Electronic properties of transuranium compounds with HoCoGa₅-type tetragonal crystal structure

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Abstract. By using a relativistic linear augmented-plane-wave method with the one-electron potential in the local-density approximation, we investigate energy-band structures and the Fermi surfaces of transuranium compounds NpTG₅, PuTG₅ and AmCoGa₅ with transition metal atoms T. It is found that the energy bands in the vicinity of the Fermi level are mainly due to the large hybridization between 5f and Ga 4p electrons. For PuTG₅, we observe several cylindrical sheets of Fermi surfaces with large volume for T = Co, Rh and Ir. The de Haas–van Alphen (dHvA) frequencies are theoretically estimated for PuCoGa₅. It is also found that the Fermi surfaces of NpFeGa₅, NpCoGa₅ and NpNiGa₅ are similar to those of UCoGa₅, UNiGa₅ and PuCoGa₅, respectively, except for small details. For AmCoGa₅, the Fermi surfaces are found to consist of large cylindrical electron sheets and small closed hole sheets, similar to PuCoGa₅. The similarity is basically understood by the change of electron numbers inside the Fermi surfaces on the basis of a rigid-band picture. We compare our theoretical Fermi surfaces with the experimental results on NpTG₅.
1. Introduction

Recently, electronic properties of actinide compounds have attracted renewed attention in the research field of condensed matter physics. Among numerous kinds of actinide compounds, the material group with HoCoGa$_5$-type tetragonal crystal structure, referred to as ‘115’, has been intensively investigated both from experimental and theoretical sides. A characteristic trend in the recent investigation of actinide compounds has been a rapid expansion of the research frontier from uranium to transuranium materials, typically found in a series of actinide 115 systems.

First, let us briefly survey the properties of U-115. For several transition metal ions T, UTGa$_5$ are antiferromagnetic (AF) metals or Pauli paramagnets [1]–[12]. Among them, neutron scattering experiments have revealed that UNiGa$_5$ exhibits the G-type AF phase, while UPdGa$_5$ and UPtGa$_5$ have the A-type AF state [8, 12]. Note that G-type indicates a three-dimensional Néel state, while A-type denotes a layered AF structure in which spins align ferromagnetically in the $ab$-plane and AF along the $c$-axis. On the other hand, for T = Co, Rh, Ir, Fe, Ru and Os, magnetic susceptibility is almost independent of temperature, since these are Pauli paramagnets. It is quite interesting that the magnetic structure is different for U-115 compounds which differ only by the substitution of transition metal ions.

Now we move on to transuranium 115 compounds. The rapid progress in the research from uranium to transuranium systems has been triggered by the discovery of superconductivity of Pu-115 compound PuCoGa$_5$ [13, 14]. It has been reported that the superconducting transition temperature $T_c$ of PuCoGa$_5$ is 18.5 K, which is an amazingly high value even compared with other well-known intermetallic compounds. The coefficient of electronic specific heat $\gamma$ is estimated as $\gamma = 77 \text{ mJ mol}^{-1} \text{ K}^{-2}$, moderately enhanced relative to that for normal metals, suggesting that PuCoGa$_5$ is considered to be a heavy-fermion superconductor, but with moderately enhanced effective mass. In PuRhGa$_5$, superconductivity has been also found [15, 16]. Although the value of $T_c = 8.7 \text{ K}$ is lower than that of PuCoGa$_5$, it is still high enough compared with other heavy-fermion superconductors. Another Pu-115 material, PuIrGa$_5$, has been also synthesized, but up to 10 GPa and down to 1.4 K, superconductivity has not been found [17].

Recently, Np-115 compounds NpTGa$_5$ (T = Fe, Co, Rh and Ni) have been synthesized and several kinds of physical quantities have been successfully measured [18]–[27]. Especially,
the de Haas–van Alphen (dHvA) effect has been observed in NpNiGa$_5$ [19], which is the first observation of dHvA signal in transuranium compounds. For NpCoGa$_5$ and NpRhGa$_5$, the dHvA oscillations have been also detected and plural number of cylindrical Fermi surfaces are found [21, 22]. For NpFeGa$_5$, the magnetic moment at Fe site has been suggested in neutron scattering experiments [23] and it has been also detected by $^{57}$Fe Mössbauer spectroscopy [25]. The magnetic structure of Np-115 compounds also depends sensitively on transition metal ion [23, 26, 27]: G-AF for NpNiGa$_5$, A-AF for NpCoGa$_5$ and NpRhGa$_5$, and C-AF for NpFeGa$_5$. Here C-type indicates a situation in which the ferromagnetic chains along the $c$-axis are antiferromagnetically coupled in the $ab$-plane. Note also that in the neutron scattering experiment for NpNiGa$_5$, the G-AF peak due to canted magnetic moments of Np ions grows after the FM transition occurs [23]. It is characteristic of U-115 and Np-115 compounds that the magnetic properties are sensitive to the choice of transition metal ions.

Quite recently, the experimental research on actinide 115 systems has been further developed and the frontier has reached the americium compound AmCoGa$_5$ [28]. Interestingly enough, from the resistivity measurement, superconductivity has been suggested to occur below 2 K, but unfortunately, it has not been confirmed yet by other experimental techniques, mainly due to the difficulty caused by the self-heating effect. It may be interesting to consider the possibility of superconductivity in Am-115 also from the theoretical viewpoint.

In order to elucidate the mechanism of superconductivity and magnetism of such actinide compounds, it is necessary to develop a microscopic theory based on an appropriate $f$-electron model. A prescription to construct a microscopic $f$-electron model has been obtained on the basis of a $j$-$j$ coupling scheme [29]. The spirit of the prescription is that the many-body effect is further included into the band-structure calculation results. In actual calculations, since the energy dispersion obtained by the band-structure calculations are very complicated, the $f$-electron kinetic term is simply reconstructed by using a tight-binding approximation based on the $j$-$j$ coupling scheme so as to reproduce the band structure around the Fermi energy. Here, we note that all 5 $f$ electrons are assumed to be itinerant. Note also that it is quite natural to use the $j$-$j$ coupling scheme for the construction of the microscopic $f$-electron model, since the total angular momentum $j$ is the label for one-electron state in the relativistic band-structure calculations in which the Dirac equations are directly solved.

