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FLAPW Band Structure Calculations and the Fermi Surfaces in Rare-Earth Compounds

Hisatomo HARIMA

Department of Condensed Matter Physics, The Institute of Scientific and Industrial Research
Osaka University, Ibaraki, Osaka 567-0047

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In order to obtain Fermi surfaces and compare them with experimental results, FLAPW band structure calculations are performed for LaAg, $LaCu_2$, $LaPb_3$, $LaNi_2Al_5$, $YbSn_3$, $YbPb_3$, $YbSb_2$, $LaFe_4P_{12}$ and $La_3Pd_{20}Ge_6$. The results show fairly good agreement with experiments especially in LaAg, $LaCu_2$ and $YbSn_3$. Full potential treatment is important for LaAg, $LaCu_2$, $LaFe_4P_{12}$ and $La_3Pd_{20}Ge_6$.

KEYWORDS: Fermi surfaces, Band theory, FLAPW method, Rare-earth compounds, de Haas-van Alphen effects

§1. Introduction

Recently lots of Fermi surface studies have been performed on various rare-earth and actinide compounds.[1] To clarify electronic structures and Fermi surfaces, band structure calculations have been carried out by various methods. Among them, a full potential LAPW (FLAPW) method is one of the most reliable methods within the local density approximation. In fact, the FLAPW method succeeds in giving a good description about Fermi surface topology for hexagonal $NbSe_2$.[2] The result shows that the muffin-tin approximation (MTA) underestimates the relative energy in Nb d-bands and Se p-bands and does not give Fermi surfaces corresponding to the measured dHvA effects. In this paper, the results by LAPW (using the MTA) and FLAPW band structure calculations of various rare-earth compounds are compared with the dHvA measurements.

§2. Method of Calculation

Band structure calculations are carried out using an FLAPW method with the local density approximation (LDA) for the exchange correlation potential. For the LDA, the formula proposed by Gunnarsson and Lundqvist[3] is used. For the band structure calculation, we used the program codes; TSPACE[4] and KANSAI-94.[5] The scalar relativistic effects are included for all electrons and the spin-orbit interactions are included for valence electrons as a second variational procedure.

§3. Lanthanum Compounds

In LaAg and $LaCu_2$, the FLAPW calculations show different Fermi surfaces with the LAPW calculations. LaAg has a small electron pocket around the X point. The size of the Fermi surfaces is very sensitive to the relative energy of La d-bands and Ag s-bands and depends on the choice of the muffin-tin radii within the MTA. The FLAPW result has succeeded in predicting the correct size of the Fermi surfaces.[6] In $LaCu_2$, full potential treatment improves the band structure in the vicinity of the H point in the hexagonal Brillouin zone, resulting topologically different Fermi surfaces. The agreement of the angular dependence with the dHvA frequencies is quite satisfying.[7]

Both in $LaPb_3$ and $LaNi_2Al_5$, significant changes of Fermi surfaces are not found in the FLAPW calculations. However, a slight change is necessary to explain the dHvA measurement of $LaPb_3$[8] and $PrPb_3$,[9] probably because the LDA overestimates hybridisation in the Σ axis in $LaPb_3$. The FLAPW Fermi surfaces of $LaNi_2Al_5$ have been compared with the dHvA measurements of $CeNi_2Al_5$.[10] The dHvA frequencies in higher magnetic fields can be explained well by the Fermi surfaces of $LaNi_2Al_5$. More detailed comparison with the recent dHvA measurements of $LaNi_2Al_5$ is now in progress.[11]

§4. Ytterbium Compounds

In $YbSn_3$, $YbPb_3$ and $YbSb_2$, Yb atoms behaves as +2 with the closed $4f$ shell. Band structures of such

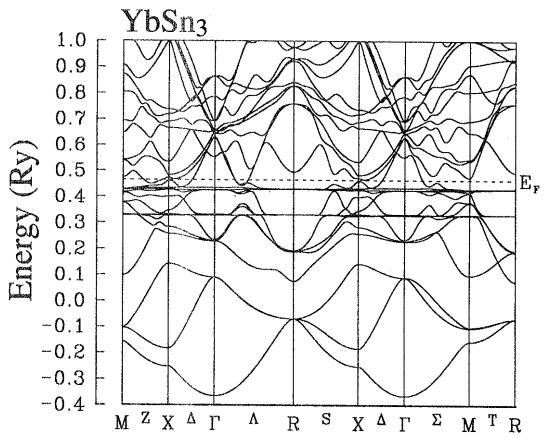

Fig. 1. FLAPW energy band structure of $YbSn_3$. The Fermi level 0.4652 Ry is denoted by E_F. Yb $4f$-bands are situated at around 0.42 Ry ($j = 7/2$) and 0.33 Ry ($j = 5/2$).

