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Electronic Structures of Strongly Correlated Itinerant-Electron System

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Fermi surfaces and mass enhancement factor in strongly correlated itinerant-electron compounds, $CeCo_2$, $CeRh_2$, $CeRu_2$, YCo_2 and $LuCo_2$, are calculated. The calculated Fermi surfaces of $CeCo_2$ and $CeRu_2$ are in good agreement with the dHvA measurements. Mass enhancement factors, $\tilde{\gamma}$, are calculated by using a method of perturbation expansion with respect to electron-electron interactions based on Fermi liquid theory. The values of electronic specific-heat coefficient calculated by using $\tilde{\gamma}$ reproduce well the experimental values for $CeCo_2$, $CeRu_2$, YCo_2 and $LuCo_2$.

Key word: mass enhancement factor, Fermi surface, heavy fermion compounds, cubic Laves phase

1 Introduction

Density functional theory (DFT) within the local density approximation (LDA) have provided good descriptions for electronic states in many compounds. Calculated Fermi surfaces in strongly correlated electron (SCE) system, *d*-electron and / or *f*-electron compounds, are in good agreement with experimental ones [1]. It is naturally accepted because DFT describes correctly physical quantities including many-body effects in ground states such as Fermi surfaces. On the other hand, the values of physical quantities in low-energy excited states, such as an electronic specific-heat coefficient (γ) and cyclotron effective masses, are hardly reproduced [1]. The reason is that DFT do not guarantee the many-body effects in low-energy excitation. In SCE system, most of such effects are caused by electron-electron interactions between SCEs. It is important how to approach many-body effects in understanding the physical property in such system.

One of the method for considering the many-body effects has been given by Yamada *et al.* [2] In the method, a calculation of enhancement for γ is equivalent to obtain an enhancement factor for SCE, $\tilde{\gamma}_k$. A simple method to calculate $\tilde{\gamma}_k$ is a second-order perturbation. However it is still difficult to calculate $\tilde{\gamma}_k$ numerically in a three-dimensional system, because it is necessary to carry out multiple integrals with respect to momentum. An enhancement factor can be calculated numerically only from a density of states (DOS) for SCEs if $\tilde{\gamma}_k$ is replaced by $\tilde{\gamma}(=\sum\tilde{\gamma}_k)$, in which $\boldsymbol{k}$-dependence is neglected.

Series of the cubic Laves-phase compounds have shown various physical properties such as mass enhancement, superconductivity and metamagnetism. Band structure calcualtions have been reported for $CeCo_2$, $CeRh_2$ and $CeRu_2$ [3]. Their Ce-*f* electrons are shown itinerant properties because of hybridization between Ce-*f* and conduction electrons, though be localized at the rare earth atom in general. The fermi surfaces of $CeCo_2$ and $CeRu_2$ are in good agreement with the dHvA experiments.

YCo_2 and $LuCo_2$ are well-known as a strongly enhanced Pauli paramagnets like Pd metal, in which Co-*d* electrons are strongly correlated ones. Band structure calcualtions have been reported for YCo_2 and $LuCo_2$ [4]. Recently, different characteristics of magnetic properties of $CeCo_2$ and a group of YCo_2 and $LuCo_2$ have been experimentally revealed. ^{59}Co NMR measurement for YCo_2, [5, 6] for $LuCo_2$ [7] and NMR and NQR measurements for $CeCo_2$ [8, 9] indicate: (1) the measured value of χ associated with Co-3*d* for $CeCo_2$ is much smaller than that for YCo_2 and $LuCo_2$; (2) $(T_1T)^{-1}$ of $CeCo_2$ is proportional to K_{3d}^2, where K_{3d} is a Knight shift with Co-3*d* electrons, but proportional to K_{3d} in YCo_2 and $LuCo_2$. While the experimental values of γ ($\gamma_{\rm exp}$) are very similar in YCo_2, $LuCo_2$ and $CeCo_2$, the calculated values of γ ($\gamma_{\rm band}$) are much smaller than those. Then large enhancement in effective masses at the Fermi energy are expected.

This paper is reported the Fermi surfaces and the enhanced values of γ ($\gamma_{\rm ehs}$) for cubic Laves-phase compounds $CeCo_2$, $CeRh_2$, $CeRu_2$, YCo_2, $LuCo_2$.

