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Anisotropic superconductivity in $PrOs_4Sb_{12}$

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Abstract. Recently two anisotropic superconducting gap functions have been observed in the skutterudite $PrOs_4Sb_{12}$. These order parameters are spin-triplet. There are at least 2 distinct phases in a magnetic field, bearing some resemblance to superfluid ^{3}He. Here we present an analysis of the thermodynamic properties in these two superconducting states within the weak-coupling BCS theory.

PACS. 74.20.Fg BCS theory and its development

1 Introduction

Superconductivity in the body-centered cubic heavy-fermion (HF) skutterudite $PrOs_4Sb_{12}$ was discovered in 2002 by Bauer et al. [1–3]. Since then many experimental and theoretical studies of this compound have been reported. This compound possesses several interesting and unusual characteristics: two distinct phases (the A phase and B phase) in a magnetic field, nodal superconductivity with point nodes, and triplet pairing with chiral symmetry breaking [6–8]. The phase diagram is still controversial. In Figure 1 recent measurements by Measson et al. [9] are shown.

It was recently observed that the magnetothermal conductivity data [4,7] in this compound are consistent with anisotropic superconductivity using the gap functions

$$\boldsymbol{\Delta}_A(\mathbf{k}) = \mathbf{d}e^{\pm i\phi_i}\frac{3}{2}(1-\hat{k}_x^4-\hat{k}_y^4-\hat{k}_z^4)). \quad (1)$$

$$\boldsymbol{\Delta}_B(\mathbf{k}) = \mathbf{d}e^{\pm i\phi_3}(1-\hat{k}_z^4). \quad (2)$$

Here $e^{\pm i\phi_i}$ is one of $e^{i\phi_1} = (\hat{k}_y + i\hat{k}_z)/\sqrt{\hat{k}_y^2+\hat{k}_z^2}, e^{i\phi_2} = (\hat{k}_z + i\hat{k}_x)/\sqrt{\hat{k}_z^2+\hat{k}_x^2}, e^{i\phi_3} = (\hat{k}_x + i\hat{k}_y)/\sqrt{\hat{k}_x^2+\hat{k}_y^2}$. The factor of 3/2 ensures proper normalization of the angular dependence of the order parameter. In equation (2) the nodal direction is chosen to be parallel to (001).

We note that the proposed order parameter (2) lies outside of the usual classification scheme [11], in which order parameters correspond to a single irreducible representation of the rotation group. However, this hybrid order parameter appears to be necessary to reproduce the observed B-phase gap structure. A similar situation has been observed in the borocarbide superconductors [12].

The cubic symmetry of $PrOs_4Sb_{12}$ suggests order parameters which are invariant under the T_h cubic tetrahedral symmetry group applicable to this crystal [10], as well as reflections (containing the origin) about the planes of the crystal parallel to the cube faces [13]. As suggested in [10], one possible invariant is $\hat{k}_x^2\hat{k}_y^2+\hat{k}_y^2\hat{k}_z^2+\hat{k}_z^2\hat{k}_x^2$. This belongs to the A_1 representation of T_h [10].

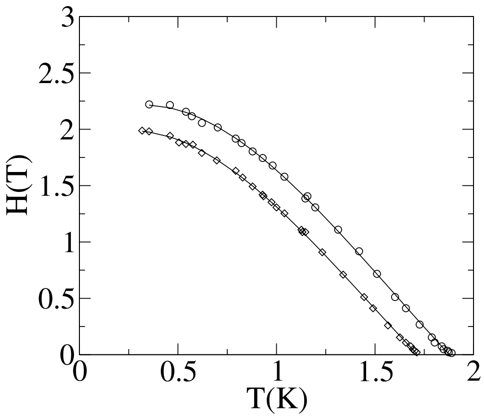

Fig. 1. Phase diagram by Measson et al. [9]. The top line and points are the upper critical field H_{c2}, while the lower ones are the phase boundary H'.

