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Coupled s-Wave and d-Wave States in the Heavy Fermion
Superconductor $U_{1-x}Th_xBe_{13}$

by

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Coupled s-Wave and d-Wave States in the Heavy Fermion

Superconductor $U_{1-x}Th_xBe_{13}$

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Abstract

In the heavy-fermion superconductor $U_{1-x}Th_xBe_{13}$, superconducting states coexist for thorium concentrations $0 \leq x \leq 0.06$. Assuming s-wave and d-wave symmetries for these states, we derive a Ginzburg-Landau free energy expression which couples s- and d-wave states and is rotationally invariant, in contrast to the free energy expression proposed by Kumar and Wolfle (Phys. Rev. Lett. 59, 1954 (1987)). We discuss in detail the consequences that follow from our free energy relation. In particular, we predict that in the above system there are two eigenfrequencies associated with the dynamics of phase oscillations (internal Josephson effect) which are characteristic of the s-wave and d-wave states.

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In the heavy-fermion system $U_{1-x}Th_xBe_{13}$, different superconducting states have been found¹⁻⁷ to coexist for impurity concentration x in the range $0 < x \leq x_2$ ($x_2 \approx 0.06$). Two superconducting transitions are observed in specific heat^{1,2} and critical field^{3,4} measurements. For undoped UBe_{13} , the higher temperature transition occurs at $T_h = 0.87$ K, whereas the lower transition occurs at $T_l = 0.55$ K. Upon doping with thorium beyond a critical concentration $x_1 \approx 0.017$, i.e., $x_2 > x > x_1$, the high-temperature and low-temperature superconducting states are reversed from that in the undoped material.^{3,4} We assume the two superconducting states in question to possess even-parity spin-singlet s- and d-wave components. We are aware that an interpretation of some of the properties of the superconducting state of the UBe_{13} system in terms of an odd-parity state can also be forwarded;⁸ however, it is our interest to study the relevance of s- and d-wave coupling in this material.

The study of coupling between s-wave and d-wave states is also relevant to other heavy-fermion systems, e.g., La-doped UBe_{13} ⁷ and UPt_3 .⁹ For the high- T_c superconductors, the relevance of s-wave and d-wave coupling is suggestive from the microscopic theories^{10,11} based on the Hubbard model. High resolution X-ray scattering experiments¹² on the 123-superconductor, $YBa_2Cu_3O_7$, indicate that there might exist a special interference between d- and s-wave states. The coupling of superconducting states is reminiscent of the classic A- to-B transition in superfluid 3He which involves coupling between different spin-polarization states of the odd-parity triplet pairing of the quasi-particles.^{13,14} Superconductivity due to d-wave and s-wave states has also been considered by Anderson and Morel¹⁵ and by Mermin and Stare.¹⁶

In this paper we develop a model to explain the important electronic and magnetic transitions observed in the $U_{1-x}Th_xBe_{13}$ system. The higher temperature transition in pure UBe_{13} is due to a d-wave state, while the lower transition is due to an s-wave state coexisting with the d-wave state. We assume, following Kumar and Wolfe (KW),¹⁷ that thorium doping reduces the transition temperature for the anisotropic d-waves due to impurity scattering whereas the transition temperature for the isotropic s-waves is hardly affected. Thus, for $x > x_1$, the d-wave transition temperature $\bar{T}_2 (= T_l)$ is reduced to a value lower than that of the s-wave transition temperature $T_0 (= T_h)$.

