 10^{-5}$ cm. If the size of the domain satisfies $a < \delta$, then most of the attenuation of sound takes place within the domain. In this case, the coefficient of attenuation α is $\propto \omega^2$, and its ratio to the attenuation of sound in a spatially uniform phase is:[31,32]

$$\frac{\alpha}{\alpha_n} \sim \frac{\lambda^2 a^2}{\delta^4} \frac{V_k'}{V}.$$

Here V_k'/V is the ratio of the volume occupied by the droplets of the new phase with size $a < \delta$ to the volume of the sample. For droplets with sizes $a > \delta$, most of the attenuation of sound takes place in a "skin depth" of thickness $\sim \delta$ near the boundary of the domain. The attenuation coefficient satisfies $\alpha \propto \omega^{1/2}$, and

$$\frac{\alpha}{\alpha_n} \sim \frac{\lambda^2}{a\delta} \frac{V_k''}{V},$$

where V_k'' is the volume of a droplet of the new phase with size $a > \delta$. It is not hard to see that the quantity α can exceed α_n by several orders of magnitude. The observed dependence of α on frequency ($\propto \omega^{1-\gamma}$) shows that droplets with size $a > \delta$ provide the more significant contribution to the ultraviolet attenuation.

In conclusion, we would like to thank G. E. Volovik for his active interest in this work. One of us (V.P.M.) also expresses his thanks to A. Leggett and J. Flouquet for useful discussions.

APPENDIX

We here present a full analysis of the GLF for a mixture of the representations A_2 and F_1. The order parameters which transform according to these representations, i.e., $\hat{\Delta}_a(\hat{\mathbf{k}})$ and $\hat{\Delta}_b(\hat{\mathbf{k}})$ (where $\hat{\mathbf{k}} = \mathbf{k}/k_F$) in the notation of Ref. 19, are described in the following way: in the spatially-even case ($S = 0$, i.e., singlet pairing)

$$\hat{\Delta}_a(\hat{\mathbf{k}}) = i\Delta_a \Psi_g(\hat{\mathbf{k}})\hat{\sigma}_y, \quad \hat{\Delta}_b(\hat{\mathbf{k}}) = i\sum_{i=1}^{3} \eta_i \Phi_g^{(i)}(\hat{\mathbf{k}})\hat{\sigma}_y;$$

in the spatially-odd case ($S = 1$, i.e., triplet pairing)

$$\hat{\Delta}_a(\hat{\mathbf{k}}) = i\Delta_a(\hat{\boldsymbol{\sigma}}\boldsymbol{\Psi}_u(\hat{\mathbf{k}}))\hat{\sigma}_y, \quad \hat{\Delta}_b(\hat{\mathbf{k}}) = i\sum_{i=1}^{3} \eta_i(\hat{\boldsymbol{\sigma}}\boldsymbol{\Phi}_u^{(i)}(\hat{\mathbf{k}}))\hat{\sigma}_y.$$

Here, $\Psi_g(\hat{\mathbf{k}})$, $\Psi_u(\hat{\mathbf{k}})$ are respectively the spatially-even and spatially-odd basis functions for the representation A_2; analogously, $\Phi_g^{(i)}(\mathbf{k})$ and $\Phi_u^{(i)}(\mathbf{k})$ ($i = 1,2,3$) are spatially-even and spatially-odd basis functions for the three-dimensional representation F_1; $\hat{\sigma}$ are Pauli matrices. In order to find the complex amplitudes Δ_a and η_i, it is necessary to minimize a GLF of the form,

$$F = F_a + F_b + F_{ab}. \tag{2}$$

Here F_a and F_b are functionals of $\hat{\Delta}_a$ and $\hat{\Delta}_b$ (see Ref. 19), while F_{ab} is a term corresponding to the interaction of these representations:

$$F_a = \alpha_a \Delta_a^2 + \beta_a \Delta_a^4, \tag{3}$$

$$F_b = \alpha_b |\boldsymbol{\eta}|^2 + \beta_1 |\boldsymbol{\eta}|^4 + \beta_2 |\boldsymbol{\eta}^2|^2 + \beta_3(|\eta_1|^4 + |\eta_2|^4 + |\eta_3|^4), \tag{4}$$

$$F_{ab} = \gamma_1(\eta_1^*\eta_2^*\eta_3 + \eta_1^*\eta_2\eta_3^* + \eta_1\eta_2^*\eta_3^*)\Delta_a + \gamma_2|\boldsymbol{\eta}|^2\Delta_a^2 + \gamma_3(\boldsymbol{\eta}^*\boldsymbol{\eta}^*)\Delta_a^2 + \text{c.c.} \tag{5}$$