This combination can be recast as $1-\hat{k}_x^4-\hat{k}_y^4-\hat{k}_z^4$, thus forming the basis of the proposed order parameter of the A phase. Furthermore, in weak-coupling BCS theory the quasiparticle density of states and the thermodynamics depend only on $|f|$ [16], the magnitude of the angle-dependent part of the order parameter. For this reason, an order parameter which breaks chiral symmetry still retains the essential features of the cubic symmetry, and is in fact necessitated by the triplet pairing observed in this compound [6–8]. Triplet pairing requires that the orbital wavefunction be antisymmetric under particle interchange. The phase factor proposed in equation (1) meets this requirement.

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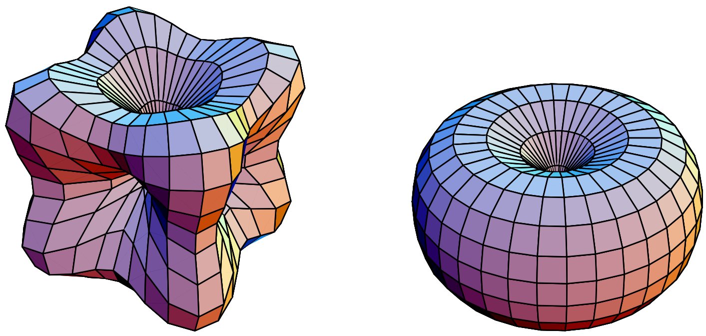

Fig. 2. A-phase (left) and B-phase (right) order parameters.

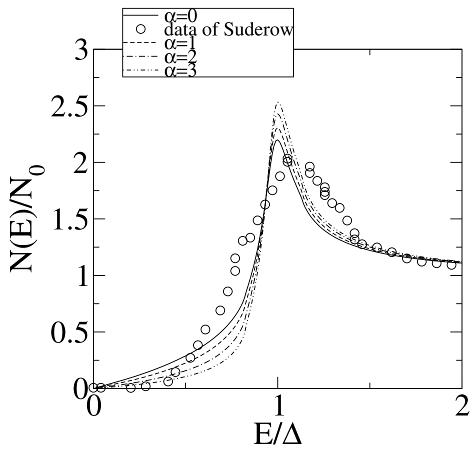

Fig. 3. Comparison of quasiparticle DOS in B phase with STM data of Suderow et al. [18].

The proposed B-phase order parameter breaks the cubic symmetry more manifestly. Nevertheless, there is almost certainly a relationship between the A and B phase, particularly since the zero-field transition temperatures are so nearly equal. The simplest relationship consistent with the nodes at [001] and [00-1], despite requiring a hybrid representation, would suggest $|f| \sim 1-\hat{k}_z^4$. This order parameter again would have a phase factor included in f to ensure antisymmetry under interchange. These proposed order parameters are illustrated in Figure 2 [17].

We note that while the B-phase is the prevalent phase in zero magnetic field, the A-phase exists at all temperatures below T_c for fields between H^* (the phase boundary) and H_{c2}.

Presented above is a comparison of the predicted B-phase DOS with scanning tunneling microscopy (STM) data taken by Suderow et al. [18] at $T = 0.19$ K. The predicted B-phase DOS differs somewhat from that presented in [7] due to the use of the angular-dependent quasiparticle density-of-states, as well as an accounting for the energy and directional resolution of the STM. Here we have assumed that the STM performed measurements along the nodal directions, where α is the size of the momentum cone within the STM's spatial resolution. Here we have assumed Δ to take the B-phase weak-coupling value of 3.3 K.

We note that the observed small DOS for $E < \Delta/3$ can be reproduced by choosing α to be 3.0. We observe fair agreement, with some differences apparent surrounding the quasiparticle peak at $E = \Delta$. A lowering and broadening of the central peak at $E = \Delta$, as would be expected from the use of a more realistic Fermi surface (instead of the sphere assumed here), would yield a better fit to the data. In the following we analyze both phases over the entire temperature range from $T = 0$ to T_c.

2 Weak-coupling BCS theory

We focus on the superconductivity in the A and B-phases of $PrOs_4Sb_{12}$, using the $\Delta(\mathbf{k})$ given by equations (1) and (2) with $|\mathbf{d}| = \Delta(T)$.