In even-parity spin-singlet superconducting states, the energy gap function $\Delta(\vec{k})$ is related to the anomalous thermal average $\langle c_{\vec{k}\uparrow}^\dagger c_{-\vec{k}\downarrow} \rangle$ of the microscopic theory, $c_{\vec{k}\uparrow}^\dagger$ being the electron annihilation operator with wave vector \vec{k} and spin \uparrow . We expand $\Delta(\vec{k})$ as a linear combination of the basis function set $\{\phi_j(\hat{k})\}$, where $\phi_0(\hat{k}) = (C_x + C_y + C_z)/\sqrt{3}$, $\phi_1(\hat{k}) = (C_y - C_x)$ and $\phi_2(\hat{k}) = (C_x + C_y - 2C_z)/\sqrt{3}$ with $C_i = \cos(k_i a)$ ($i = x, y, z$ and a the lattice constant):

$$\Delta(\vec{k}) = \sum_{j=0}^2 \eta_j(k) \phi_j(\hat{k}) \quad , \quad (1a)$$

$$\eta_j(k) = \Delta_j(k) \exp(i\theta_j) \quad , \quad j = 0, 1, 2 \quad . \quad (1b)$$

We restrict our analysis to angular momentum states $l \leq 2$ upon which the basis functions $\phi_j(\hat{k})$ can be replaced by the spherical harmonics $Y_j(\hat{k})$. Then $j = 0$ labels the s-states and $j = 1, 2$ label the two d-state components of the E_g irreducible representation of the octahedral point group, O_h . The order



parameter amplitudes $\Delta_j(k)$ and the phases θ_j are all real. Y_0 is a constant, whereas Y_1 and Y_2 are analogous to the $d_{x^2-y^2}$ and d_{z^2} orbitals, respectively, of atomic physics. The symmetry group of the gap function is¹⁸⁻²⁰ $G \times R \times U(1)$, where G is the crystal point group, R is the time reversal operator, and $U(1)$ is a gauge transformation group. In coordinate space, the Ginzburg-Landau free energy of the superconducting states (relative to the normal state) can be written as $F = \int d^3r (F_L + F_G)$, where the free energy density F_L is restricted to fourth order in the order parameters and F_G contains the gradient terms. We write,²¹ $F_L = F_{KW} + F_E$, where,

$$F_{KW} = \alpha_0 \Delta_0^2 + \beta_0 \Delta_0^4 + \alpha_2 \Delta_2^2 + \beta_2 \Delta_2^4 + \gamma_2 \Delta_0^2 \Delta_2^2 + \gamma_2 \delta_2 \Delta_0^2 \Delta_2^2 \cos^2 \theta_2 + \gamma_1 \Delta_0 \Delta_2^3 \cos \theta_2, \quad (2a)$$

and

$$F_E = \alpha_2 \Delta_1^2 + \beta_2 \Delta_1^4 + \gamma_2 \Delta_0^2 \Delta_1^2 + \gamma_2 \delta_2 \Delta_0^2 \Delta_1^2 \cos^2 \theta_1 + \gamma_1 \Delta_1^2 \Delta_2^2 + \gamma \delta \Delta_1^2 \Delta_2^2 \cos^2(\theta_1 - \theta_2) - \gamma_1 \Delta_0 \Delta_1^2 \Delta_2 [2 \cos \theta_2 + \cos(2\theta_1 - \theta_2)] \quad (2b)$$

In deriving Eq. (2) we have adopted the notation of Kumar and Wolfe (KW)¹⁷ to facilitate comparison with their work; our Eq. (2a) is identical to their Eq. (1). Note that Eq. (2a) alone, without Eq. (2b), is not rotationally invariant, since the d-wave order parameter amplitudes Δ_1 and Δ_2 transform as partner components of the irreducible two-dimensional representation E_g of the cubic group O_h . Equation (2) belongs to the product representation $A_{1g} \times E_g$. Note also that the coupling terms $\Delta_0 \Delta_2^3$ and $\Delta_0 \Delta_1^2 \Delta_2$ arise from the fact that $\int d\Omega Y_2^3(\hat{k}) \neq 0$ and $\int d\Omega Y_2(\hat{k}) Y_1(\hat{k})^2 \neq 0$, respectively; these two terms transform into each other under symmetry

operations of the O_h group. The gauge invariance of Eq. (2) follows from the fact that θ_1 and θ_2 are measured relative to θ_0 , the phase angle of the s-wave state.