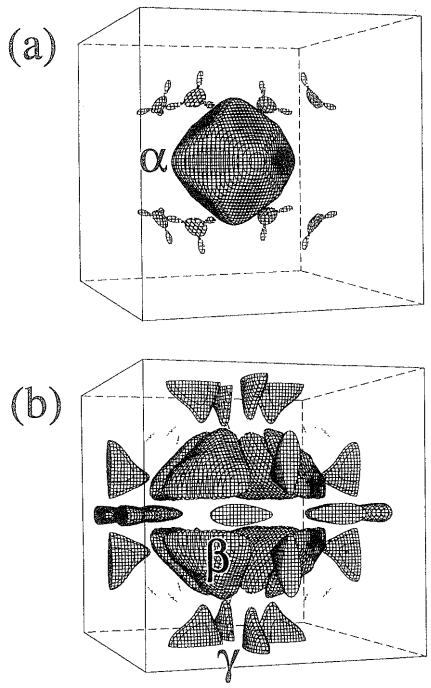

Fig. 2. (a) The 14th hole Fermi surface and (b) the 15th electron Fermi surfaces in $YbSn_3$.

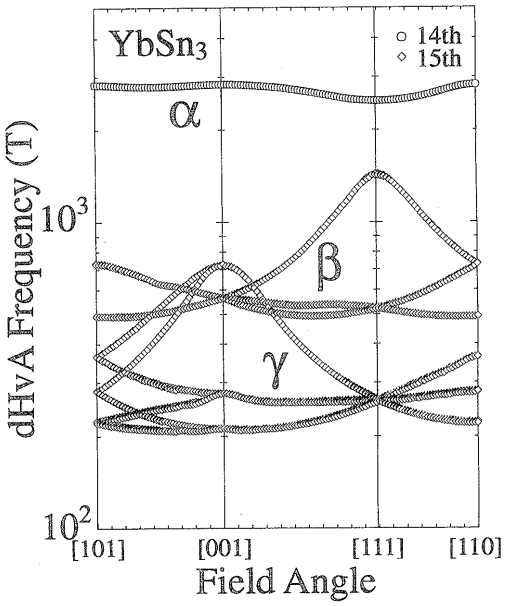

Fig. 3. Calculated angular dependence of the dHvA frequencies in $YbSn_3$.

non-magnetic Yb-compounds, as shown in Fig. 1, Yb $4f$-bands occasionally lie very close to the Fermi level to affect Fermi surfaces due to the insufficient treatment of Yb $4f$-electrons in the LDA. Then the energy level of Yb can be determined by a slight change of a number of the occupied $4f$ electrons. Actually the number of $4f$ electrons in $YbPb_3$ is increased to be 13.91 in the FLAPW method from 13.75 in the LAPW method, in which the $4f$-bands are closer to the Fermi level. In $YbSn_3$ and $YbSb_2$, the number is unchanged and both methods give similar Fermi surfaces.

Figures 2 and 3 show Fermi surfaces and the angular dependence of the dHvA frequencies of $YbSn_3$. The result shows fairly good agreement with experiments.[12] Therefore influence of the $4f$-bands is negligible in this case.

In the dHvA measurements of $YbPb_3$, one branch is very similar to α branch of $YbSn_3$ in Fig. 3, but the other branches are different,[13] indicating that the electron Fermi surfaces are modified by the spin-orbit interactions in $YbPb_3$. Downward shift of $4f$-bands is found ineffective in $YbPb_3$. These branches in the dHvA measurements can be explained by model Fermi surfaces based on the FLAPW method.[13]

In band structure calculations of $YbSb_2$, the both methods obtain Yb $4f$-bands lying very close to the Fermi level, which affect Fermi surfaces. Then we have carried out another self-consistent calculation with $4f$-level artificially shifted downward by 0.2 Ry, which gives Yb $4f_{j=7/2}$ level 0.1 Ry below the Fermi level after self-consistent step. The calculated Fermi surfaces give a good description of the dHvA experiments.[14]

§5. $LaFe_4P_{12}$ and $La_3Pd_{20}Ge_6$

5.1 $LaFe_4P_{12}$

$LaFe_4P_{12}$ belongs to ternary metal phosphides MT_4P_{12} with the skutterudite structure filled by a lanthanide or actinide atom. The space group is #204, T_h^5. The dHvA signals have been observed for $LaFe_4P_{12}$ and $NdFe_4P_{12}$.[15] The band structure calculations of $LaFe_4P_{12}$ have been briefly reported.[16]