Then, within the weak-coupling theory the gap equation is given by

$$\lambda^{-1} = 2\pi T \langle f^2\rangle^{-1} \sum_n \left\langle \frac{f^2}{\sqrt{\omega_n^2 + \Delta^2 f^2}} \right\rangle \quad (3)$$

$$= \langle f^2\rangle^{-1} \int_0^{E_0} dE \left\langle \frac{f^2}{\sqrt{E^2 - \Delta^2 f^2}} \right\rangle \tanh\left(\frac{E}{2T}\right) \quad (4)$$

where λ is a dimensionless coupling constant, E_0 is the cut-off energy, and ω_n is the Matsubara frequency. Here, for the A-phase $f = \frac{3}{2}(1-\hat{k}_x^4-\hat{k}_y^4-\hat{k}_z^4)$, and $\langle \ldots \rangle$ denotes $\int d\Omega/4\pi$. For the B-phase, $f = 1 - z^4$, and $\langle \ldots \rangle$ denotes $\int_0^1 dz \ldots$. The frequency sum in equation (3) is cut off at $\omega_n = E_0$.

In the vicinity of $T = T_c$ and $T = 0K$, equations (3) and (4) can be solved analytically. For $T \to T_c$ we obtain

$$T_c = \frac{2\gamma}{\pi} E_0 e^{-1/\lambda} = 1.136 E_0 e^{-1/\lambda} \quad (5)$$

$$\Delta^2(T) \approx \frac{2\langle f^2\rangle (2\pi T_c)^2 \ln(T/T_c)}{7\zeta(3)\langle f^4\rangle} \quad (6)$$

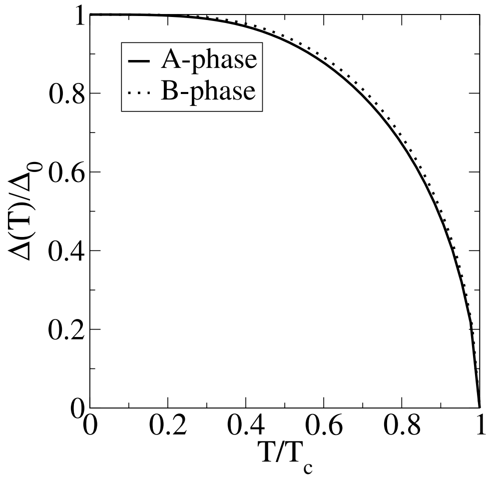

Fig. 4. $\Delta(T)$ for the A and B phases.

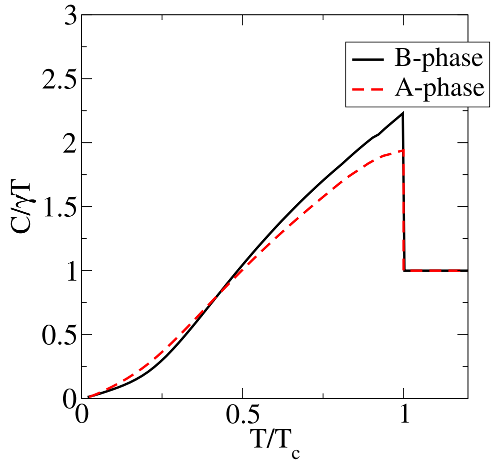

Fig. 5. Specific heat $C_s/\gamma_S T$ for $PrOs_4Sb_{12}$.

where $\gamma = 1.78\ldots$ is the Euler constant and

$$\Delta(0)/T_c = \frac{\pi}{\gamma} \exp\left[-\langle f^2\rangle^{-1}\langle f^2 \ln(f)\rangle\right] \quad (7)$$
$$= 2.364, \text{A-phase} \quad (8)$$
$$= 1.938, \text{B-phase}. \quad (9)$$

In the low-temperature regime $T/\Delta(0) \ll 1$ one obtains

$$\ln(\Delta(T)/\Delta(0)) = -\frac{7\pi\zeta(3)}{8}\left(\frac{T}{\Delta(0)}\right)^3 \quad \text{A-phase} \quad (10)$$

$$\ln(\Delta(T)/\Delta(0)) = -\frac{135\pi\zeta(3)}{512}\left(\frac{T}{\Delta(0)}\right)^3 \quad \text{B-phase}. \quad (11)$$

In Figure 4 numerical solutions of $\Delta(T)/\Delta(0)$ are shown for both phases over the entire temperature range.