By applying the prescription to uranium compound such as U-115, it has been clarified that electronic structure of uranium compounds is effectively described by a two-orbital Hubbard model based on the $j$-$j$ coupling scheme [30]. With increasing $f$-electron number, it has been pointed out that this two-orbital Hubbard model can be also applied to some neptunium and plutonium compounds. For instance, quite recently, magnetic structure of Np-115 [31] and octupole ordering of NpO$_2$ [32]–[34] have been discussed based on the two-orbital Hubbard model.

Although it is important to develop the microscopic analysis of actinide compounds based on the simple $f$-electron model, close attention should be always paid to correct information about the electronic properties around the Fermi energy obtained by the relativistic band-structure calculations. Those two types of researches, microscopic analysis of the simple $f$-electron model and band-structure calculations, should be complementary to each other in order to make significant progress in our understandings on magnetism and superconductivity of actinide compounds.

In this paper, we study electronic properties of neptunium, plutonium and americium compounds such as NpTGa$_5$, PuTGa$_5$ and AmCoGa$_5$ by using a relativistic linear
augmented-plane-wave (RLAPW) method with the one-electron potential in the local-density approximation (LDA). We assume that all $5f$ electrons are itinerant in our treatment. Furthermore, in order to clarify the variation of the Fermi-surface structure due to the change of the band filling originating from the change of $d$- and $f$-electron numbers of transition metal and actinide ions, we simply assume the non-magnetic phase in our calculations, although AF phases have been found in some actinide 115 compounds. We observe that the energy bands in the vicinity of Fermi level are given by the large hybridization between $5f$ and Ga 4$p$ electrons in actinide 115 compounds. For PuTGa$_5$, several cylindrical sheets of Fermi surfaces with large volume are found in common for $T =$ Co, Rh and Ir. For Np-115, we find that the Fermi surfaces of NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$ are similar to those of UCoGa$_5$, UNiGa$_5$ and PuCoGa$_5$, respectively. It is also found that the Fermi surfaces of AmCoGa$_5$ are rather similar to those of PuCoGa$_5$. Basically, the similarity is understood by the rigid-band nature of the 115 structure, since the topology of the Fermi surfaces in the band-structure calculations seems to depend on the sum of $f$-electron number of actinide ions and $d$-electron number of transition metal ions.

The organization of this paper is as follows. In section 2, we briefly explain the method of our band-structure calculations and lattice parameters for PuTGa$_5$, NpTGa$_5$ and AmCoGa$_5$. In section 3, we show the results for electronic band structure and the Fermi surfaces of PuTGa$_5$ ($T =$ Co, Rh and Ir), NpTGa$_5$ ($T =$ Fe, Co and Ni) and AmCoGa$_5$. In section 4, we will discuss our present results of Pu-, Np- and Am-115 materials in comparison with that of UCoGa$_5$. We also discuss the dHvA results on NpTGa$_5$ from the band-theoretical viewpoint. Finally, in section 5, we will summarize this paper.

2. Method of band calculation

In order to calculate the electronic energy-band structure of transuranium compounds, in general, it is necessary to include relativistic effects such as relativistic energy shifts, relativistic screening effects and spin–orbit interaction [35]. Based on the Dirac equation, such relativistic effects can be fully taken into account in the energy-band structure calculation [36]–[38]. Among several kinds of band-structure calculation techniques, in this paper we adopt the RLAPW method [39]. The exchange and correlation potential is considered within the LDA, while the spatial shape of the one-electron potential is determined in the muffin-tin approximation. The radius of the APW spheres are chosen to be as large as possible, as long as they do not overlap with each other. The self-consistent calculation is performed by using the lattice constants which are determined experimentally. It is stressed that all $5f$ electrons in PuTGa$_5$, NpTGa$_5$ and AmCoGa$_5$ are assumed to be itinerant in our band-structure calculations. Again we note that the non-magnetic state is assumed in our calculations.

In figure 1, we show the HoCoGa$_5$-type tetragonal crystal structure which belongs to the space group P4/mmm (no 123) and D$_{4h}^1$. Note that one molecule is contained per primitive cell. In the HoCoGa$_5$-type tetragonal structure, positions of atoms in the unit cell are given by (0, 0, 0) for the actinide atom, (0, 0, 1/2) for the transition metal atom, (1/2, 1/2, 0) for the Ga atom at 1$c$ site and (0, 1/2, ±$z$) for the Ga atom at 4$i$ site, where $z$ is a parameter determined from x-ray diffraction experiments and the labels for atoms are referred to in figure 1. The lattice constants, $a$ and $c$ and the $z$ parameters for NpTGa$_5$, PuTGa$_5$ and AmCoGa$_5$ are listed in table 1.

The iteration process for solving the Dirac one-electron equation starts with the crystal charge density that is constructed by superposing the relativistic atomic charge densities for
In each iteration step for the self-consistent calculation processes, a new crystal charge density is constructed using 18 \( k \) points, which are uniformly distributed in the irreducible 1/16 Brillouin zone.
part of the Brillouin zone. At each $k$ in the Brillouin zone, 431 plane waves are adopted under the condition $|k + G| \leq 4.4(2\pi/a)$ with $G$ the reciprocal lattice vector and angular momentum up to $\ell_{\text{max}} = 8$ are taken into account.

3. Band calculation results

3.1. Overview

Before proceeding to the exhibition of the band calculation result for each compound, let us take a survey of the energy-band structure in common with actinide 115 materials. In general, in the relativistic energy band calculation, the spin–orbit interaction makes the narrow 5$f$ bands split into two subbands, corresponding to the total angular momentum $j = 5/2$ (lower bands) and $7/2$ (upper bands). The magnitude of the splitting between those groups is estimated as $\sim 1.0$ eV for actinide 115 compounds, which is almost equal to the spin–orbit splitting in the atomic 5$f$ state.

