Possible type of heavy-electron superconductivity in $PrOs_4Sb_{12}$

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Abstract

Unconventional nature of superconducting state of $PrOs_4Sb_{12}$, Pr-based heavy electron compound with the filled Skutterudite structure, is shown to be explained by taking into account a structure of the crystalline-electric-field level and of the Fermi surface determined by the band structure calculation. In particular, the anisotropic pairing with a full gap on the Fermi surface, suggested by the measurement of the NQR relaxation rates $1/T_1$, is shown to be possible in the manifold of chiral "p"- or "d"-wave pairing, and to be compatible with the absence of the coherence peak and the pseudo-gap behavior of $1/T_1T$ far above the superconducting transition temperature.

Key words: $PrOs_4Sb_{12}$; unconventional superconductivity; non-Kramers doublet; quadrupolar Kondo lattice

Recently, the superconductivity has been found in heavy electron compound $PrOs_4Sb_{12}$ with the crystal structure of the filled Skutterudite [1]. Quite recently, a measurement of the longitudinal relaxation rates $1/T_1$ of NQR at Sb site has been performed and very unusual temperature (T) dependence was revealed both $T < T_c$ and $T > T_c$ [2], while the normal state properties had been known to be also quite unconventional [1,3]: 1) $1/T_1$ exhibits exponential T-dependence at $T < 1.3T_c$, giving the superconducting gap Δ in the low temperasture limit as $2\Delta/k_BT_c \simeq 5.3$. 2) There is no trace of the coherence (Hebel-Slichter) peak around $T = T_c$ at all. 3) pseudo-gap behavior is seen in $1/T_1T$ at $T_c < T < 2T_c$, in which the resistivity ρ also shows a pseudo-gap behavior [1]. These behaviors seem to suggest that a novel type of heavy electron superconductivity is realized in this compound.

The purpose of this contribution is to propose a scenario explaining such anomalous behaviors in a unified way on the basis of the crystalline-electric-field (CEF) level, inferred from the experiment [1] and theoretical study [4].

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The CEF level scheme proposed by Bauer *et al.* is given in the point group T_h as follows [1]: The ground state is the non-Kramers doublet $\Gamma_{23}^{\pm}$ ($\Gamma_3^{\pm}$ in the representation of the point group O_h)

$$|\Gamma_{23}^{+}\rangle = \sqrt{\frac{7}{24}}(|4\rangle + |-4\rangle) - \sqrt{\frac{5}{12}}|0\rangle, \tag{1}$$

$$|\Gamma_{23}^{-}\rangle = \frac{1}{\sqrt{2}}(|2\rangle + |-2\rangle), \tag{2}$$

and the first excited state with the excitation energy Δ_{23} =11 K is one of the triplet states $\Gamma_4^{(1)}$ or $\Gamma_4^{(2)}$ (Γ_4 or Γ_5 in the representation of the point group O_h), the wavefunction of which has the same form as

$$|\Gamma_4^{(i)}\rangle = \begin{cases} A_1^{(i)}(|-4\rangle - |4\rangle) + A_2^{(i)}(|-2\rangle - |2\rangle), \\ B_1^{(i)}|\mp 3\rangle + B_2^{(i)}|\mp 1\rangle \\ \qquad + B_3^{(i)}|\pm 1\rangle + B_4^{(i)}|\pm 3\rangle, \end{cases} \tag{3}$$

where the coefficients $A^{(i)}$'s and $B^{(i)}$'s are not universal but depend on the details of the CEF parameters [4]. Other excited CEF levels have high excitation energies greater than 100K so that their effects are negligible in the low temperature region of the order of T_c.

At temperatures T <10K, these lowest excited CEF levels give a considerable contribution not only to the thermodynamic quantities but also to the NQR relaxation rate $1/T_1$, since the "spin-flip" process can occur between the states forming $\Gamma_4^{(i)}$, e.g., between $|\pm 1\rangle$ and $|\pm 2\rangle$, giving the NQR relaxation. It is noted that each CEF level is broadened due to the hybridization with the conduction electrons so that the energy conservation law is satisfied in the NQR or NMR relaxation process. If the temperature is decreased well below Δ_{23}=11K, these relaxation processes are gradually killed, leading to the pseudo-gap behavior. However, the usual relaxation process due to the quasiparticles of the Fermi liquid still remains. Therefore, T-dependence of $1/T_1$ at $T < T_c$ will be complicated in general.