In Eqs. (3)–(5) we introduce the notation:

$$\alpha_a = a_a[1 - T/T_{ca}(x)], \quad \alpha_b = a_b[1 - T/T_{cb}(x)];$$

$T_{ca} > T_{cb}$ for $x > x_m$, and $T_{ca} < T_{cb}$ for $x < x_m$. The values of $\beta_1, \beta_2, \beta_3$ were chosen in such a way that stability is guaranteed for the phase $\hat{\Delta}_b(\hat{\mathbf{k}})$ with symmetry $H_b = D_3(C_3) \times \tilde{R}$ in the absence of the phase $\hat{\Delta}_a(\hat{\mathbf{k}})$ (in particular, $\beta_2 < 0$; see Ref. 19). Since the choice of the overall phase of the order parameter is not significant, we will assume that the coefficient Δ_a is real.

For further exposition, it is convenient to cast η_i in the form

$\eta_i = \Delta_b n_i e^{i\psi_i}$.

Here $\Delta_b = |\eta|$, n_i are the components of a unit vector. Expressing (4), (5) in this notation, we obtain

$$F_b = \alpha_b\Delta_b^2 + \beta_1\Delta_b^4 + (\beta_2+\beta_3)\Delta_b^4(n_1^4+n_2^4+n_3^4) + 2\beta_2\Delta_b^4\{n_1^2n_2^2\cos[2(\psi_1-\psi_2)] + n_2^2n_3^2\cos[2(\psi_2-\psi_3)] + n_1^2n_3^2\cos[2(\psi_1-\psi_3)]\}, \quad (4')$$

$$F_{ab} = 2\gamma_1\Delta_a\Delta_b^3 n_1n_2n_3[\cos(\psi_1+\psi_2-\psi_3) + \cos(\psi_2+\psi_3-\psi_1) + \cos(\psi_1+\psi_3-\psi_2)] + 2\gamma_2\Delta_a^2\Delta_b^2 + 2\gamma_3\Delta_a^2\Delta_b^2(n_1^2\cos 2\psi_1 + n_2^2\cos 2\psi_2 + n_3^2\cos 2\psi_3). \quad (5')$$

It is clear from equations obtained by variation of the functional (2)–(5) in the region $x<x_m$ that for temperatures $T_{cb}(x) \gtrsim T > T_{ca}(x)$ the phase $\hat{\Delta}_b(\hat{\mathbf{k}})$, for which $|n_1| = |n_2| = |n_3| = 3^{1/2}$, $\psi_i = 0$, $\Delta_b^2 \propto (1 - T/T_{cb})$, has some phase $\hat{\Delta}_a(\hat{\mathbf{k}})$ mixed into it; the amplitude of this admixture satisfies $\Delta_a^2 \propto (1 - T/T_{cb})^3$ (these results are derived in direct analogy with those obtained in Ref. 12). This perturbation possesses symmetry $H_a = O(T)\times\widetilde{R}$. The symmetry of the general solution $\hat{\Delta}_a + \hat{\Delta}_b$, which corresponds to an extremum of the free energy, is left unchanged by transformations of the group $H_b = D_3(C_3)\times\widetilde{R}$, because $H_b \subset H_a$. Furthermore, because for $T \lesssim T_{cb}$ the inequality $\Delta_a \ll \Delta_b$ is fulfilled, the functional $F_a + F_{ab}$ is small compared to the functional F_b, and the solution $\hat{\Delta}_a + \hat{\Delta}_b$ obviously remains a minimum point. However, the first term in (5′) partially lifts the eightfold degeneracy connected with the choice of signs of the n_i; for $\gamma_1 < 0$ the phases with values of $3^{1/2}(n_1,n_2,n_3)$ equal to $(1,1,1)$, $(1,-1,-1)$, $(-1,1,-1)$, and $(-1,-1,1)$ are energetically favored, while for $\gamma_1 > 0$ the favored phases are those with $3^{1/2}(n_1,n_2,n_3)$ equal to $(-1,-1,-1)$, $(-1,1,1)$, $(1,-1,1)$, and $(1,1,-1)$.