2 Method of Band Calculation

We have carried out band structure calculations by using a full-potential linear augmented plane wave (FLAPW) method. The crystal structure of compounds is a $MgCu_2$-type which is called a cubic Laves-phase. The space group is $Fd\bar{3}m$. The exchange-correlation interactions of electrons are taken into account by the LDA, in the form proposed by Moruzzi *et al.* [10] We adopted the relaxed-core approximation to the Xe-core states except $5p$ electrons on Ce atom, the Ar-core on Co atoms and the Kr-core on Rh and Ru atoms. The scalar relativistic effects are taken into account for all electrons and the spin-orbit interactions are included for valence electrons as a second variational procedure. The final potential is constructed self-consistently from eigenstates at 19 sampling $\boldsymbol{k}$-points in the irreducible 1/48 Brillouin zone. To obtain a final band structure, we selected 231 sampling $\boldsymbol{k}$-points in the irreducible 1/48 Brillouin zone.

3 Fermi Surfaces

We have calculated the Fermi surfaces with the extremal cross-sectional areas and the cyclotron effective masses for the compounds. The results are shown in our previous papers in detail [3]. The calculated Fermi surfaces for $CeCo_2$ and $CeRu_2$ can explain many observed branches and show similar topology. In $CeRh_2$, the uncertainty of assignment remains in several branches. It seems that the LDA calculation for $CeRh_2$ fails to obtain Fermi surfaces measured by the dHvA experiment. The calculated cyclotron masses in heavy fermion compounds are, as usual, smaller than the observed ones, because of mass enhancement.

4 Density of States and Enhancement Factor

A theory for mass enhancement in strongly correlated system is given by Yamada *et al* [2] in the framework of Fermi liquid theory on the basis of a periodic Anderson model. The theory is a reasonable one to describe the essential properties of the system and can be applied for real compounds if the non-correlation term in Hamiltonian is replaced with that in DFT.

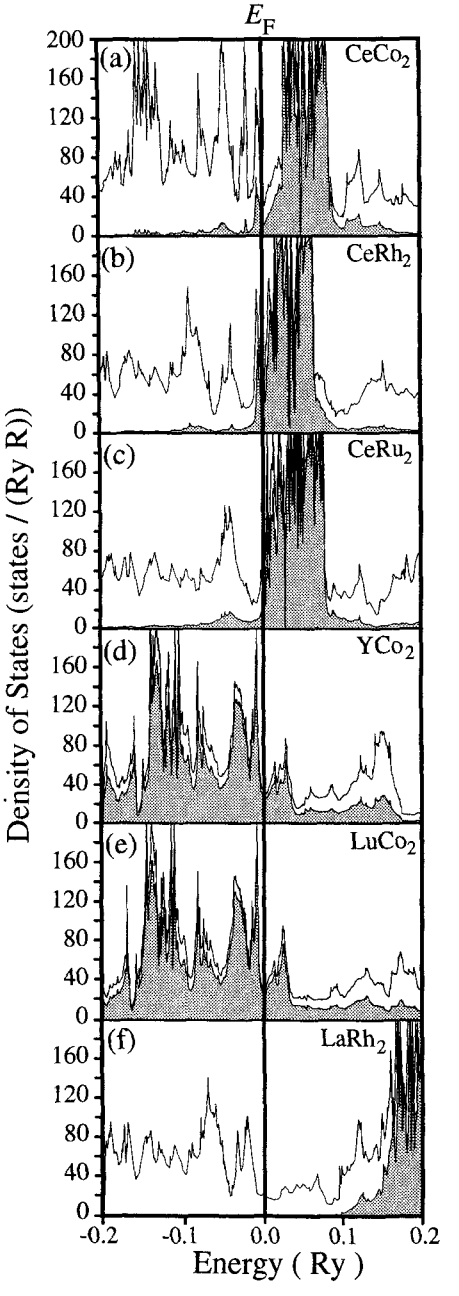

Figure 1: Calculated density of states (DOS) for (a) $CeCo_2$, (b) $CeRh_2$, (c) $CeRu_2$, (d) YCo_2, (e) $LuCo_2$ and (f) $LaRh_2$. The solid lines and shadow areas show the total and Ce-f (for f electron-case) or Co-d partial DOS (for d electron-case), respectively.

Table 1: Results of DOSs and enhancement calculations with experimental data. $\gamma_{\rm band}$ is the electronic specific-heat coefficient, i.e., the total DOS ($\rho(E_{\rm F}^{\rm o})$) obtained from a band calculation. R is the contribution of the component of SCE on Ce- or Co-site to the $\gamma_{\rm band}$. $\tilde{\gamma}$ is the enhancement factor according to partial DOS of SCE. (see text). $\gamma_{\rm ehs}$ is the value of γ taking into account the effects of$\tilde{\gamma}$ ($\gamma_{\rm ehs} = \gamma_{\rm band}\{1 + R_d(\tilde{\gamma} - 1)\}$). $\gamma_{\rm exp}$ is the experimental value.

