Derivation of RKKY Interaction between Multipole Moments in CeB₆ by the Effective Wannier Model based on the Bandstructure Calculation

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We have investigated the electronic states of CeB₆ and have directly calculated the RKKY interaction on the basis of the 74-orbital effective Wannier model which includes 14 Ce-f orbitals and 60 conduction (c) orbitals of Ce-d, s and B-p, s derived from the density-functional theory bandstructure calculation. By using not only the c-band dispersion but also the f-c mixing matrix elements of the Wannier model, the realistic couplings for all 15 active multipole moments in Γ₄ quartet subspace are obtained in the wavevector q-space and real-space. Both of the Γ₄ multipole quadrupoles (Oₓz, Oₓz, Oₓz) and the Γ₄ octupole Tₒₙ couplings are maximally enhanced with q = (π, π, π) which naturally explains the phase II of the antiferro-quadrupolar ordering at Tₒ = 3.2 K, and are also enhanced with q = (0, 0, 0) corresponding to the elastic softening of C₄₄. Also the couplings of the Γ₄ multipole quadrupoles Tₒₚ and Tₒₜ are quite large for q = (π, 0, 0), (0, π, 0) and (0, 0, π), which yields the antiferro-octupolar ordering of a possible candidate for phase IV of CeLa₁₋₁B₆. The intersite vector dependence of the RKKY couplings exhibit different long-range, oscillating, isotropic and anisotropic behaviors depending on the types of the multipole moments. The present approach enables us to provide the information about the possible multipole ordering in an unbiased way and is easily available for other localized f electron materials once the c states and f-c mixing elements are given from the bandstructure calculation.

1. Introduction

CeB₆ has been known as a typical and remarkable compound exhibiting a rich phase diagram of the multipole orderings²⁻⁴ and extensively studied experimentally⁵⁻⁻¹¹ and theoretically.¹²⁻²¹ Due to the large spin-orbit coupling (SOC) and the cubic crystalline electric field (CEF), the ground state of CeB₆ is the antiferro-quadrupolar (AFQ) ordering of the Ce²⁺ ion which is the Γ₄ quartet separated from the excited Γ₂ doublet by 540 K,⁸ and has a inherently the freedom of 15 active multipole moments as shown in Table I.

Up to now, three phases exist in temperature T and external magnetic field H plane of CeB₆. Normal phase (phase I) from room temperature down to a few K with H = 0 is a typical Kondo lattice metal with a highly-enhanced specific heat coefficient⁶⁰ C/T = 250 m mol/K². With decreasing H, phase II emerges at a critical temperature Tₒ = 3.2 K with the ordering wavevector q = (π, π, π) and is confirmed by the antiferro-quadrupolar (AFQ) ordering of the Γ₄ multipole quadrupoles (Oₓz, Oₓz, Oₓz). The ordering tendency of the Γ₄ multipole moment is supported from the elastic softening of C₄₄ at low temperature.¹⁵⁻¹⁸ Interestingly, Tₒ increases with increasing the applied field H, where the Γₒ₆ octupole Tₒ₆₆₆ moment is induced by H in addition to the Γ₄ multipole quadrupoles, which is well understood by the analysis of NMR.²¹ The phase III is a antiferro-magnetic (AFM) ordering of the Γ₄ magnetic moments (σₓ, σᵧ, σz) at Tₙ = 2.3 K with the double-σ₆ structure of Q₁ = (0, 0, 0) and Q₂ = (1/2, 1/2, 0, 0).

Phase IV electron state of CeB₆ is believed to be almost localized in Ce³⁺-ion from the several experiments of the magnetic and transport properties. More directly, the Fermi-surface (FS) has been observed in the de Haas-van Alphen (dHvA) experiments,²⁶⁻²⁷ the angle resolved photoemission spectroscopy (ARPES)²⁸⁻³⁰ and the high-resolution photoemission tomography.³¹ They has indicated an ellipsoidal FS centered at X point in the Brillouin zone (BZ) which is almost the same as that of LaB₆ with the 4f⁰ state. Hence the 4f state in CeB₆ is localized and hardly participates in the formation of FS.