We now investigate the concentration region $x > x_m$ for temperatures $T < T_{cb}(x) < T_{ca}(x)$, in which the phase $\hat{\Delta}_b(\hat{\mathbf{k}})$ with symmetry $H_b = D_3(C_3)\times\widetilde{R}$, which transforms according to the representation F_1, appears against a background of the phase $\hat{\Delta}_a(\hat{\mathbf{k}})$, which transforms according to the representation A_2 $(H_a = O(T)\times\widetilde{R})$. Here an important role is played by the sign of the coefficient γ_3 when the GLF is minimized.

1. Let $\gamma_3 < 0$. It is clear that the minimum of the GLF is attained for $\psi_1 = \psi_2 = \psi_3 = 0$ and, depending on the sign of γ_1, for the same values of n as the ones listed above for the region $x<x_m$. Substituting these values of $\psi_i = 0$ and $|n_i| = 3^{1/2}$ in the functional (2)–(5), we obtain

$$F = \alpha_a\Delta_a^2 + \alpha_b\Delta_b^2 + \beta_a\Delta_a^4 + (\beta_1+\beta_2+\beta_3/3)\Delta_b^4 + (2/\sqrt{3})\gamma_1\Delta_a\Delta_b^3 + 2(\gamma_2+\gamma_3)\Delta_a^2\Delta_b^2. \quad (6)$$

Minimizing (6) with respect to Δ_a, Δ_b, we find that the phase transition which takes us from the phase with order parameter $\hat{\Delta}_a(\hat{\mathbf{k}}) + \hat{\Delta}_b(\hat{\mathbf{k}})$ to the phase with order parameter $\hat{\Delta}_a(\hat{\mathbf{k}})$ is first-order. This result corresponds to the first-order phase transition investigated in Sec. 2, paragraph B for a symmetry change $O(T)\times\widetilde{R} \to D_3(C_3)\times\widetilde{R}$, which is illustrated by the phase diagram in Fig. 3.

2. For the case $\gamma_3 > 0$, the picture of the phase transitions is somewhat more complex. A further illustration is constructed using the following logical argument. Having guessed that the phases ψ_i take on the general value $\psi_1 = \psi_2 = \psi_3 = \psi$, we will investigate under what conditions these values of the phases correspond to a minimum of the GLF (2)–(5); we will then investigate the existence of other minima.

Thus, setting $\psi_1 = \psi_2 = \psi_3 = \psi$ and minimizing (4′), (5′), we find

$$\cos\psi = -\frac{3}{4}\frac{\gamma_1}{\gamma_3}\frac{\Delta_b}{\Delta_a}n_1n_2n_3, \quad (7)$$

only if

$$\left|\frac{3}{4}\frac{\gamma_1}{\gamma_3}\frac{\Delta_b}{\Delta_a}n_1n_2n_3\right| < 1,$$

which is always valid near $T_{cb}(x)$. Substituting (7) into (2)–(5), we obtain

$$F = \alpha_a\Delta_a^2 + \alpha_b\Delta_b^2 + \beta_a\Delta_a^4 + (\beta_1+\beta_2)\Delta_b^4 + \beta_3\Delta_b^4(n_1^4+n_2^4+n_3^4) - (9\gamma_1^2/4\gamma_3)\Delta_b^4 \times (n_1n_2n_3)^2 + 2(\gamma_2-\gamma_3)\Delta_b^2\Delta_a^2. \quad (8)$$

A further minimization of (8) with respect to $\mathbf{n}$ gives the solution $|n_1| = |n_2| = |n_3| = 3^{-1/2}$ for $\beta_3 > -\gamma_1/2\gamma_3$ (recall that $\gamma_3 > 0$). Investigating first the region $\beta_3 > -\gamma_1^2/2\gamma_3$, we finally obtain a functional of the form

$$F = \alpha_a\Delta_a^2 + \alpha_b\Delta_b^2 + \beta_a\Delta_a^4 + (\beta_1+\beta_2+\beta_3/3 - \gamma_1^2/12\gamma_3)\Delta_b^4 + 2(\gamma_2-\gamma_3)\Delta_b^2\Delta_a^2, \quad (9)$$

from which it follows that a second-order phase transition occurs with the appearance of an amplitude Δ_b for

$$T^* = [a_b\beta_a T_{cb} - (\gamma_2-\gamma_3)a_a T_{ca}][a_b\beta_a - (\gamma_2-\gamma_3)a_a]^{-1}.$$

