Correlation effect on effective model with realistic electronic structure for heavy fermion compound YbAl$_3$

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Abstract. Based on the recently proposed effective Hamiltonian for typical heavy fermion compound YbAl$_3$, the correlation effect is systematically investigated in the physical properties. The band part of the Hamiltonian consists of conduction bands described by nearly free electron method, hybridization between the conduction and 4f-electrons, and the localized 4f-states of Yb ions on the lattice site. The correlation effect is considered by using the self-consistent perturbation theory with local approximation. The temperature dependence of the specific heat coefficient is calculated, and the anomalous behaviors are found in the low temperature region in accord with the experiments. We show that the anomalies may originate from the correlation effect and the structure of the non-interacting density of states.

1. Introduction
In the analyses in heavy fermions, the periodic Anderson model (PAM) and its extensions are often employed, which can describe the physical properties qualitatively. However, the momentum dependence of the hybridization between conduction and localized f-orbitals is often neglected. Use of such a model yields an appearance of a hybridization gap above or below the Fermi level, and this simplification may lead to the serious inconsistency between the experimental and theoretical results. Therefore, we proposed an effective Hamiltonian for describing the band structure of heavy fermion compounds with only a few adjustable parameters, and apply it to the typical heavy fermion compound YbAl$_3$ [1]. The obtained band structure is almost consistent with the first principle calculation [2], and the overall structure of the optical conductivity corresponds to the experimental result [3]. However, the amplitude of the specific heat coefficient is only a third of that of the experimental result [4], which may be due to the absence of the correlation effect.

In this paper, we focus on the heavy fermion metal YbAl$_3$ and investigate the correlation effect. Applying the self-consistent second order perturbation theory with local approximation to the constructed effective Hamiltonian, we calculate physical quantities. In particular, we concentrate the temperature dependence of the specific heat coefficient.

2. Model of YbAl$_3$ and Method
YbAl$_3$ shows that the mean valence is in the range from 2.65 to 2.8, which indicates this material is a valence fluctuation compound. The Sommerfeld coefficient $\gamma$ is about 40 mJ·K$^{-2}$·mol$^{-1}$.
With a decrease of temperature, the specific heat divided by temperature $c_v/T$ shows prominent temperature dependence; the peak structure appears at $T \sim 80$ K and the amplitude increases at the very low temperature region ($T < 25$ K). The latter was interpreted as the effect of the lattice coherence [5].

We introduce the effective Hamiltonian for YbAl$_3$, which can be written as follows $H^{\text{eff}} = H^{\text{band}} + H^{\text{int}}$ where $H^{\text{band}}$ ($H^{\text{int}}$) represents the band (interaction) part. $H^{\text{band}}$ consists of the conduction band, the localized 4f orbitals, and the hybridization between the conduction and the localized 4f-electrons terms. $H^{\text{int}}$ consists of the Coulomb interaction between the localized 4f-electrons of Yb ions.

Let us discuss the band part of the Hamiltonian for YbAl$_3$. Each electronic configuration is [Ne](3s)$^2$(3p)$^1$ for Al and [Xe](4f)$^{14}$(5d)$^0$(6s)$^2$ for Yb atom. There are 25 electrons per unit cell of YbAl$_3$ except the closed shells. The mean configuration of the localized 4f-electrons lies in between $f^{14}$ to $f^{13}$, which indicates that YbAl$_3$ shows the valence fluctuation and becomes the metallic system. Most of the conduction electrons consist of s-electrons, which spread over in the crystal system on the weak periodic potentials. Therefore, we employ the nearly free electron (NFE) method with the empty-core pseudo-potentials screened by RPA in order to describe the conduction bands. The atomic potential and the core radius in each atom are employed from Harrison’s textbook [6]. The degenerated localized 4f-electron levels split into the total angular momentum $J=7/2$ and $J=5/2$ levels because of the strong spin-orbit coupling. The difference between both energy levels is of the order of 0.1 eV. $J=7/2$ level $E_7^{7/2}$ is located near and below the Fermi level, while $J=5/2$ level is far from the Fermi level [4]. Although $J=5/2$ level may not affect the physical properties in the low temperature region, we also include $J=5/2$ term in the Hamiltonian and set the energy difference between both levels as 0.1 eV. Thus $E_7^{7/2}$ is a tuning parameter in the present method. The plane wave and the localized 4f-electron can be expanded by the spherical harmonics, so that the matrix element of the hybridization term is described by this spherical harmonics. We neglect the wave vector $k$-dependence of the radial part and regard this part as a constant which denotes the hybridization amplitude $V$, for simplicity. Note here that although we neglect the $k$-dependence of the radial part, we keep the angular dependence of $k$ completely.

Thus we stress that the effective Hamiltonian for the band energy of YbAl$_3$ contains only two adjustable parameters, $J = 7/2$ level $E_7^{7/2}$ and the hybridization strength $V$. Diagonalizing this effective Hamiltonian $H^{\text{band}}$ for the band energy, we can obtain the band structure. For $E_7^{7/2} = 0.6$ Ry and $V = 0.03$ Ry, the overall band structure around and below the Fermi level and the topology of the Fermi surface correspond to the LDA results [2] except that around M point in the Brillouin zone [1].

