ORIGIN OF THE METAL–INSULATOR TRANSITION IN $PrRu_4P_{12}$*

Hisatomo Harima

Institute of Scientific and Industrial Research, Osaka University
567-0047 Ibaraki, Japan

Katsuhiko Takegahara

Department of Materials Science and Technology, Hirosaki University
036-8561 Hirosaki, Japan

Kazuo Ueda

Institute for Solid State Physics, University of Tokyo, 277-8581 Kashiwa, Japan

and Stephanie H. Curnoe

Department of Physics and Physical Oceanography
Memorial University of Newfoundland, A1B 3X7 St. John's NF, Canada

(Received July 10, 2002)

$PrRu_4P_{12}$ 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.

PACS numbers: 71.30.+h, 71.20.–b, 71.20.Eh, 71.15.–m

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, $PrRu_4P_{12}$ shows a metal–insulator

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

(MI) transition at $T_{\rm MI} = 60$ K [2], $PrFe_4P_{12}$ 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 $PrOs_4Sb_{12}$ [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 $PrFe_4P_{12}$ and $PrRu_4P_{12}$ have been discussed as anti-quadrupolar ordering [9, 10]. However, no distinct magnetic anomaly has been observed at the M-I transition in $PrRu_4P_{12}$ [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 Γ_1-type lattice distortion should be considered [15].

2. Band structure calculations

To investigate the origin of the M-I transition of $PrRu_4P_{12}$, band structure calculations are performed by using the FLAPW-LDA+U method [16] with many types of Γ_1-type lattice distortions.

$PrRu_4P_{12}$ 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 $T_{\rm MI}$, so the occupied $4f$ states are assumed as the singlet ground state ($\Gamma_1 = \Gamma_7(j = 5/2) \times \Gamma_7(j = 5/2)$ 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.

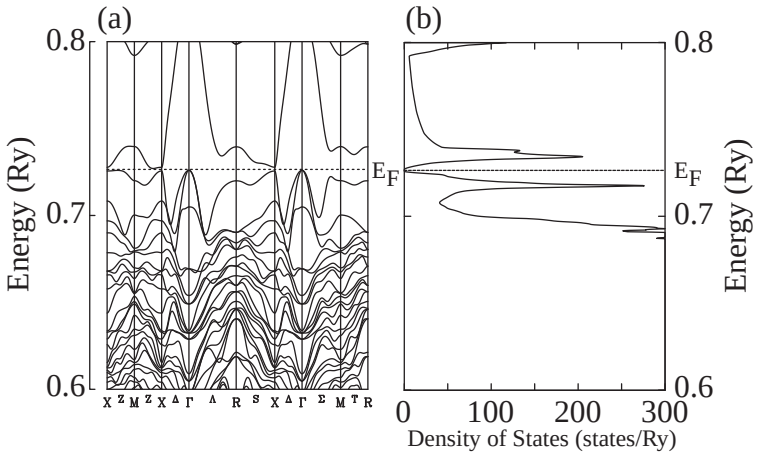

Fig. 1. The calculated (a) band structure and (b) the density of states for $PrRu_4P_{12}$ with the Γ_1-type distortion, with the 1mRy band gap width.

Here we consider the $4f$ states do not participate the phase transition in $PrRu_4P_{12}$, assuming the singlet ground state. Therefore such the nesting is expected in another compound, *e.g.*, $LaRu_4P_{12}$. However, the existence of the other Fermi surfaces [19] would suppress such the nesting in $LaRu_4P_{12}$, while anti-quadrupolar ordering cooperates with the nesting in $PrFe_4P_{12}$, though also another Fermi surface appears.

3. Conclusion

The band structure calculations reveal that $PrRu_4P_{12}$ 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.

This work was partly supported by a Grant-in-Aid for Science Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

REFERENCES

[1] G.D. Mahan, *Solid State Physics*, Vol. 51, ed. H. Ehreinreich, F. Spaepen, Academic Press, New York 1998, p. 81.

[2] C. Sekine, T. Uchiumi, I. Shirotani, T. Yagi, *Phys. Rev. Lett.* **79**, 3218 (1997).

[3] H. Sato, Y. Abe, H. Okada, T.D. Matsuda, H. Sugawara, Y. Aoki, *Phys. Rev.* **B62**, 15125 (2000).

[4] H. Sugawara, T.D. Matsuda, K. Abe, Y. Aoki, H. Sato, S. Nojiri, Y. Inada, R. Settai and Y. Ōnuki, *J. Magn. Magn. Mater.* **226-230**, 48 (2001).

[5] E.D. Bauer, N.A. Frederick, P.-C. Ho, V.S. Zapf, M.B. Maple, *Phys. Rev.* **B65**, 100506(R) (2002).

[6] M.B. Maple, P.-C. Ho, V.S. Zapf, N.A. Frederick, E.D. Bauer, Y.M. Yuhasz, F.M. Woodward, J.W. Lynn, *J. Phys. Soc. Jpn.* **71**, Suppl. 23 (2002).

[7] H. Sugawara, Y. Abe, Y. Aoki, H. Sato, M. Hedo, R. Settai, Y. Ōnuki, H. Harima, *J. Phys. Soc. Jpn.* **69**, 2938 (2000).

[8] H. Harima, K. Takegahara, *Physica B* **312&313**, 843 (2002).

[9] S.H. Curnoe, H. Harima, K. Takegahara, K. Ueda, *Physica B* **312&313**, 837 (2002).

[10] H. Harima, K. Takegahara, S.H. Curnoe, K. Ueda, *J. Phys. Soc. Jpn.* **71** Suppl. 70 (2002) .

[11] C. Sekine, T. Inaba, K. Kihou, I. Shirotani, *Physica B* **281&282**, 300 (2000).

[12] C. Sekine, T. Inaba, I. Shirotani, M. Yokoyama, H. Amitsuka, T. Sakakibara, *Physica B* **281&282**, 303 (2000).

[13] K. Matsuhira, T. Takikawa, T. Sakakibara, C. Sekine, I. Shirotani, *Physica B* **281&282**, 298 (2000).

[14] K. Matsuhira, Y. Hinatsu, C. Sekine, I. Shirotani, *Physica B* **312&313**, 829 (2002).

[15] S.H. Curnoe, K. Ueda, H. Harima, K. Takegahara, *J. Phys. Chem. Solids* **63**, 1207 (2002).

[16] H. Harima, *J. Magn. Magn. Mater.* **226-230**, 83 (2001).

[17] C. Sekine, private communications.

[18] C.H. Lee, H. Matsuhata, A. Yamamoto, T. Ohta, H. Takazawa, K. Ueno, C. Sekine, I. Shirotani, T. Hirayama, *J. Phys.: Condens. Matter* **13**, L45 (2001).

[19] H. Harima, *Prog. Theor. Phys. Suppl.* **138**, 117 (2000).