The dHvA results indicate $LaFe_4P_{12}$ has a nearly spherical Fermi surface and a multiply connected one. In the LAPW result, only the 48th band, originating mainly from P p-states, cuts the Fermi energy, while in the FLAPW result the 47th band from Fe d-states cuts it as well, because Fe d-bands are shifted upward slightly in the latter. Both of the 48th hole Fermi surfaces are multiply connected and the 47th Fermi surface given by the FLAPW is spherical. Therefore, the FLAPW result explains the measurement better. The detailed results both of the experiments and the calculations will be published.[17]

5.2 $La_3Pd_{20}Ge_6$

The dHvA effects are measured in $Ce_3Pd_{20}Ge_6$[18] and $La_3Pd_{20}Ge_6$.[19] The angular dependence of the dHvA frequencies are similar indicating the $4f$ electrons in $Ce_3Pd_{20}Ge_6$ are localized.

The space group of the crystal structure is #225, O_h^5 where 4a-site and 8c-site are occupied by La atoms, 32f-site and 48h-site by Pd atoms and 24e-site by Ge atoms. Therefore the unit cell contains 116 atoms/cell corresponding to 29 atoms/primitive cell.

A large number of valence electrons requires lots of basis functions in calculations. The LAPW calculations with up to 2000 basis functions and the FLAPW calcu-

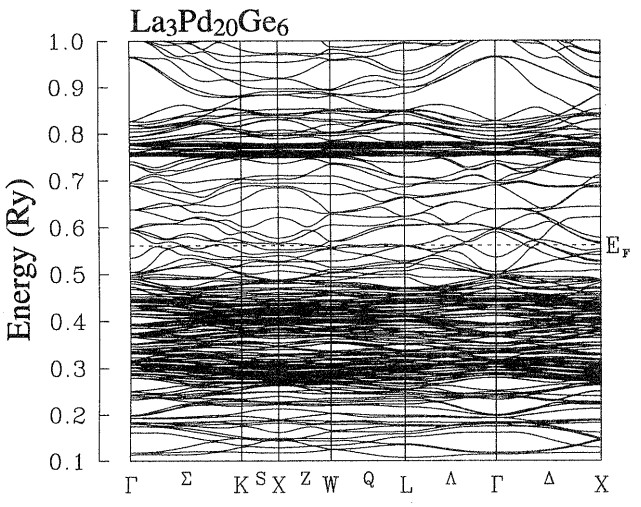

Fig. 4. FLAPW energy band structure of $La_3Pd_{20}Ge_6$. The Fermi level 0.5608 Ry is denoted by E_F

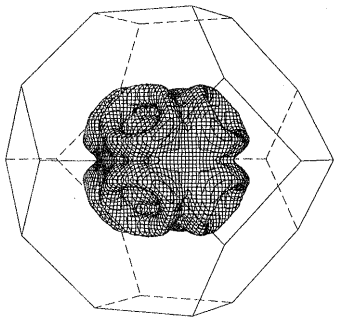

Fig. 5. The 116th hole Fermi surface in $La_3Pd_{20}Ge_6$.

lation with up to 1500 basis functions have been carried out. In the LAPW band structure, unoccupied La-I (4a) $4f$-bands and La-II (8c) $4f$-bands are situated around 0.1 Ry and 0.3 Ry above the Fermi level, respectively. While both unoccupied La $4f$-bands are located around 0.2 Ry above the Fermi level in the FLAPW result, as shown in Fig. 4. Then Fermi surfaces are quite different in the two results.

In the FLAPW result, there are mainly two kinds (the 116th and the 117th) of closed hole Fermi surfaces around the Γ point and one small closed electron Fermi surface (the 118th) around the X point. Angular dependence of the dHvA frequencies indicates that the 116th hole Fermi surface, as shown in Fig. 5, corresponds to the experimental frequencies around 1×10^7Oe. In the experiments,[19] frequencies larger than 2.3×10^7Oe have not been detected so far. However, the main frequencies from the 117th Fermi surface, which is the largest, are calculated to be around 4-5 $\times 10^7$Oe. $La_3Pd_{20}Ge_6$ is an uncompensated metal. The 116th hole, the 117th hole and the 118th electron Fermi surfaces have 6.5%, 44% and 0.5% in volume in the Brillouin zone. Therefore, larger frequencies should exist, originating in the 117th Fermi surface.

§6. Conclusion

The FLAPW calculations have been carried out for various rare-earth compounds. Through comparison of Fermi surfaces with the experiment, significant improvement is found in FLAPW calculations of LaAg, $LaCu_2$, $LaFe_4P_{12}$ and $La_3Pd_{20}Ge_6$.

Acknowledgements

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