Concerning the energy-band structure in the vicinity of $E_F$, we emphasize that there always occurs a large hybridization between actinide 5$f$ and Ga 4$p$ states occurs, which is an important common feature of 115 compounds. Since the Bloch states on the Fermi surface mainly consist of 5$f$ and 4$p$ electron states, the Fermi level $E_F$ should be adjusted in order to change 5$f$ electron number. Thus, it is basically possible to explain the Fermi-surface structure of actinide 115 compounds based on the so-called $f$–$p$ model [40] by simply changing the Fermi level for 5$f$ electron number. In fact, as we will see later, the Fermi-surface structure is understood by simply changing $E_F$ in the same energy-band structure. It is a characteristic point of actinide 115 materials that such a rigid-band picture holds. The similarity and difference among 115 materials are discussed in the following subsections.

3.2. PuTGa$_5$

First, let us discuss the calculated results for Pu-115 compounds, since PuCoGa$_5$ and PuRhGa$_5$ have been recently focused as ‘high-$T_c$’ $f$-electron superconductors. In particular, the possibility of $d$-wave pairing has been discussed intensively from NMR measurements [41, 42]. Note that a part of our band-calculation results of PuCoGa$_5$ has been already published [43]. Band-structure calculation results for PuTGa$_5$ ($T =$ Co, Rh and Ir) were also reported by another group [44, 45].

Among Pu-115 materials, first we focus on PuCoGa$_5$. In figure 2(a), we depict the energy-band structure along the symmetry axes in the Brillouin zone in the energy region from $-0.5$ to $1.0$ Ryd. Three Pu 6$p$ and 25 Ga 3$d$ bands in the energy range between $-1.0$ and $-0.6$ Ryd are not shown in figure 2, since those bands are irrelevant to the present discussion. The Fermi level $E_F$ is located at 0.446 Ryd. For other 115 materials, we will not show the figure of the energy-band structure such as figure 2(a), since the overall structure observed in the order of Ryd is almost the same among 115 materials. In the following, we show only the fine structure around the Fermi level, for the discussion on the Fermi-surface structure.

The number of the valence electrons in the APW sphere is partitioned into the angular momenta as listed in table 3. There are 8.17 valence electrons outside the APW sphere in the primitive cell. The total density of states (DOS) at $E_F$ is evaluated as $N(E_F) = 97.3$ states Ryd cell$^{-1}$. By using this value, the theoretical specific heat coefficient $\gamma_{\text{band}}$ is
Figure 2. (a) Energy-band structure for PuCoGa$_5$ calculated by using the self-consistent RLAPW method. $E_F$ indicate the position of the Fermi level. (b) Energy-band structure near the Fermi level in a magnified scale.

Table 3. The number of valence electrons for PuCoGa$_5$ in the Pu APW sphere, the Co APW sphere and the Ga APW sphere partitioned into angular momenta.

|    | s    | p    | d    | f    |
|----|------|------|------|------|
| Pu | 0.39 | 6.15 | 1.81 | 5.24 |
| Co | 0.43 | 0.44 | 7.46 | 0.01 |
| Ga(1c) | 0.95 | 0.69 | 9.92 | 0.01 |
| Ga(4i) | 3.73 | 2.82 | 39.75 | 0.06 |

estimated as 16.9 mJ K$^{-2}$ mol$^{-1}$. We note that the experimental electronic specific heat coefficient $\gamma_{\text{exp}}$ is 77 mJ K$^{-2}$ mol$^{-1}$ [13]. We define the enhancement factor for the electronic specific heat coefficient as $\lambda = \gamma_{\text{exp}}/\gamma_{\text{band}} - 1$, and in the present case, we obtain $\lambda = 3.6$. The disagreement between $\gamma_{\text{band}}$ and $\gamma_{\text{exp}}$ values is ascribed to electron correlation effect and electron–phonon interactions, which are not fully taken into account in the present LDA band theory.

Now we discuss the Fermi surfaces of PuCoGa$_5$. In figure 2, the lowest 14 bands are fully occupied. The next four bands are partially occupied, while higher bands are empty. Then, as shown in figure 2(b), 15th, 16th, 17th and 18th bands cross the Fermi level and they construct the hole or electron sheet of the Fermi surface. The Fermi surfaces are shown in figure 3(a). The Fermi surface from the 15th band consists of one hole sheet centred at the $\Gamma$ point. The 16th band has two kinds of sheets. One is a set of small hole pockets, each of which is centred at the X point. Another is a large cylindrical hole sheet which is centred at the $\Gamma$ point. The 17th band forms a large cylindrical electron sheet which is centred at the M point. The 18th band has a slender cylindrical electron sheet which is also centred at the M point. These electron sheets are characterized by two-dimensional Fermi surfaces. The number of carriers contained in these Fermi-surface sheets are 0.040, 0.563 holes cell$^{-1}$, 0.519 and 0.084 electrons cell$^{-1}$ in the 15th, 16th, 17th and 18th bands, respectively. The total number of holes is equal to that of electrons, which indicates that PuCoGa$_5$ is a compensated metal. Note that the Fermi surfaces of PuTGa$_5$ are similar to those of CeTIn$_5$ [40]. This similarity can be understood based on the electron–hole
relation in the $j$-$j$ coupling scheme \cite{43}, since one $f$ electron is included in the $j = 5/2$ sextet for Ce$^{3+}$ ion, while five $f$ electrons are contained for Pu$^{3+}$ ion.

