

ORIGIN OF THE METAL-INSULATOR TRANSITION IN $\text{PrRu}_4\text{P}_{12}$ *

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$\text{PrRu}_4\text{P}_{12}$ shows a metal-insulator (M-I) transition with a structural phase transition and without magnetic anomaly. To investigate the origin of the M-I transition, we have calculated the band structures by using the FLAPW-LDA+U method with many types of lattice distortions. The result shows an insulator when P ions are slightly distorted with Γ_1 mode, suggesting the M-I transition is caused by the perfect 3 dimensional nesting of the Fermi surface.

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1. Introduction

The filled skutterudites RT_4X_{12} (R=light Rare earth, Yb, Th and U; T=Fe, Ru and Os; X= P, As and Sb) have recently attracted much attention as improved thermoelectric materials [1] and for the variety of the electrical and magnetic properties. Among them, $\text{PrRu}_4\text{P}_{12}$ shows a metal-insulator

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(MI) transition at $T_{\text{MI}} = 60$ K [2], $\text{PrFe}_4\text{P}_{12}$ undergoes a non-magnetic ordering showing heavy fermion behaviour under magnetic fields [3,4]. Very recently, a new class of heavy fermion superconductivity has been reported in $\text{PrOs}_4\text{Sb}_{12}$ [5,6]. Such the interesting physical properties are expected to reflect the Fermi surface property and the $4f^2$ states under the crystalline field. In fact, the Fermi surfaces of RT_4P_{12} (T=Fe, Ru) shows nesting property with $q = (1, 0, 0)$ [7,8].

The phase transitions of $\text{PrFe}_4\text{P}_{12}$ and $\text{PrRu}_4\text{P}_{12}$ have been discussed as anti-quadrupolar ordering [9,10]. However, no distinct magnetic anomaly has been observed at the M-I transition in $\text{PrRu}_4\text{P}_{12}$ [11–14]. Then, it has been suggested that the charge density wave (CDW) is a possible origin of the M-I transition [14]. When the origin of the transition is non-magnetic one, the T_1 -type lattice distortion should be considered [15].

2. Band structure calculations

To investigate the origin of the M-I transition of $\text{PrRu}_4\text{P}_{12}$, band structure calculations are performed by using the FLAPW-LDA+U method [16] with many types of T_1 -type lattice distortions.

$\text{PrRu}_4\text{P}_{12}$ crystallises in a unique body-centered cubic (BCC) structure of a space group $Im\bar{3}$ (T_h^5 , # 204). The atomic parameters are obtained as $a=8.0424$ Å for the lattice constant and $u = 0.3576$, $v = 0.1444$ for $24g$ -site occupied by P [17]. The structural phase transition with $q = (1, 0, 0)$ is observed suggesting that the unit cell becomes doubled to be a simple cubic structure $Pm\bar{3}$ (T_h^1 , # 200) [18]. However, the details of the distortion have remained undetermined experimentally. Now only P distortions are considered. In $Pm\bar{3}$, $12j$ -site ($0, u + \delta_u^A, v + \delta_v^A$) and $12k$ -site ($1/2, 1/2 + u + \delta_u^B, 1/2 + v + \delta_v^B$) are occupied by 24 P ions. When $\delta_u^A = \delta_u^B$ and $\delta_v^A = \delta_v^B$, the lattice becomes back to $Im\bar{3}$. The calculations are performed with a variety of two parameters; $\delta_u = \delta_u^A = -\delta_u^B$ and $\delta_v = \delta_v^A = -\delta_v^B$.

The details of the method follow the previous LDA+U calculation [8]. Now the calculations for the system containing 34 atoms are performed by using up to about 2,100 LAPW basis functions. The local symmetry for Pr-site is unchanged and no magnetic anomaly is observed at T_{MI} , so the occupied $4f$ states are assumed as the singlet ground state ($T_1 = T_7(j = 5/2) \times T_7(j = 5/2)$ in O_h) for the starting potential, then determined self-consistently. The occupied and unoccupied $4f$ states are located below and above the Fermi level, respectively, therefore they do not affect directly the band structure near the Fermi level.

The original Fermi surface in $Im\bar{3}$ has a very good nesting property with the volume of a half of the BCC Brillouin zone [8], however, for many cases a small number of carriers survives when the Fermi surface is nested. There

are two inequivalent Z axes, which are due to no four fold symmetry axes in $Pm\bar{3}$, and a band overlap between bands on the two Z axes tends to remain in the case of $\delta_v = 0$. In the case of $\delta_u = 0$, the degeneracy of the bands around the X points are hardly lifted. Therefore opening a band gap prefer that both of δ_v and δ_u are non zero. Moreover, δ_v and δ_u should have another sign, then will keep the Pr-P distance almost unchanged.

One successful result is obtained, as shown in Fig. 1, for $\delta_u = 0.003a$ and $\delta_v = -0.004a$. Only 0.5% distortion wipes out a whole of the carrier, resulting in an insulator, indicating this is the 3 dimensional *perfect* nesting system. This type of the P distortion must be observed experimentally.

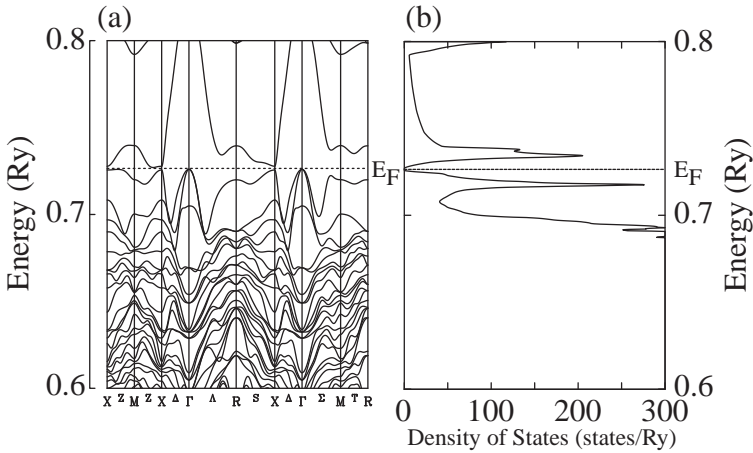


Fig. 1. The calculated (a) band structure and (b) the density of states for $\text{PrRu}_4\text{P}_{12}$ with the Γ_1 -type distortion, with the 1mRy band gap width.

Here we consider the $4f$ states do not participate the phase transition in $\text{PrRu}_4\text{P}_{12}$, assuming the singlet ground state. Therefore such the nesting is expected in another compound, *e.g.*, $\text{LaRu}_4\text{P}_{12}$. However, the existence of the other Fermi surfaces [19] would suppress such the nesting in $\text{LaRu}_4\text{P}_{12}$, while anti-quadrupolar ordering cooperates with the nesting in $\text{PrFe}_4\text{P}_{12}$, though also another Fermi surface appears.

3. Conclusion

The band structure calculations reveal that $\text{PrRu}_4\text{P}_{12}$ could be an insulator with Γ_1 -type P-distortion in the doubled unit cell. It shows the origin of the M-I transition is the 3 dimensional Fermi surface *perfect* nesting.

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