It is remarked here that the magnetic susceptibility is enhanced even if the mass enhancement arises from the degeneracy of the non-Kramers doublet, electric quadrupolar moment, provided that there exist perturbations breaking the particle-hole symmetry such as the repulsion among conduction electrons, as shown by the numerical renormalization group calculations for the impurity model [7] This is in marked contrast with the case of heavy electrons based on f^2-configuration with the singlet CEF ground state [8].

Thus, we have to be careful with drawing the conclusion on the **k**-dependence of the superconducting gap Δ_k because it is expected that the effect of the CEF level itself gives rise to a considerable part of the quasi-exponential T-dependece as discussed above. Nevertheless, it is quite probable that the superconducting gap Δ_k is finite over the Fermi surface, suggesting that the conventional s-wave pairing or BW-like paring in the odd-parity manifold if the Fermi surface is closed around the Γ-point. Absence of the coherence peak in $1/T_1$ suggests that the pairing is not s-wave although the possibility of the strong coupling effect is not completely ruled out. The strong on-site repulsion, the possible origin of the heavy electron state, cannot be avoided in a manifold of the conventional s-wave pairing state. The BW-like pairing is not promoted either due to a microscopic reason which will be discussed elsewhere.

It is the most natural way to reconcile these seemingly conflictive aspects that the relevant Fermi surface is missing in the direction, in which the superconducting gap vanishes, due to the band effect. Actually, the band structure calculations in $PrOs_4Sb_{12}$ [5] shows that the Fermi surface of the band, which is expected to bocome heavy electrons through the correlation effect, is missing in the direction of [1,1,1] and [1,0,0], or its equivalent as shown in Fig. 1. Then, among the states compatible with the cubic symmetry [6], possible types of pairing are as follows:

$$\hat{\Delta}_k = \Delta[p_x(k) + \varepsilon p_y(k) + \varepsilon^2 p_z(k)]\sigma_z, \tag{4}$$

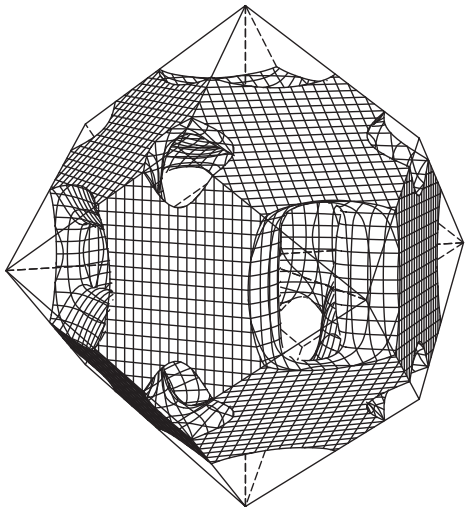

Fig. 1. Fermi surface of $PrOs_4Sb_{12}$ relevant to the heavy electrons given by band structure calculation [5].

$$\hat{\Delta}_k = \Delta[p_x(k) + \mathrm{i}p_y(k)]\sigma_z, \tag{5}$$

in the odd-parity class, and

$$\Delta_k = \Delta[d_{x^2-y^2}(k) + \mathrm{i}d_{z^2}(k)], \tag{6}$$

$$\Delta_k = \Delta[d_{xy}(k) + \varepsilon d_{yz}(k) + \varepsilon^2 d_{zx}(k)], \tag{7}$$

$$\Delta_k = \Delta[d_{xz}(k) + \mathrm{i}d_{yz}(k)], \tag{8}$$

in the even-parity class, respectively. Here, σ_z is the z-component of the Pauli matrix, $\varepsilon \equiv e^{\mathrm{i}2\pi/3}$, $p(k)$'s are bases of irreducible representations with "p"-symmetry, and $d(k)$'s are those with "d"-symmetry. These gaps vanish along the direction of [1,1,1] or [1,0,0], and its equivalent. However, since there exists no Fermi surface in those directions, $1/T_1$ exhibits an exponential T-dependence in general.

This work was supported in part by the Grant-in-Aid for COE Research Program (10CE2004) of the Monbukagaku-sho.

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