As usual, we assume that $\alpha_0 = A_0(T-T_0)$ and $\alpha_2 = A_2(T-T_2)$, where T_0 and T_2 are the uncoupled s-wave and d-wave critical temperatures, respectively, and that the other coefficients are independent of temperature. The coefficients β_0 and β_2 are assumed positive and large, relative to the other fourth-order coefficients, to insure stability. We assume the condition $T_2 > T_0$ for the undoped material, whereas the reverse is true for the doped material with $x_2 > x > x_1$ ($x_2 = 0.06$ and $x_1 = 0.017$). We consider the following seven possible superconducting states: (I) $\Delta_1 \neq 0, \Delta_2 = \Delta_0 = 0$, (II) $\Delta_2 \neq 0, \Delta_1 = \Delta_0 = 0$, (III) $\Delta_1, \Delta_2 \neq 0, \Delta_0 = 0$, (IV) $\Delta_0 \neq 0, \Delta_1 = \Delta_2 = 0$, (V) $\Delta_0, \Delta_1 \neq 0, \Delta_2 = 0$, (VI) $\Delta_0, \Delta_2 \neq 0, \Delta_1 = 0$ and finally (VII) $\Delta_0, \Delta_1, \Delta_2 \neq 0$.

For a transition to a superconducting d-state, the case $T_2 > T_0$ and $T = T_2^-$ is appropriate. For (a) $\gamma\delta > 0$ and $\theta_2 = \theta_1 \pm \pi/2$, state I or state III is favored depending on whether $\gamma > 2\beta_2$ or $2\beta_2 - |\gamma| > 0$, respectively; for (b) $\gamma\delta < 0$ and $\theta_2 = \theta_1 \pm \pi$, state I or state III is favored depending on whether $\gamma(1+\delta) > 2\beta_2$ or $2\beta_2 - |\gamma(1+\delta)| > 0$, respectively. Near $T_2 (> T_0)$ state I is stable with respect to non-vanishing Δ_0 and Δ_2 since these two order parameters produce a second-order effect through $\Delta_0\Delta_2$. State II is normally degenerate with state I in the absence of coupling; however, the (s+d)-coupling lifts this degeneracy and makes state II unstable with respect to state VI. With a decrease of temperature, state III goes over to state VII since coupling to Δ_0 becomes important. In view of the restrictions on the fourth-order coefficients imposed in the previous paragraph, we favor state III as the higher-temperature d-state for the undoped material. Taking $\gamma < 0$,

$\gamma_1 > 0$, $\gamma_2 < 0$, $\delta < 0$ and $\delta_2 < 0$, we have minimized Eq. (2) numerically in the five-dimensional space of the order parameters and the relative phase angles. Our plots for the normalized order parameters and the phase angle θ_1 , as a function of temperature are given in Fig. 1; θ_2 exhibits a constant value of $\pi/2$.

For a superconducting transition from the normal to the s-wave state, as occurs in the doped material, the choice $T_0 > T_2$ with $T = T_0^-$ is appropriate. Taking all other parameters exactly the same as in Fig. 1, state IV is favored. With a decrease of temperature, the d-wave states get populated at a temperature \bar{T}_2 which is slightly up-shifted from T_2 . The s-wave and d-wave order parameters coexist giving rise to state VII. Note that state V is unstable with respect to state VII, since the linear term involving Δ_2 acts as an "applied field" and lowers the free energy. Our numerical results appropriate for the doped materials are plotted in Fig. 2. For this figure we find that state VII is stable with respect to state VI.²²