In figure 3(a), we also depict various kinds of extremal orbits on the Fermi surfaces. Among them, $a$ denotes an orbit running around the small closed hole sheet centred at the $\Gamma$ point in the 15th band, while $b_1$ and $b_2$ indicate a couple of dHvA frequency branches which originate from the 16th hole sheets and they exist in the narrow range between $1.5 \times 10^7$ and $3 \times 10^7$ Oe. Their centre is located at the X point. On the other hand, $\alpha_1$ and $\alpha_2$ are the orbits running around the cylinder along the V-axis in the 17th band, while $\beta_1$, $\beta_2$ and $\beta_3$ are the orbits running around the cylinder along the $\Lambda$-axis in the 16th band. Finally, $\gamma_1$, $\gamma_2$ and $\gamma_3$ denote the orbits running around the cylinder along the V-axis in the 18th band. These orbits exist in the large range of angles in the vicinity of the [0 0 1] direction.

In figure 3(b), we show the angular dependence of the theoretical dHvA frequency in PuCoGa$_5$. The area of the extremal cross-section of the Fermi surface $A$ is related to the dHvA frequency $F$ by the well-known formula $F = (c\hbar/2\pi)A$. The Fermi surface produces many dHvA frequencies in the wide frequency range between $1 \times 10^7$ and $22 \times 10^7$ Oe, as shown in figure 4. The small hole sheet in the 15th band possesses dHvA frequencies in the range between $1.5 \times 10^7$ and $2 \times 10^7$ Oe. The Fermi surface in the 16th, 17th and 18th band possesses many extremal cross-section in the limited range of angles because of its rugged shape. Due to the similarity of the Fermi-surface structure, the branches labelled by $\alpha_i$, $\beta_i$, $a$ and $b_i$ ($i = 1$ and 2) are the same as those of CeTIn$_5$ \cite{46, 47}. The angular dependence of the branch $\alpha_i$ of PuCoGa$_5$ is similar to that of the branch $\alpha_i$ of CeTIn$_5$, although the magnitude of the branch $\alpha_i$ of PuCoGa$_5$ is slightly larger than that of CeTIn$_5$. The branch $\beta_i$ of PuCoGa$_5$ with the $\Gamma$-point centre exhibits the angular dependence similar to that of the branch $\beta_i$ of CeTIn$_5$ with the M-point centre, although the magnitude of the branch $\beta_i$ of PuCoGa$_5$ is a little larger than that of CeTIn$_5$. We suggest that the branch $a$ and $b_i$ of PuCoGa$_5$ may correspond to $g(\epsilon)$ and $h(\gamma)$ of CeIrIn$_5$(CeCoIn$_5$),
respectively. The dHvA experiments on Pu-115 compounds have not yet been carried out due to several experimental difficulties, but we believe that such experiments will be done in the near future. On that occasion, the present theoretical results will be helpful for the experimentalists.

Here, we briefly mention the result of Shick et al [48] using a relativistic local spin density approximation + Hubbard $U$ (LSDA + $U$) method. In order to consider the effect of local moment in the paramagnetic phase, the LSDA + $U$ result in the AF phase has been compared with that of the non-magnetic LDA calculation. Their treatment on the AF phase is based on a semi-localized 5$f$-electron model. All the Fermi-surface sheets calculated by the LSDA + $U$ method are closed in topology, in sharp contrast to the cylindrical Fermi-surface sheets from non-magnetic LDA calculations.
Table 4. The number of valence electrons for PuCoGa$_5$ in the Pu APW sphere, the Co APW sphere and the Ga APW sphere partitioned into angular momenta.

|   | s   | p  | d   | f  |
|---|-----|----|-----|----|
| Pu| 0.33| 6.16| 1.85| 5.22|
| Rh| 0.35| 0.35| 7.38| 0.02|
| Ga(1c)| 0.99| 0.75| 9.94| 0.01|
| Ga(4i)| 3.73| 2.84| 39.77| 0.07|

Now we consider other Pu-115 materials, PuRhGa$_5$ and PuIrGa$_5$. In figures 4(a) and (b), we depict the energy-band structure near the Fermi energy for PuRhGa$_5$ and PuIrGa$_5$, respectively. Note that the values of the Fermi level are 0.436 and 0.453 Ryd for PuRhGa$_5$ and PuIrGa$_5$, respectively. In table 4, we list the number of the valence electrons in the APW sphere distributed into the angular momenta. There are 8.31 and 8.64 valence electrons outside the APW sphere in the primitive cell for PuRhGa$_5$ and PuIrGa$_5$, respectively. The theoretical electronic specific-heat coefficients $\gamma_{\text{band}}$ are 13.2 and 11.5 mJ K$^{-2}$ mol$^{-1}$ for PuRhGa$_5$ and PuIrGa$_5$, respectively, which are smaller than that of PuCoGa$_5$.

Since the 15th, 16th, 17th and 18th bands are partially occupied both for PuRhGa$_5$ and PuIrGa$_5$, as in the case of PuCoGa$_5$, these four bands construct the Fermi surface. This point can be intuitively understood, when we recall that the number of valence electrons in the transition metal ion is unchanged among three Pu-115 materials. The hole and electron sheets of the Fermi surface in PuRhGa$_5$ and PuIrGa$_5$, respectively, are shown in figures 4(c) and (d). The Fermi surface from the 15th band consists of one hole sheet centred at the $\Gamma$ point. The 16th band has a large cylindrical hole sheet which is centred at the $\Gamma$ point and two equivalent small hole pockets at the X point for PuRhGa$_5$, while for PuIrGa$_5$, this Fermi surface possesses crossed arms, which are connected like a jungle-gym to the next Brillouin zone. The 17th band has a large electron sheet which is open in the [0 0 1] direction and looks like a cylinder running along the V-axis. The Fermi surface from the 18th band consists of one hole sheet centred at the A point. Note that for PuCoGa$_5$, the 18th band has a slender cylindrical electron sheet centred at the M point. In this sense, two-dimensionality is considered to be better in PuCoGa$_5$.

The number of carriers contained in the Fermi-surface sheets for PuRhGa$_5$ (PuIrGa$_5$) are 0.040 (0.025), 0.627 (0.579) holes cell$^{-1}$, 0.594 (0.537) and 0.073 (0.067) electrons cell$^{-1}$ in the 15th, 16th, 17th and 18th bands, respectively. The total number of holes is equal to that of electrons, again indicating that PuRhGa$_5$ and PuIrGa$_5$ are also compensated metals.