The values of $\Delta(0)_A$ and $\Delta(0)_B$ obtained from the weak-coupling theory offer a possible explanation for the multiphase diagram of $PrOs_4Sb_{12}$. The value of the condensation energy at $T = 0$ is given by

$$E_0 = -\frac{1}{2}\langle |f|^2\rangle N_0 \Delta_0^2 \quad (12)$$

where f is the angular-dependent part of the order parameter and N_0 the normal state density of states at the Fermi level. For the A-phase, $\langle |f|^2\rangle = 3/7$, whereas for the B-phase, $\langle |f|^2\rangle = 32/45$. This yields two distinct condensation energies:

$$E_0^A = -1.17 N_0 (T_c^A)^2, \quad (13)$$
$$E_0^B = -1.32 N_0 (T_c^B)^2. \quad (14)$$

If one uses the experimental values [9] for T_c^{A} and T_c^{B} as 1.887 K and 1.716 K, respectively, one finds that E_0^{A} is slightly lower than E_0^{B}, contrary to observation. On the other hand, if we assume $T_c^{\mathrm{A}} = T_c^{\mathrm{B}}$, which the simplest interaction would give, we find $E_0^{\mathrm{B}} < E_0^{\mathrm{A}}$. The difference between our assumed and the measured T_c would in this case be due to some unknown external perturbation, such as the effect of the crystalline electric field (CEF), not accounted for in this treatment. One possibility is that the CEF affects superconductivity in the two phases differently, resulting in a difference in measured transition temperatures. Indeed, if we assume that, absent such an effect, we would have $T_c^{\mathrm{A}} = T_c^{\mathrm{B}}$ we find $\Omega_0^{\mathrm{B}} < \Omega_0^{\mathrm{A}}$ for all temperatures.

Upon evaluating $\Delta(T)$, the thermodynamics of the system can be analyzed following reference [19]. Let us start with the entropy:

$$S_s = -4\int_0^\infty dE\, N(E)(f\ln f + (1-f)\ln(1-f)), \quad (15)$$

where f is the Fermi-Dirac distribution $(1+e^{\beta E})^{-1}$ with $\beta = 1/k_B T$. $N(E)$ is the quasiparticle density of states,

$$N(E) = N_0 \mathrm{Re}\left\langle \frac{|E|}{\sqrt{E^2 - \Delta^2 f^2}}\right\rangle. \quad (16)$$

The electronic specific heat can be derived from the entropy via

$$C_s = T\frac{\partial S}{\partial T}. \quad (17)$$

In Figure 5 we show $C_s/\gamma_S T$ versus T/T_c for both phases. Here $\gamma_S = 2\pi^2 N_0/3$ is the Sommerfeld constant. We find the jump $\Delta C/C$ at T_c to be approximately 0.93 for the A-phase and 1.20 for the B-phase. In addition, as expected, the low-temperature specific heat is predicted to be proportional to T^2 [5] for both phases. Unfortunately, the presence of a Schottky specific heat peak [2] at low temperature makes assessment of the T^2 prediction difficult.

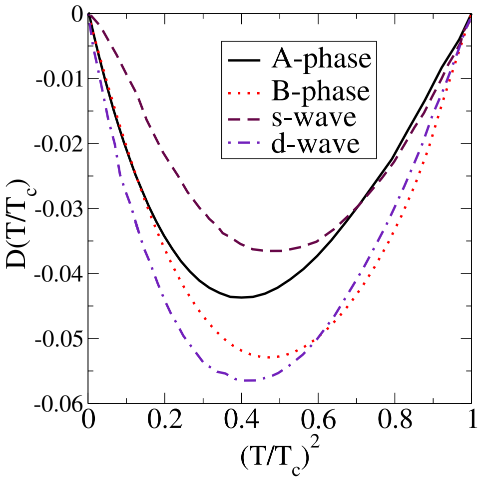

Fig. 6. Deviation of critical field from parabolic dependence for $p+h$, s, and d-wave superconductors.