The d-wave superconducting states have some characteristic signatures such as power-law temperature dependencies in specific heat, muon spin rotation, nuclear magnetic resonance and ultrasonic sound attenuation experiments. These are associated with vanishing of the gap function along certain symmetry lines or planes near the Fermi surface. Another characteristic signature is the dynamics of the relative phase angles θ_1 and θ_2 due to (s+d)-coupling, similar to the internal Josephson effect in superfluid ³He.^{13,14} The particle numbers N_1 and N_2 (relative to N_0) are conjugate to the phase angles θ_1 and θ_2 and obey the commutation relations $[\theta_i, N_j] = i\delta_{ij}$. We generalize Eqs. (2) to include the chemical potentials and linearize the associated Hamilton's equations about θ_1^0 and θ_2^0 , the minimum solutions. The characteristic phase oscillation frequencies ω^2 are then obtained by solving

the secular equation, $(a_{11}-\omega^2)(a_{22}-\omega^2) - a_{12}a_{21} = 0$, where $a_{ij} = (\partial^2 F_L / \partial \theta_i \partial \theta_j)$, $i, j = 1, 2$. The solution of the characteristic equation is analogous to that of a double pendulum²³ in which the higher frequency normal mode is connected with a symmetric vibration and the lower frequency one with an asymmetric vibration. For solutions corresponding to Fig. 1, we find that the high-frequency mode appears at the first transition, whereas the low frequency mode appears at the second transition. In contrast, both the normal mode solutions for Fig. 2 appear at the second transition (since the first transition is to a pure s-wave state, there is no coupling). These characteristic phase oscillations thus provide a method to identify the superconducting states. At low temperatures, the high-frequency fundamental vibration can couple to even-harmonics (the odd-harmonics are absent) of the low-frequency fundamental mode, thus producing a resonance. Our predictions in this regard are in sharp contrast to that of KW, who argued for a single dynamic frequency. We find that the fundamental frequencies increase monotonically as $T \rightarrow 0$, and we do not find any evidence for non-monotonic behavior as suggested by KW.

Finally, we would like to comment on the gradient terms F_G of the free energy density. For an applied magnetic field \vec{H} along the z-direction, $\vec{H} = (0, 0, H)$, we choose the vector potential in the Landau gauge, $\vec{A} = (0, A_y, 0)$, $A_y = xH$. With $\Phi_0 = hc/2e$, we obtain the invariant terms by setting up covariant differential operators. For the sake of simplicity, we assume a one dimensional coordinate dependence of the order parameter amplitudes: $\eta_j(\vec{r}) = \eta_j(x) = \Delta_j(x) \exp(i\theta_j)$, $j = 0, 1, 2$, denote derivatives with respect to x by primes and obtain²⁴ for $F_G = G_{KW} + G_E$

$$G_{KW} = |\alpha_0| \xi_0^2 [\Delta_0'^2 + (2\pi/\Phi_0)^2 A_y^2 \Delta_0^2] + |\alpha_2| \xi_2 [\Delta_2'^2 + \Delta_1'^2 + (2\pi/\Phi_0)^2 A_y^2 (\Delta_2^2 + \Delta_1^2)] , \quad (3a)$$

and

$$G_E = |\alpha_2| \xi_d^2 \left\{ \Delta_2'^2 - \Delta_1'^2 + \sqrt{3} \Delta_1' \Delta_2' \cos(\theta_2 - \theta_1) + (2\pi/\Phi_0)^2 A_y^2 [\Delta_2^2 - \Delta_1^2 - \sqrt{3} \Delta_1 \Delta_2 \cos(\theta_2 - \theta_1)] \right\} +$$

$$|\alpha_2| \xi_{sd}^2 [\Delta_0' (\sqrt{3} \Delta_1' \cos \theta_1 - \Delta_2' \cos \theta_2) - (2\pi/\Phi_0)^2 A_y^2 \Delta_0 (\sqrt{3} \Delta_1 \cos \theta_1 + \Delta_2 \cos \theta_2)] . \quad (3b)$$

Here Φ_0 is the flux quantum; ξ_0, ξ_2, ξ_d and ξ_{sd} are coherence lengths. We have separated the terms in Eq. (3) in the same spirit as that of Eq. (2). Note that the coefficients of the ξ_{sd} -terms mix the s- and d-waves; note also that there is an extra d-wave invariant whose coefficient is ξ_d^2 , which was not considered by KW. Employing Eqs. (2) and (3), we obtain the differential Ginzburg-Landau (GL) equations and use them to derive expressions for the upper and the lower critical fields H_{c_2} and H_{c_1} , respectively.