Among three Pu-115 compounds, we have not found significant differences in the Fermi-surface structure, when Co is substituted by Rh or Ir, except for the two-dimensionality in the minor Fermi-surface sheet from the 18th band. This is naively explained by the fact that Co, Rh and Ir belonging to the same family in the periodic table include the same number of valence electrons. However, PuCoGa$_5$ and PuRhGa$_5$ are superconductors with $T_c = 18.5$ and 8.7 K, respectively, while PuIrGa$_5$ is considered to be non-superconducting at least up to now. Since the energy scale for superconductivity should be smaller than the width of quasi-particle band, it is very difficult to point out the difference in superconducting nature with such small energy scale only from the band-structure calculation with larger energy scale. To conclude the point concerning superconductivity, it is necessary to perform more careful investigations based on a microscopic $f$-electron model by taking account of the difference in the band-structure. We postpone such a problem to the future.
Table 5. The number of valence electrons for NpTGa$_5$ in the APW spheres for Np, Fe, Co, Ni and Ga partitioned into angular momenta

|   | s    | p   | d   | f    |   | s    | p   | d   | f    |   | s    | p   | d   | f    |
|---|------|-----|-----|------|---|------|-----|-----|------|---|------|-----|-----|------|
| Np| 0.32 | 6.11| 1.91| 4.17 |   | 0.38 | 6.12| 1.91| 4.15 |   | 0.32 | 6.041| 1.79| 4.13 |
| Co| 0.41 | 0.43| 7.49| 0.01 |   | 0.48 | 0.49| 8.39| 0.01 |   | 0.40 | 0.38 | 6.40| 0.01 |
| Ga(1c)| 0.98 | 0.73| 9.93| 0.01 |   | 0.94 | 0.67| 9.92| 0.01 |   | 1.08 | 0.90 | 9.98| 0.02 |
| Ga(4i)| 3.75 | 2.90| 39.75| 0.06 | 3.72 | 2.79| 39.73| 0.05 | 3.85 | 2.92 | 39.79| 0.06 |

3.3. NpTGa$_5$

Let us turn our attention to NpTGa$_5$ with T = Fe, Co and Ni. It is quite impressive that Np-115 compounds have been synthesized and several kinds of physical quantities have been intensively measured [18]–[25]. In particular, the dHvA signals have been successfully detected and the comparison with band-calculation results is important. Note that a part of the results on NpCoGa$_5$ has been reported [43, 45]. Again we emphasize that the present results are obtained by assuming the non-magnetic state for Np-115. We also assume that all 5$f$ electrons are itinerant in our calculations.

In figures 5(a)–(c), we show the energy-band structure along the symmetry axes in the Brillouin zone near the Fermi energy for NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$. The values of the Fermi level are found to be 0.43, 0.46 and 0.46 Ryd for NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$, respectively. At the first glance, there is no qualitative difference in the energy-band structure among three Np-115 compounds. However, due to the slight increase of the Fermi level in the order of NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$, the Fermi-surface structure is sensitively changed among Np-115 materials, as we will see later.

The total DOS is calculated at $E_F$ as $N(E_F) = 133.6, 230.4$ and 171.9 states Ryd cell$^{-1}$ for NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$, respectively, leading to $\gamma_{\text{band}} = 23.1, 39.9$ and 29.8 mJ K$^{-2}$ mol$^{-1}$. Note that $\gamma_{\text{exp}} = 60.0$ mJ K$^{-2}$ mol$^{-1}$ for NpCoGa$_5$ [20] and 100.0 mJ K$^{-2}$ mol$^{-1}$ for NpNiGa$_5$, $\lambda$ is equal to 0.5 and leading to $\lambda = 2.4$. The number of the valence electrons in the APW sphere is listed in table 5. For NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$, there are 8.03, 8.11 and 8.44 valence electrons outside the APW sphere in the primitive cell, respectively. The Np APW sphere contains about 4.13, 4.17 and 4.15 electrons in the $f$ state for T = Fe, Co and Ni, respectively.

Next we discuss the Fermi surfaces of three Np-115 compounds. For NpFeGa$_5$, we note that the lowest 14 bands are fully occupied, the next two bands are partially occupied, and the higher bands empty. Then, the 15th and 16th bands construct the Fermi surfaces, as shown in figure 5(d). These Fermi-surface sheets are small in the size and closed in topology. Note that there exists no open orbit on any sheets.

For NpCoGa$_5$, as shown in figure 5(e), the lowest 14 bands are fully occupied. The next three bands are partially occupied, while higher bands are empty, indicating that the 15th, 16th and 17th bands crossing the Fermi level form the hole or electron sheet of the Fermi surface. The Fermi surface from the 15th band consists of small hole sheets centred at the $\Gamma$ point. The 16th band constructs a large cylindrical hole sheet centred at the $\Gamma$ point, which exhibits a complex network consisting of big ‘arms’ along the edges of the Brillouin zone. The 17th band constructs a small electron sheet.
**Figure 5.** Energy-band structure near the Fermi level for (a) NpFeGa$_5$, (b) NpCoGa$_5$, and (c) NpNiGa$_5$. Fermi surfaces for (d) NpFeGa$_5$ with 15th band hole sheets and 16th band electron sheets (e) NpCoGa$_5$ with 15th band hole sheets, 16th band hole sheets, and 17th band electron sheets, and (f) NpNiGa$_5$ with 15th band hole sheets, 16th band hole sheets, 17th band electron sheets, and 18th band electron sheets.
For NpNoGa$_5$, we show the Fermi surfaces formed by 15th, 16th, 17th and 18th bands. As shown in figure 5(d), the Fermi surface from the 15th band includes one small hole sheet centred at the $\Gamma$ point. The 16th band constructs a large cylindrical hole sheet centred at the $\Gamma$ point, while two equivalent small hole sheets are centred at $X$ points. The 17th band has a large electron sheet. Each electron sheet lies across the $V$-axis and looks like a cushion. If the 16th band hole Fermi surface of NpCoGa$_5$ is almost occupied by an electron and the volume of the 17th band electron Fermi surface is slightly enlarged, these Fermi surfaces correspond to the 16th band hole Fermi surfaces and the 17th band electron ones of NpNiGa$_5$, respectively.