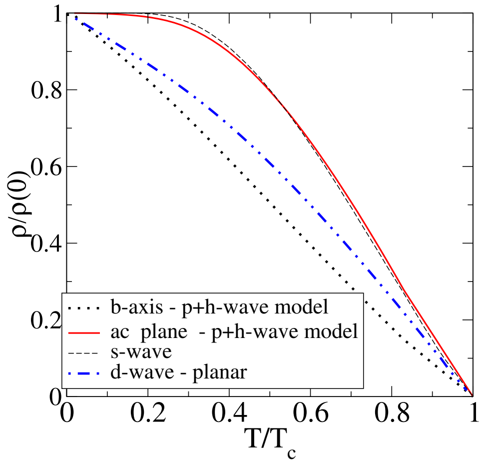

Fig. 7. Predicted superfluid densities for $PrOs_4Sb_{12}$.

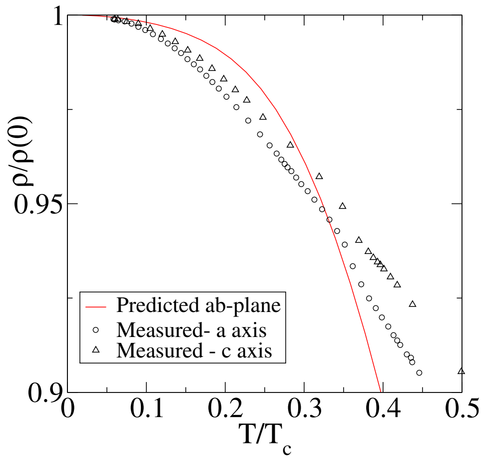

Fig. 8. Superfluid densities for $PrOs_4Sb_{12}$.

Also the thermodynamical critical field $H_c(T)$ can be obtained from

$$F_S(T) - F_N(T) = -\int_T^{T_c} dT\,(S_S(T) - S_N(T)) \quad (18)$$

$$= -\frac{1}{8\pi} H_c^2(T). \quad (19)$$

Here S_S(T) and $S_N = \gamma_N T$ are the entropies in the superconducting and normal state respectively. We show $D(\frac{T}{T_c}) = H_c(T)/H_c(0) - (1 - (T/T_c)^2)$ for both phases in Figure 6. The function $D(\frac{T}{T_c})$ for both the A-phase and B-phase cases is slightly larger than for the isotropic s-wave case and somewhat smaller than for the d-wave case [20], with the B-phase case slightly larger than the A-phase case.

While there are a few reports of $H_{c2}(T)$ for the A-phase and $H^*(T)$, the phase boundary between the A phase and the B phase [1,4,9,21–23], no experimental data are available for $H_c(T)$.

Finally the superfluid density is given by

$$\frac{\rho_{s\parallel}(T)}{\rho_{s\parallel}(0)} = 1 - 3\int_0^\infty \frac{dE}{2T}\text{sech}^2(E/2T) \times \text{Re}\left\langle z^2 \frac{E}{\sqrt{E^2 - \Delta^2(T) f^2}}\right\rangle \quad (20)$$

and

$$\frac{\rho_{s\perp}(T)}{\rho_{s\perp}(0)} = 1 - \frac{3}{2}\int_0^\infty \frac{dE}{2T}\text{sech}^2(E/2T) \times \text{Re}\left\langle (1-z^2) \frac{E}{\sqrt{E^2 - \Delta^2(T) f^2}}\right\rangle \quad (21)$$