The upper critical field is the highest field at which the normal state free energy equals that of the mixed state. For simplicity, we set $\xi_{sd}^2 = 0$ and obtain:

$$H_{c_2} = (\Phi_0/2\pi) [c(\xi_2^4 - \xi_d^4) + 2\xi_2^2 \xi_0^2] / [(c+2)(\xi_2^4 - \xi_d^4) \xi_0^2] , \quad (\text{for Fig. 1}), \quad (4a)$$

$$H_{c_2} = (\Phi_0/2\pi) (1+2c') / [(\xi_0^2 + 2c' \xi_2^2)^2 - 12c' \xi_d^4]^{1/2} , \quad (\text{for Fig. 2}) . \quad (4b)$$

The coefficients c and c' are defined as Δ_0/Δ and Δ/Δ_0 respectively ($\Delta = \Delta_1 - \Delta_2$) and are related in a complicated way to the nonlinear terms of F_L . Since c and c' are less than one, the effective coherence length is dominated by ξ_2^2

or ξ_0^2 as the case may be. Setting them to zero we recover the expected limits. In deriving Eq. (4) we have found the ground state simple harmonic oscillator solutions to the linearized GL equations to be adequate.

The lower critical field, i.e., the field at which the Meissner state is thermodynamically stable, is directly proportional to the super-electron density. Consequently, the transition to the mixed (s+d)-wave state should be accompanied by an increase in the temperature coefficient of H_{c1} as observed in Ref. 3. By assuming the large $\nu(-\lambda/\xi)$ solution for H_{c1}^{25} and the London form for the supercurrent, we can write $H_{c1} = \Phi_0 \ln \kappa_{\text{eff}} / (2\pi\lambda_{\text{eff}}^2)$, with $\lambda_{\text{eff}}^{-2} = (\lambda'_0)^{-2} + (\lambda'_2)^{-2} + (\lambda'_d)^{-2}$, where $(\lambda'_0)^{-2} = 16\pi^2 \alpha_0 \xi_0^2 \Delta_0^2 / \Phi_0^2$, $(\lambda'_2)^{-2} = 32\pi^2 \alpha_2 \xi_2^2 \Delta^2 / \Phi_0^2$ and $(\lambda'_d)^{-2} = (\sqrt{3}/2)(\lambda'_2)^{-2} (\xi_d/\xi_2)^2 \cos(\theta_1 - \theta_2)$. The large effect of the relative phase difference $(\theta_1 - \theta_2)$ on $(\lambda'_d)^{-2}$ may explain the anomaly in κ_{eff} near the lower transition temperature reported in Ref. 3.²⁶

In summary, we have derived, from general symmetry principles, the Ginzburg-Landau free energy for the mixed $A_{1g} \times E_g$ representation of the octahedral point group relevant for pure and thorium-doped UBe_{13} . By detailed numerical work, we have obtained the nature of the superconducting states that might exist in this system. We have predicted the existence of a symmetric and an asymmetric normal-mode phase oscillation frequency characteristic of the coupled states. Finally, we have examined the behavior of the critical fields for this system.