The numbers of carriers contained in these Fermi-surface sheets are summarized as follows: for NpFeGa$_5$, those are 0.094 holes cell$^{-1}$ and 0.094 electrons cell$^{-1}$ in the 15th and 16th bands, respectively. For NpCoGa$_5$, we find 0.014, 1.044 holes cell$^{-1}$ and 0.058 electrons cell$^{-1}$ in the 15th, 16th and 17th bands, respectively. Finally, for NpNiGa$_5$, we obtain 0.006, 0.430 holes cell$^{-1}$, 0.427 and 0.009 electrons cell$^{-1}$ in the 15th, 16th, 17th, and 18th bands, respectively. Since the total number of holes is equal to that of electrons, NpFeGa$_5$ and NpNiGa$_5$ are compensated metals. Note that there are just 0.094 holes cell$^{-1}$ and the compensating number of electrons, indicating that NpFeGa$_5$ is a semi-metal. On the other hand, for NpCoGa$_5$, we find that the total number of holes is not equal to that of electrons, which means that NpCoGa$_5$ is an uncompensated metal.

Here, we discuss similarity and difference among three Np-115 compounds. For the purpose, let us examine the trend in the change of $d$ electron number in transition metal atoms. The numbers of electrons with the $d$ character contained in the Fe, Co and Ni APW spheres are 6.40, 7.49 and 8.39 in NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$, respectively, indicating clearly that the number is increased by unity among three Np-115 compounds. Since one more $d$ electron is added to NpCoGa$_5$, the Fermi level for NpNiGa$_5$ is shifted upward in comparison with that of NpCoGa$_5$.

To understand the change in the electronic properties among three Np-115 compounds, it is useful to see the total and partial DOS, as shown in figure 6. We should note that there is no qualitative difference in the energy-band structure among Np-115 compounds: the 5$f$ bands are split into two subbands by the spin–orbit interaction and due to the hybridization between 5$f$ and 4$p$ electrons, finite DOS always appear at the Fermi level, indicating that Np-115 compounds are metallic in our band-structure calculations. Note, however, that the peak structure in the DOS are located just at or near the Fermi energy in common with three Np compounds. This fact may be related to the easy appearance of magnetism in Np-115 materials. When the transition metal atom is substituted, the position of $E_F$ is slightly shifted upward with increasing $d$ electron in the order of NpFeGa$_5$, NpCoGa$_5$ and NpNiGa$_5$.

The partial DOS for 3$d$ electrons are shown as shaded region in figure 6. We can observe that the center of the 3$d$ band approaches gradually to $E_F$ in the order of Ni, Co and Fe. In the case of NpFeGa$_5$, the main part of the 3$d$ electron state is near the Fermi level, indicating that the 3$d$ electron has contribution to electronic properties including magnetism. This result may be related to the experimental fact that magnetic moment appears at Fe site in NpFeGa$_5$, while there is no moment for Co and Ni sites [23, 26]. To conclude this point theoretically, it is necessary to analyse the model further including 3$d$ electrons in addition to 5$f$ and 4$p$ electrons. Such an analysis is a future problem.
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Figure 6. Density of states for NpFeGa₅, NpCoGa₅ and NpNiGa₅. Solid curves and shaded region show the total and the 3d DOS, respectively. The dashed line indicates the Fermi energy, from which the energy is measured.

Table 6. The number of valence electrons for AmCoGa₅ in the Am APW sphere, the Co APW sphere and the Ga APW sphere partitioned into angular momenta.

|  | s   | p   | d   | f  |
|---|-----|-----|-----|----|
| Am | 0.39| 6.16| 1.68| 6.39|
| Co | 0.43| 0.42| 7.45| 0.01|
| Ga(1c) | 0.95| 0.67| 9.92| 0.01|
| Ga(4i) | 3.73| 2.77| 39.75| 0.05|

3.4. AmCoGa₅

Now we show our calculated result of the energy-band structure of AmCoGa₅ within the framework of the itinerant-electron model for the 5f electrons. In figure 7(a), we depict the energy-band structure near the Fermi level, which is located at 0.437 Ryd. The number of the valence electrons in the APW sphere is partitioned into the angular momenta, as listed in table 6. There are 8.22 valence electrons outside the APW sphere in the primitive cell. The total DOS at $E_F$ is evaluated as $N(E_F) = 62.8$ states Ryd cell⁻¹. By using this value, the theoretical specific heat coefficient $\gamma_{\text{band}}$ is estimated as $10.9$ mJ K⁻² mol⁻¹.

In figure 7(a), the lowest 15 bands are fully occupied. The next four bands are partially occupied, while higher bands are empty. Namely, 16th, 17th, 18th and 19th bands crossing the Fermi level construct the hole or electron sheet of the Fermi surface, as shown in figure 7(b). The Fermi surface from the 16th band consists of two equivalent small hole sheets centred at the X points and one hole sheet centred at the Γ point. It consists of eight small closed sheets which have the mirror-inversion symmetry with respect of $\{100\}$, $\{010\}$ and $\{001\}$ planes lying between them. The 17th band has three kinds of sheets. One is a set of two equivalent electron-like pockets, each of which is centred at the R point. Another is an electron pocket centred at the Z point. There is also a large cylindrical electron sheet centred at the M point, which characterizes the two-dimensional Fermi surface. The 18th band has a slender cylindrical electron sheet which is also centred at the M point. The 19th band constructs a very small electron sheet, which is centred at the M point. The number of carriers contained in these Fermi-surface sheets are 0.0500 holes cell⁻¹, 0.7798, 0.2698, and 0.0004 electrons cell⁻¹ in the 16th, 17th, 18th and 19th bands, respectively. The total number of holes is not equal to that of electrons, indicating that AmCoGa₅ is an uncompensated metal.
Figure 7. (a) Energy-band structure near the Fermi level for AmCoGa\textsubscript{5}. (b) Calculated Fermi surfaces of AmCoGa\textsubscript{5} for 16th band hole sheets, 17th band electron sheets, 18th band electron sheets and 19th band electron sheets.

