ORIGIN OF THE METAL–INSULATOR TRANSITION IN PrRu$_4$P$_{12}$

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PrRu$_4$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 $T_1$ 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$_4$X$_{12}$ (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, PrRu$_4$P$_{12}$ shows a metal–insulator

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(M1) transition at $T_{MI} = 60$ K [2], PrFe$_4$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 PrOs$_4$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$_4$P$_{12}$ (R=Fe, Ru) shows nesting property with $q = (1,0,0)$ [7,8].

The phase transitions of PrFe$_4$P$_{12}$ and PrRu$_4$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 PrRu$_4$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 $I'_{1}$-type lattice distortion should be considered [15].

2. Band structure calculations

To investigate the origin of the M-I transition of PrRu$_4$P$_{12}$, band structure calculations are performed by using the FLAPW-LDA+U method [16] with many types of $I'_{1}$-type lattice distortions.

PrRu$_4$P$_{12}$ crystallises in a unique body-centered cubic (BCC) structure of a space group $I\bar{m}$3 ($T_{h}^{0}$, # 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 $(0, u + \delta^{A}_{u}, v + \delta^{A}_{v})$ and 12k-site $(1/2, 1/2 + u + \delta^{B}_{u}, 1/2 + v + \delta^{B}_{v})$ are occupied by 24 P ions. When $\delta^{A}_{u} = \delta^{B}_{u}$ and $\delta^{A}_{v} = \delta^{B}_{v}$, the lattice becomes back to $I\bar{m}$3. The calculations are performed with a variety of two parameters; $\delta^{A}_{u} = \delta^{A}_{v} = -\delta^{B}_{u}$ and $\delta^{A}_{u} = \delta^{A}_{v} = -\delta^{B}_{v}$.

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 ($I_{1} = I_{7}(j = 5/2) \times I_{7}(j = 5/2)$ 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 $I\bar{m}$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 $Pm\overline{3}$, 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 $\delta_v = 0$, the degeneracy of the bands around the X points are hardly lifted. Therefore opening a band gap prefer that both of $\delta_v$ and $\delta_u$ are nonzero. Moreover, $\delta_v$ and $\delta_u$ 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 PrRu$_4$P$_{12}$ with the $\Gamma_1$-type distortion, with the 1mRy band gap width.

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

3. Conclusion

The band structure calculations reveal that PrRu$_4$P$_{12}$ could be an insulator with $\Gamma_1$-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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REFERENCES