Acknowledgments

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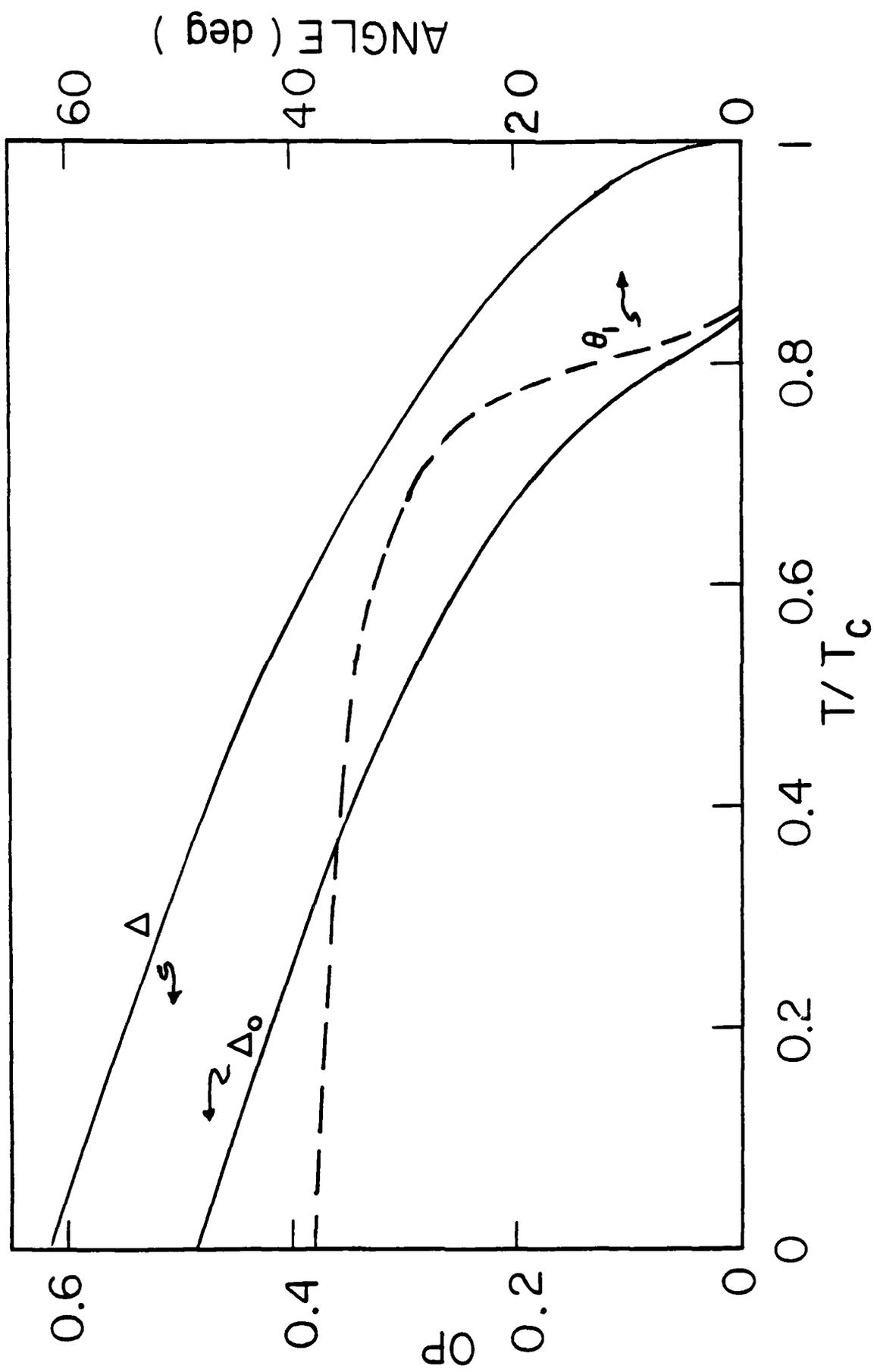
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21. The symmetry-classified (s+d)-coupled free energy for several crystal point groups have been recently obtained by D. Sahu, A. Langner and T. F. George (unpublished).
22. We were also unable to obtain a state IV to state VI transition for several other parameter sets.
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26. The values for Δ_0^2 and Δ^2 in these expressions are generally not equal to the single order parameter-state values.

Figure Captions

1. Temperature dependence of order parameters (OP) Δ_0 and Δ ($-\Delta_1-\Delta_2$) and phase angle θ_1 for critical temperatures $T_2/T_c = 1$ and $T_0/T_2 = 0.75$ and phase angle $\theta_2 = \pi/2$. This figure is appropriate for undoped UBe_{13} . The x-axis and the left-hand side y-axis variables are in dimensionless units.
2. Same as in Figure 1, but with $T_0/T_c = 1$ and $T_2/T_0 = 0.75$. This figure is appropriate for $U_{1-x}Th_xBe_{13}$.