We remark that the Fermi surfaces of AmCoGa\textsubscript{5} seem to be similar to those of PuCoGa\textsubscript{5}, since we can see the multiple two-dimensional cylindrical Fermi-surface sheets with a constricted part around the M point. For a better understanding of the Fermi surface, we attempt to explain the relationship between AmCoGa\textsubscript{5} and PuCoGa\textsubscript{5}. On the basis of the itinerant 5\textit{f} electron model, AmCoGa\textsubscript{5} has one more Bloch electron per primitive cell in comparison with PuCoGa\textsubscript{5}. Therefore, in AmCoGa\textsubscript{5}, the Fermi-level shift upward, indicating that relatively the position of 5\textit{f} bands move downward. As a result, we find that the 15th band hole sphere of PuCoGa\textsubscript{5} disappears and the size of the 16th band hole sheet becomes smaller. On the other hand, the sizes of the 17th and 18th band cylindrical electron sheets become large. Note that in AmCoGa\textsubscript{5}, a small electron sheet appears in the 19th band.

The same similarity in the Fermi-surface structure can be found between Ce- and Th-115 materials [49]. If we assume the trivalent rare-earth or actinide ion in 115 structure, we find zero and one 5\textit{f} electron for Th and Ce ions, respectively. It has been found that the Fermi surfaces of ThRhIn\textsubscript{5} constructed from the four bands contain the 5\textit{f} components due to the large hybridization between \textit{f} and \textit{p} electrons near the Fermi level. In fact, in our band-calculation results, we have found 0.55 5\textit{f} electrons in the Th atom. Then, the Fermi-surface structure of Th-115 becomes similar to that of Ce-115. If we are based on the electron–hole symmetry in the \textit{j}–\textit{j} coupling scheme and the assumption that all 5\textit{f} electrons are itinerant, the similarity between Ce-115 and Th-115 is considered to have the same origin as that between Pu-115 and Am-115.

However, Opahle et al [45] reported that the Fermi-surface structure of AmCoGa\textsubscript{5} is different from that of PuCoGa\textsubscript{5}. Although the figures for band structure and Fermi-surface of AmCoGa\textsubscript{5} were not shown in [45], their results for PuCoGa\textsubscript{5} were found to be quite similar to ours and thus, their Fermi surfaces of AmCoGa\textsubscript{5} are deduced to be different from ours. One reason for this discrepancy is the treatment of 5\textit{f} electrons. Namely, we have treated itinerant 5\textit{f} electrons, while Opahle et al considered localized ones. In general, the tendency of localization of 5\textit{f} electron in Am is stronger than that in Pu, but in actual compounds, it is difficult to determine
Figure 8. (a) Energy-band structure near the Fermi level for UCoGa$_5$. (b) Calculated Fermi surfaces of UCoGa$_5$ for 15th band hole sheets and 16th band electron sheets.

which is the better approximation, the localized or itinerant picture. It will be necessary to clarify which picture better explains the experimental results consistently.

4. Discussion

In the previous section, we have shown the band-structure calculation results for PuTGa$_5$, NpTGa$_5$ and AmCoGa$_5$. In order to obtain deep insight into the electronic properties of these 115 compounds, it is useful to compare the results with the electronic energy-band structure of UCoGa$_5$, as shown in figure 8(a) [50, 51]. Note that three U 6$p$ and 25 Ga 3$d$ bands are omitted.

The Fermi energy $E_F$ is located at $E_F = 0.461$ Ryd. The total DOS at $E_F$ of UCoGa$_5$ is calculated as $N(E_F) = 48.4$ states Ryd cell$^{-1}$, which corresponds to $\gamma_{\text{band}} = 8.4$ mJ K$^{-2}$ mol$^{-1}$.

In UCoGa$_5$, the 15th and 16th bands form the Fermi surfaces, as shown in figure 8(b). The Fermi surfaces from the 15th band have one sheet centred at the $\Gamma$ point, two equivalent sheets centred at the X points, and the sheets across the S-axis. We show a set of the 16 electron sheets of the Fermi surfaces in the 16th band. Each electron sheet across the T-axis looks like a cushion. The total number of holes is equal to that of electrons, which means that UCoGa$_5$ is a compensated metal.

Except for details, we can observe that the sheets of the Fermi surface of UCoGa$_5$ with small size and closed topology are similar to those of NpFeGa$_5$ (see figure 5). We also remark that NpCoGa$_5$ is considered to be UCoGa$_5$ plus one more $f$-electron from the viewpoint of the Fermi surface topology, since the small-pocket parts are the remnants of UCoGa$_5$ and the large-volume Fermi surface contains one additional electron. Interestingly enough, the Fermi surfaces of UNiGa$_5$ [5] are quite similar to figure 5, since both of UNiGa$_5$ and NpCoGa$_5$ are regarded as UCoGa$_5$ plus one more electron, if we simply ignore the difference in the original character, $d$ or $f$, of the additional one electron. In this picture, NpCoGa$_5$ is UCoGa$_5$ plus one $f$-electron and NpFeGa$_5$ is regarded as NpCoGa$_5$ minus one $d$-electron. Thus, the bands forming the Fermi
surfaces of UCoGa$_5$ become the same as those of NpFeGa$_5$. In fact, the Fermi-surface sheets of both UCoGa$_5$ and NpFeGa$_5$ consist of several kinds of pockets with small volumes, although the actual shape of Fermi surfaces of UCoGa$_5$ is not very similar to that of NpFeGa$_5$.