Next, let us consider the interaction term of the Hamiltonian $H^{\text{int}}$. The Coulomb interaction between the localized 4f-electrons may play important role in some physical phenomena, the energy level $E_7^{7/2}$ is located near the Fermi level. The bases which diagonalize the band term of the Hamiltonian makes the matrix element of the Coulomb interaction very complicated representation, so that we approximate this element independent of the band index for simplicity. The interaction term is given by $H^{\text{int}} = U \sum_{i, \alpha, \beta, \gamma, \delta} a_{i\alpha}^\dagger a_{i\beta} a_{i\gamma}^\dagger a_{i\delta}$ where $a_{i\alpha}^\dagger$ ($a_{i\alpha}$) is a creation (annihilation) operator of a diagonalized band with site $i$ and band index $\alpha$, and $U$ represents the strength of the Coulomb interaction. Since the width of the diagonalized bands is greater than 1 Ry, we extract the low energy bands within $|E_{k\alpha} - E_F| < 0.1$ Ry in order to discuss precisely the correlation effect, in which $E_{k\alpha}$ is the energy eigenvalue of $H^{\text{band}}$ with the wave-vector $k$ and band index $\alpha$, and $E_F$ denotes the Fermi level at absolute zero temperature. Note that there are 14 bands within this energy range, which are used for the effective Hamiltonian $H^{\text{eff}}$ in the present study.

In order to investigate the correlation effect, we employ to the self-consistent second order
perturbation theory combined with local approximation. It is because the 4f electrons have the strong localized character and the amplitude of the Coulomb interaction is generally renormalized to a smaller value at low energy region in heavy fermion metallic compounds. Note here that the band off-diagonal terms in the self energy vanish within the present scheme.

3. Results

Let us discuss the physical properties of YbAl$_3$. The main adjustable parameters are chosen as $E_{\text{f}}^{7/2} = 0.6$ Ry and $V = 0.03$ Ry, respectively. The other employed parameters in the band term of the Hamiltonian are the same as in ref. [1]. Figure 1 shows the total density of states (DOS) near the Fermi level. The split peaks originate from the contribution of the 4f $J = 7/2$ levels. Although the 4f electrons remain the strong localized character where almost all the 4f orbitals are occupied, the hybridization between the conduction and 4f electrons gives rise to the broadening of the DOS at the low energy region. The particle number of the 4f electrons per site is about 13.43 for $U = 0$ [1]. We also stress that a hybridization gap never appears in the DOS since the $k$-dependence of the hybridization is taken into account. With an increase of the Coulomb interaction $U$, the DOS is smeared and the peak width at the very low energy region becomes narrower, which results from dynamical effect of the self-energy. It is well-known that this effect gives rise to the mass enhancement.

Next, let us consider the specific heat divided by temperature $c_v/T$ (Fig. 2), in which the specific heat $c_v$ is numerically estimated by the temperature derivative of the internal energy obtained within the present scheme. For the non-interacting case, the maximum appears at $T \sim 0.002$ Ry ($\sim 300$ K). With a further decrease of temperature, $c_v/T$ decreases monotonically and becomes constant at the very low temperature region ($T < 0.0007$ Ry). At low temperature region, the specific heat are given by $c_v = \gamma T + \alpha T^3$, in which the coefficient $\gamma$ (specific heat coefficient) indicates the degree of the mass enhancement, and the coefficient $\alpha$ is obtained from the derivative of the DOS at the Fermi level [7, 8]. With an increase of temperature from absolute zero temperature, $c_v/T$ increases (decreases) for $\alpha > 0$ ($\alpha < 0$). When the structure of the DOS is a Lorentzian-like form where the peak position is located slightly below the Fermi level, the coefficient $\alpha$ favors the positive value [8]. Thus, although the position is twice higher than the experiment [5], we conclude that the origin of the peak $c_v/T$ at $T \sim 0.002$ Ry is due to the band structure (or the structure of DOS).
With an increase of the interaction $U$, the enhancement of $c_v/T$ occurs at the very low temperature region ($T < 0.0005$ Ry). While the electron correlation plays very important role for the enhancement of $c_v/T$, the effect vanishes except the very low temperature region because the effective Coulomb interaction is remarkably reduced due to the strong renormalization effects in the metallic systems of heavy fermions. However, since the specific heat is numerically obtained from the energy derivative, it is very difficult to estimate the accurate values of $c_v/T$ at very low temperature. It is well-known that the temperature dependence of the energy is also given by $E(T) = a_F T^2 + b_F T^4$ within the Fermi liquid theory. Thus, we obtain these constants by using the energy fitting with least squares method within the very low temperature region ($T < 0.0003$ Ry), so that we can evaluate the specific heat as $c_v(T) = 2a_F T + 4b_F T^3$. The inset of figure 2 shows the specific heat divided by temperature combined with the fitting data. The fitting data are saturated near absolute zero temperature. Thus we can obtain the specific heat coefficient $\gamma (\sim 40 - 50 \text{ mJ} \cdot \text{K}^{-2} \cdot \text{mol}^{-1})$ for $U = 0.005$ Ry, which roughly corresponds to the experimental result.

Although the peak structure at $T \sim 0.002$ Ry for the non-interacting case is shifted to the lower temperature region with an increase of the interaction, this peak never vanishes. We stress that the anomalies of the structure of $c_v/T$ has two kinds of origins. One is the correlation effect at the very low temperature region ($T \ltimes 0.0003$ Ry) and the other is the band structure at the low temperature region ($T \sim 0.002$ Ry). Thus the overall structure of $c_v/T$ reproduces that of the experimental result.

4. Summary

We have constructed an effective Hamiltonian of typical heavy fermion compound YbAl$_3$ and investigated the correlation effect by using the self-consistent second order perturbation theory with local approximation. The temperature dependence of the specific heat divided by temperature is calculated. The structure of $c_v/T$ may be strongly affected by the correlation effect at the very low temperature region and the structure of the DOS at the low temperature region, which is consistent with the experimental results.

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