	$\gamma_{\rm band}$ (mJ/K^2·mol)	R (%)	$\tilde{\gamma}^{\dagger}$	$\gamma_{\rm ehs}$ (mJ/K^2·mol)	$\gamma_{\rm exp}$ (mJ/K^2·mol)
			f electron case		
$CeCo_2$	14.6	33	4.76	33.3	35 [11]
$CeRh_2$	11.9	60	7.60	59.4	20 [12]
$CeRu_2$	11.6	31	5.77	28.5	30 [13]
$CeSn_3$	11.7	61	7.90	60.7	53 [1]
$LaRh_2$	3.7	3	1.08	3.7	3.7 [14]
			d electron case		
YCo_2	6.1	84	6.87	36.2	36.2 [15]
$LuCo_2$	5.7	82	6.58	31.9	26.6 [16]
$CeCo_2$	14.3	43	1.38	15.5 (33.3††)	35.0 [11]

†: calculated in the case of U_{ff}=5 eV or U_{dd}=1.8 eV
††: a value when U_{ff} between Ce-f electrons is considered

Following the theory, a coefficient of the T-linear term of the specific heat is

$$\gamma = \frac{\pi^2}{3} k_{\rm B}^2 \sum_k \left[\tilde{\gamma}_k(\mu) \rho_k^l(\mu) + \sum_\sigma \rho_{k\sigma}^c(\mu) \right], \quad (1)$$

where

$$\tilde{\gamma}_k(\omega) = 1 - \frac{\partial \Sigma_k(\omega)}{\partial \omega}, \quad (2)$$

$\Sigma_k(\omega)$, the self energy for the localized electrons. It is calculated by using the method of second-order perturbation with respect to U. Unfortunately, it is difficult to calculate $\partial\Sigma_k(\omega)/\partial\omega$ numerically for real compounds even in the second-order perturbation. The reason is that it is necessary to carry out multiple (six-fold in a three-dimensional system) integrals with respect to momentum $\boldsymbol{k}'$ and $\boldsymbol{q}$.

On the other hand, a factor $\tilde{\gamma}(= \Sigma\tilde{\gamma}_k)$ can be calculated easily compared to $\tilde{\gamma}_k$, because multiple integrals are replaced by three one-dimensional integrals over an energy. We can use $\tilde{\gamma}$, when summations of $\tilde{\gamma}_k$ and $\rho_k^f(\omega)$ with respect to $\boldsymbol{k}$ in eq.(1) are separated. Values of γ evaluated by $\tilde{\gamma}$ are the same as those using $\tilde{\gamma}_k$, when $\boldsymbol{k}$-dependence of self-energy is neglected. Details for formulation of $\tilde{\gamma}$ are shown in our previous paper [3]. The DOSs are shown in Figs. 1(a)- 1(f), respectively.

Using the partial DOSs and eq.(A.1) in ref. 3 , the enhancement factors $\tilde{\gamma}$ are calculated. The value of U for f-electron is used 5eV, because the value is evaluated U~4-5eV from XPS and BIS experiments [17, 18]

The results of the calculations are listed in Table 1. The main results are as follows:
(1) the values of $\gamma_{\rm ehs}$ for Ce-compounds are larger than $\gamma_{\rm band}$, while the values of $\gamma_{\rm band}$ and $\gamma_{\rm ehs}$ for $LaRh_2$ is equal to $\gamma_{\rm exp}$,
(2) the values of $\gamma_{\rm ehs}$ are fairly in good agreement with $\gamma_{\rm exp}$ except for $CeRh_2$.

5 Summary

We have calculated band structures for $CeCo_2$, $CeRh_2$, $CeRu_2$ YCo_2 and $LuCo_2$ using the FLAPW method with spin-orbit interactions. Using the electronic structures, we have calculated DOS and Fermi surfaces for all the compounds. The calculated Fermi surfaces of $CeCo_2$ and $CeRu_2$ explain well the dHvA measurements. Using the calculated partial DOS for SCE and the method of a second-order perturbation with respect to many-body effects, we have calculated the values of $\tilde{\gamma}$ and the enhanced electronic specific-heat coefficients $\gamma_{\rm ehs}$ for the above compounds. The results indicate that the values of $\gamma_{\rm ehs}$ for $CeCo_2$, $CeRu_2$, YCo_2 and $LuCo_2$ reproduce the values of $\gamma_{\rm exp}$. On the other hand, the calculated Fermi surfaces and the value of $\gamma_{\rm ehs}$ for $CeRh_2$ show poor agreement with the experiments. Consequently, when we calculate the values of $\gamma_{\rm ehs}$, it is important to evaluate Fermi surfaces corresponding to the experiments.

Acknowledgements

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