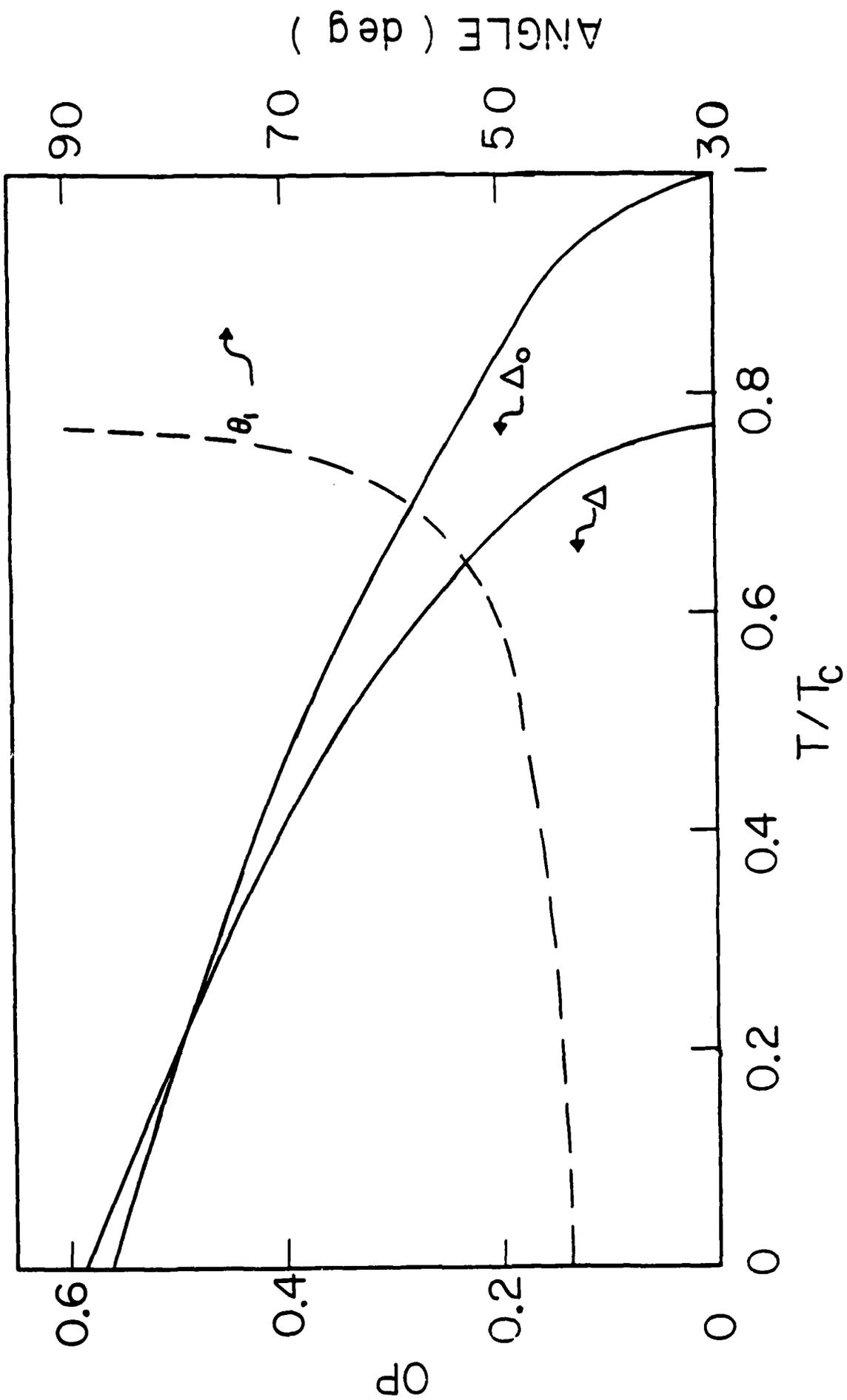


Fig. 1

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