Based on the discussion on the number of $d$ and $f$ electrons, we also point out that the Fermi surfaces of NpNiGa$_5$ should be similar to those of PuCoGa$_5$, as shown in figures 3 and 5. However, it is clearly observed that two-dimensionality in the Fermi surfaces of NpNiGa$_5$ becomes worse compared with those of PuCoGa$_5$. This may be related to the reason why NpNiGa$_5$ is not superconducting, despite being in the same group as PuCoGa$_5$ in the electron number discussion.

The above phenomenology on the $d$- and $f$-electron numbers seems to suggest that a rigid-band picture works for actinide 115 compounds. This point can be partly validated due to the fact that the band structure of these compounds is basically determined by hybridization between broad $p$ bands and narrow $f$ bands in the vicinity of the Fermi level. Based on the rigid-band picture, the electron band structure itself is not changed significantly, even when we change actinide and/or transition metal ions. Thus, just by counting the valence electrons of actinide and transition metal ions, we can easily deduce the Fermi-surface topology, starting with the results of UCoGa$_5$.

It is interesting to compare the total and partial DOS of AnCoGa$_5$ (An = U, Np, Pu and Am), as shown in figure 9. We again see that the structures of DOS are qualitatively in common with actinide 115 compounds except for details, when we change the actinide ions. In UCoGa$_5$, the Fermi energy is located in a valley, leading to the small DOS at $E_F$, consistent with a semi-metal behaviour. On the other hand, for NpCoGa$_5$, as mentioned in the previous section, the peak is located just at the Fermi energy. After including the effect of electron correlation, magnetic transition may easily occur in this case. For PuCoGa$_5$ and AmCoGa$_5$, we observe moderate values of the DOS at the Fermi energy, consistent with the paramagnetic metallic phase. We emphasize that the behaviour of the partial DOS for $f$ electrons follows that of the total DOS, suggesting that $f$ electrons mainly contribute to the electronic properties in actinide 115 compounds.
It is noted that the numbers of electrons with the $f$ symmetry contained in the actinide APW sphere are, respectively, 3.2, 4.2, 5.2 and 6.4 for UCoGa$_5$, NpCoGa$_5$, PuCoGa$_5$ and AmCoGa$_5$, increase by unity as the atomic number of actinide atom increases. This fact seems to suggest that electrons are supplied to the unoccupied $5f$ bands of UCoGa$_5$. Namely, the change in the Fermi-surface structures among actinide 115 compounds is basically understood by an upward shift of the Fermi level on the rigid electronic bands. The same change occurs between PuCoGa$_5$ and AmCoGa$_5$. Note, however, that it is also important to specify the difference among 115 compounds. In particular, the change of the magnetic structure in Np-115 compounds [23]–[26] is not clarified by the present band-structure calculations. Detailed analysis on the microscopic $f$-electron model is required [31].

Finally, let us briefly discuss the dHvA experimental results on NpNiGa$_5$ [19], NpCoGa$_5$ [21] and NpRhGa$_5$ [22], in comparison with our theoretical Fermi surfaces of Np-115. For NpNiGa$_5$, Aoki et al have observed several sets of the dHvA frequency branches in the region of the order $10^7$ Oe, which have the cyclotron effective masses from 1.8$m_0$ to 4.9$m_0$ in the magnetic-field direction [1 0 0] and [0 0 1]. Here $m_0$ is the rest mass of a free electron.

For NpCoGa$_5$, a couple of cylindrical sheets of Fermi surfaces with large volume have been detected [21], while in our band-structure calculations, one cylindrical sheet is obtained, as shown in figure 5. Namely, the dHvA results are not explained by the band calculation in the paramagnetic state. A possible explanation for this discrepancy is to consider the folding of Fermi surfaces in the magnetic Brillouin zone, since NpCoGa$_5$ exhibits the A-type AF structure with a Néel temperature $T_N = 47$ K. If the magnetic unit cell is elongated along the [0 0 1] direction and doubled with respect to the chemical unit cell as observed in UPtGa$_5$ [11], a quasi-two dimensional Fermi surface is more likely to appear due to the flat magnetic Brillouin zone.

However, in actual dHvA experiments, due to the applied magnetic field, the system is not AF, but ferromagnetic. Thus, the above explanation cannot be simply applied to this case. In addition, quite recently, the dHvA experiments have been also performed for NpRhGa$_5$, which is also A-type AF. In this material, four cylindrical sheets of Fermi surfaces have been reported [22], consistent with the folding of Fermi surfaces of NpCoGa$_5$ in the magnetic Brillouin zone. Thus, it is experimentally confirmed that a couple of cylindrical sheets of Fermi surfaces exist in NpCoGa$_5$.

It may be true that the rigid-band picture cannot explain the band-structure of all actinide 115 compounds. However, a simple tight-binding approximation based on the $j$-$j$ coupling scheme can reproduce a couple of cylindrical Fermi surfaces [31], by assuming that two of four $5f$ electrons in Np ion are active. In order to understand the dHvA experimental results on the Fermi surfaces, it is a correct direction to improve the band-structure calculations by considering the difference in the degree of itinerancy among $5f$ electrons. In fact, it was suggested that a couple of cylindrical Fermi-surface sheets are obtained in the spin-orbital polarized band-structure calculation. Another way is an application of the full-potential method to the fully relativistic band-structure calculations. Such an extension of the band-calculation method is an important future task.

5. Summary

In this paper, we have applied the RLAPW method to the self-consistent calculation of the electronic structure for PuTGa$_5$, NpTGa$_5$ and AmCoGa$_5$ on the basis of the itinerant $5f$ electron
picture, by assuming the non-magnetic phase. It has been found that a hybridization between the 5f and Ga 4p states occurs in the vicinity of $E_F$. We have calculated the dHvA frequencies for PuCoGa_5 for future experiments. The similarity in the Fermi-surface structure among actinide 115 compounds has been found to be understood based on the rigid-band picture, while the theoretical Fermi surfaces of NpCoGa_5 are different from ones in the recent dHvA experimental results. In order to understand this point, a challenging future problem is to improve the band-calculation technique.

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