In such a localized f electron picture, Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction²⁻⁶⁻⁹⁻¹⁰ plays an important role for the multipole ordering where the intersite coupling between the multipole moments of f electrons is mediated by the itinerant c band electrons.⁴⁻⁵,⁶⁻¹⁰⁻¹ⁱ The RKKY model of CeB₆ was proposed by Ohkawa³²,³³ firstly, and later developed by Shina et al.,³⁴,³⁵ where all 15 active multipole moments had been taken into account in correct symmetry, and reproduced the experimental T-H phase diagram where only nearest-neighbor couplings and the largest Γ₄ multipole couplings were assumed. This assumption was discussed from the symmetry of the RKKY couplings,³⁶,³⁷ but there was no explicit calculation for the signs and values of the couplings, and also no discussion about the long-range property of the RKKY multipole coupling of CeB₆. Later Sakurai et al.,³⁸,³⁹ studied the RKKY multipole couplings of CeB₆ microscopically such as the effect of the f⁰ and f² intermediate states, c band number dependence and the ratio of the f-c mixing elements described by the Slater-Koster (SK) parameters, but

| IRR [dimension] | vector | pseudospin | multipole |
|----------------|--------|------------|-----------|
| Γ₄z [0]       | ξ      | ρ         | 2/√3 Tₒ₆₆₆ |
| Γ₄z [1]       | η      | ρ         | 4/3 Tₒ₆₆₆ |
| Γ₄z [2]       | ζ      | ρ         | 2/√3 Tₒ₆₆₆ |

Table I. The irreducible representations (IRRs) and notations for the active multipole moments in Γ₄ subspace⁵ where J = (Jₓ, Jᵧ, Jₚ), is the dipole (octupole), Tₒ₆₆₆ = (Tₒ₆₆₆, Tₒ₆₆₆, Tₒ₆₆₆), ηₓ = −(π ± √3π)/2, ηᵧ = (π ± √3π)/2, and η (η) means even (odd) time-reversal symmetry. In this paper we call 1/2 Tₒ₆₆₆ and 1/4 (Oₓz, Oₓz) just as Tₒ₆₆₆ and (Oₓz, Oₓz), but all the multipole operators are normalized.

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plausible ordering moment types and wavevectors could not be obtained. As is often discussed in the RKKY mechanism, the c band states and their couplings with the f states in the realistic materials must be important for determining the ordering moment types and wavevectors. Therefore the microscopic description of the c band states and f-c mixing elements from the realistic bandstructure calculation is needed, though such studies are quite limited. In these studies, the c states is described by the Wannier orbitals obtained from the bandstructure calculation but the f-c mixing elements using the calculation of the RKKY coupling are treated by the SK parameter only with the nearest neighbor sites, where several arbitrary parameters and assumptions are included. Hence more decisive and widely-applicable approach reflecting the individual material properties is highly desired.

In this paper, we study the electronic states of CeB$_6$ and calculate the RKKY interaction based on the 74-orbital effective Wannier model derived from the bandstructure calculation directly. In Sec. 2, we calculate the bandstructures of CeB$_6$ and LaB$_6$ and construct the effective Wannier model of CeB$_6$, and examine the quasi-particles states and their multipole fluctuations based on the renormalized Wannier model in Sec. 3. Next in Sec. 4, we formulate the present RKKY mechanism based on the multi-orbital Kondo lattice model with both of the $\Gamma$$_3$ quartet and 60 c orbitals, and present the results of the RKKY multipole couplings for all moments as functions of the wavevector and intersite vectors. Finally we give the summary and discussion in Sec. 5.

2. Bandstructure calculation & Wannier model

2.1 DFT Bandstructure calculation of CeB$_6$ & LaB$_6$

First we calculate the electronic states of CeB$_6$ and LaB$_6$ by using the WIEN2k code$^{49-51}$ based on the framework of the density-functional theory (DFT) with the generalized gradient approximation (GGA).$^{52}$ The SOC is fully included within the second variation approximation. The crystallographic parameters are the space group $Pm\bar{3}m$ (No. 221), the lattice constant $a = 4.141$Å and the internal coordinates $(x/a,y/b,z/c) = (0,0,0)$ for Ce and $(\frac{1}{2},\frac{1}{2},u)$ for B with $u = (\sqrt{2} - 1)/2 \sim 0.2071$. In self-consistent calculation, we use 145 k-points in the irreducible part of the simple cubic BZ, the muffin-tin radii $R_{MT} = 2.50$ (1.62) a.u. for Ce (B) and the plane-wave cutoff of $R_{MT}k_{max} = 8$. For the calculation of LaB$_6$, we use the same parameters of CeB$_6$ but employ the GGA+U method with $U = 60$ eV for La-f level so as to eliminate the f weights in the c bands, since we focus the pure c band state of CeB$_6$ not bulk property of LaB$_6$.