where Re$\langle \ldots \rangle$ refers to the real part, and the subscripts $\parallel$ and $\perp$ indicate parallel and perpendicular directions to the nodal points. The superfluid density, as expected, is isotropic for the cubic symmetry-retaining A-phase, but rather anisotropic for the B-phase. These superfluid densities are shown in Figure 7. In the low-temperature regime ($T \ll \Delta$) both equations (20) and (21) can be expanded as

$$\frac{\rho_{sA}(T)}{\rho_s(0)} = 1 - \frac{\pi}{2}(\ln 2)\frac{T}{\Delta} + \ldots \quad (22)$$

$$\frac{\rho_{sB\parallel}(T)}{\rho_{s\parallel}(0)} = 1 - \frac{3\pi}{4}(\ln 2)\frac{T}{\Delta} + \ldots \quad (23)$$

$$\frac{\rho_{sB\perp}(T)}{\rho_{s\perp}(0)} = 1 - \frac{\pi^2}{16}(\frac{T}{\Delta})^2 + \ldots \quad (24)$$

Close to the transition temperature, we find

$$\frac{\rho_{sA}(T)}{\rho_{sA}(0)} \simeq \frac{6}{7}\frac{\langle f^2\rangle}{\langle f^4\rangle}(-\ln T/T_c) \quad (25)$$

$$= 1.393(-\ln T/T_c) \quad (26)$$

$$\frac{\rho_{sB\parallel}(T)}{\rho_{s\parallel}(0)} \simeq \frac{17 \cdot 13}{21 \cdot 11}(-\ln T/T_c) \quad (27)$$

$$= 0.9567(-\ln T/T_c) \quad (28)$$

$$\frac{\rho_{sB\perp}(T)}{\rho_{s\perp}(0)} \simeq \frac{31\cdot 221}{45\cdot 77}(-\ln T/T_c) \quad (29)$$
$$= 1.9772(-\ln T/T_c). \quad (30)$$

In the figure above we have compared $\frac{\rho_{sB\parallel}(T)}{\rho_{sB\parallel}(0)}$ with the data taken from Chia et al. [24], assuming that the nodal points in $\Delta(\mathbf{k})$ are aligned parallel to $\mathbf{H}$. Rather satisfactory agreement is observed for $T < T_c/3$. The differences in this regime could be due to a modest suppression of $\Delta(0)$ from the weak-coupling value cited earlier. The differences at higher temperatures are consistent with those closer to T_c (not shown), where the theoretical $\rho_{s\perp}(T)$ vanishes linearly with $T_c - T$, while Chia et al. [24] found a $\rho_{s\perp}(T)$ which vanishes with essentially infinite slope at T_c.

Recently Chia et al. [25] also reported magnetic penetration depth measurements for a range of dopings x from 0.1 to 0.8 in the compound $Pr(Os_{1-x}Ru_x)_4Sb_{12}$. Over the range from $x = 0.4$ to $x = 0.8$, exponential temperature dependence of the superfluid density was found, indicating an isotropic s-wave gap function in this regime. Of direct interest for this work, the superfluid density was found to go to zero linearly for all dopings, with no hint of the essentially infinite slope found [24] in the pure case ($x = 0$). In addition, the slope of these linear curves at T_c does not increase dramatically from $x = 0.8$ to $x = 0.1$. Further experiments at doping ranges between $x = 0$ and $x = 0.1$ are highly desirable, to examine more closely the apparent transition from nodal to conventional superconductivity taking place in this system. It would also be of value to confirm the rather unusual "infinite-slope" behavior observed in the pure sample near T_c.

3 Concluding remarks

We have worked out the weak-coupling theory of the A and B phases of the heavy-fermion superconductor $PrOs_4Sb_{12}$. A simple thermodynamic analysis offers an explanation for the appearance of the lower-symmetric B phase at lower temperatures. The present model leads to a fair description of STM data taken by Suderow et al. [18]. In addition, the present model for the B-phase describes the superfluid density determined by Chia et al. [24] for the low-temperature regime, if we assume that the nodal points in the B-phase follow the magnetic field direction in the field cooled situation [7]. Since the magnetic field is the only symmetry-breaking agent, this appears to be plausible. We will present the results of an analysis in the case of impurities shortly.

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