We now prove that the solution presented above is actually a minimum. In fact, when we substitute $\psi_1 = \psi_2 = \psi_3 = \psi$ and minimize the GLF with respect to ψ and $\mathbf{n}$, w obtain a solution with $|n_1| = |n_2| = |n_3| = 3^{-1/2}$. Conversely, setting $|n_1| = |n_3| = 3^{-1/2}$, we obtain the solution $\psi_1 = \psi_2 = \psi_3 = \psi$ which minimizes the functional (2)–(5). these conditions are sufficient to ensure that the solution is a minimum, because by carrying out this two-stage minimization of the GLF twice, the second time with the stages reversed, we have in fact subjected the GLF minimum to a test with respect to stability relative to all collective oscillation modes taken separately. In the case under discussion these modes are a real oscillatory mode $\mathbf{n}$ for fixed values of ψ and a complex oscillatory mode $\psi_1\psi_2\psi_3$ for fixed values of $\mathbf{n}$.

The phase $\hat{\Delta}_c = \hat{\Delta}_a + \hat{\Delta}_b$, corresponding to the solution $\psi_i = \psi$, $|n_i| = 3^{-1/2}$, is symmetric relative to the group $H_c = D_3(C_3)$, $H_c \subset H_a$ (see Sec. 2, Paragraph B; for clarity of discussion the phase factor for $\hat{\Delta}_b$ is separated out; $\hat{\Delta}_b \to \hat{\Delta}_b e^{i\psi}$). The phase $\hat{\Delta}_c$ is not invariant relative to the operation of time reversal R, because R interchanges $e^{i\psi}$ and $e^{-i\psi}$. Because $H_c \subset H_b$, in order to find out the nature of the transition between the phases with symmetry H_b (which arises, as we have seen for $x<x_m$) and the phase with H_c, it is necessary to investigate the factor group

$$H_b/H_c = [D_3(C_3)\times\widetilde{R}]/D_3(C_3) = (E, \widetilde{R}).$$

Because the cube of a basis function from the single nonunique one-dimensional representation of this group is not an invariant, the phase transition in question is a second-order transition. Formally, the transition takes place as the quantity ψ in (7) reduces to zero, i.e., on the line

$$\gamma_1\Delta_b(x, T)/4\sqrt{3}\gamma_3\Delta_a(x, T)=1 . \qquad (10)$$

The diagram corresponding to the phase transitions under discussion is shown in Fig. 4.

In conclusion, let us dwell briefly on the region $\beta_3 < -\gamma_1^2/2\gamma_3$, where the order parameter is not even symmetric relative to $D_3(C_3)$. Without even studying the functional (2)–(5) at the minimum, we can confirm that the transition between a phase with symmetry $H_c \subset H_a$ which arises for $T \lesssim T_{cb}(x)$, and phase with symmetry $H_b = D_3(C_3) \times \tilde{R}$, is a first-order transition. Actually, even if the group H_c satisfies the condition $H_c \subset H_b$ which is necessary for a second-order transition, our investigation shows that we always can construct a third-order invariant from basis functions of the irreducible representations of the factor-group H_b with respect to any of its subgroups H_c (excepting the already-discarded $D_3(C_3)$, and also the trivial one E, which always can be augmented by a symmetry element $U_2e^{i\pi}$, because the basis function of the F_1 representation change sign when rotated around one of the second-order axes. We present here two of the factor groups H_b/H_c:

$$[D_3(C_3)\times\tilde{R}]/(E, U_2e^{i\pi})=C_3\times\tilde{R},$$
$$[D_3(C_3)\times\tilde{R}]/(E, U_2e^{i\pi})\times\tilde{R}=C_3.$$

[1)]We should mention here a phenomenological descrption of the phase transition in $U_{1-x}Th_xBe_{13}$ in terms of a transition connected with the establishment of a coherent state in a Kondo lattice whose transition temperature for $x > 1.75\%$ lies below T_c (Ref. 10). The question of whether such a transition can occur remains open.

[2)]Let us recall that the classes of colorless groups[25] coincide with the crystal classes, while the difference between magnetic order and pure crystalline order arises because of the combination of the operator R with the translation operators.

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