The obtained bandstructures with the density-of-states (DOSs) and FSs are shown in Fig. 1 for CeB$_6$ [(a) & (b)] and LaB$_6$ [(c) & (d)]. In CeB$_6$, the large f contribution due to the 14 f spin-orbital states around Fermi energy ($E_F$) is observed with the strong peak of DOS as shown in the right panel of Fig. 1 (a). On the other hands in LaB$_6$ the f states is absent in the bandstructure and DOS [Fig. 1 (b)] as expected due to the effect of the GGA+U. Except for the f band states, the global bandstructures of CeB$_6$ and LaB$_6$ are closely resembled below and above $E_F$.

2.2 Construction of Wannier model for CeB$_6$

Next we construct the 74-orbital effective Wannier model based on the maximally localized Wannier functions (MLWFs) method$^{54-58}$ from the DFT bandstructure of CeB$_6$, where we prepare 14 f-states from Ce-f (7 orbital × 2 spin) and 60 c-states from Ce-d (5 orbital × 2 spin), Ce-s (1 orbital × 2 spin), B-p (6 site × 3 orbital × 2 spin) and B-s (6
site × 1 orbital × 2 spin) as basis functions, and set considerably wide energy window in order to ensure the good localization of Wannier orbitals in the disentanglement procedure. The obtained bandstructure of the Wannier model is plotted in Fig.1 (e) and (f) together with the DFT bandstructure of CeB₆ (black), where the Wannier model is well reproduced the DFT bandstructure up to E_F + 4 eV and the shapes of the Wannier orbitals are similar to the atomic-orbitals significantly.

The obtained model can be written by the following tight-binding (TB) Hamiltonian as,

$$H_{TB} = \sum_{ij} \sum_{nm} h_{ij}^{ff} f^\dagger_{im} f^\dagger_{jm} + \sum_{ij} \sum_{\ell \ell'} h_{ij}^{cc} c^\dagger_{i\ell} c_{i\ell'} + \sum_{ij} \sum_{ml} \left( V_{im,jf} f^\dagger_{im} c_{\ell} + h.c. \right), \quad (1)$$

where $f^\dagger_{im}$ ($c^\dagger_{\ell}$) is a creation operator for a f (c) electron with unit-cell $i$ and 14 (60) spin-orbital states $m$ ($\ell$). Here 14 f states of $m$ are represented by the CEF eigenstates as $\Gamma_8$ quartet and $\Gamma_7$ doublet with the total angular momentum $J = 5/2$, and $\Gamma_6$, $\Gamma_5$ doublets and $\Gamma_8$ quartet with $J = 7/2$. The f-f (c-c) matrix element of $h_{ij}^{ff}$ ($h_{ij}^{cc}$) includes the f (c) energy levels, SOC couplings, CEF splittings and f-f (c-c) hopping integrals, and $V_{im,jf}$ is the f-c mixing element which is finite only for the intersite terms due to the inversion symmetry. The wavevector $\mathbf{k}$-representation of $H_{TB}$ is given by,

$$H_{TB} = \sum_{k} \sum_{mm'} h_{mm'}^{ff}(\mathbf{k}) f^\dagger_{km} f_{km'} + \sum_{k} \sum_{\ell \ell'} h_{k\ell}^{cc} c^\dagger_{k\ell} c_{k\ell'} + \sum_{k} \sum_{m} \left( V_{km,jf} f^\dagger_{km} c_{\ell} + h.c. \right) = \sum_{k} \varepsilon_k a^\dagger_{k \ell} a_{k \ell}, \quad (2)$$

where $\varepsilon_k$ is the eigenenergy with $k$ and band-index $s$ and $a_{k \ell}^\dagger$ is a creation operator for an electron with $k$, $s$, which is transformed into $m$ and $\ell$ states as $a_{i \ell} = \sum_{k} u_{k \ell m} a_{k \ell m}$ where $u_{k \ell m}$ is the eigenvector component of $m$ ($\ell$) state.

Several atomic parameters are obtained from the Wannier model, such as the SOC splitting for Ce-4f between $J = 5/2$ and $J = 7/2$ states $\Delta_{SO} = 0.33$ eV close to the experimental value of 3000 K, the atomic CEF splitting between $\Gamma_8$ and $\Gamma_7$, $\Delta_{CEF} = 8.2$ meV which is smaller than the experimental value of 540 K (=46 meV). The f (c) electron number per unit-cell is $n^f = 1.24$ ($n^c = 20.86$) and the total number is $n_{tot} = 22$. All the f electron number for each CEF state becomes finite where $n^f(\Gamma_8) = 0.634$ and $n^f(\Gamma_7) = 0.205$ for $J = 5/2$ and $n^c(\Gamma_8) = 0.098$, $n^c(\Gamma_7) = 0.088$, and $n^c(\Gamma_5) = 0.216$ for $J = 7/2$, due to the considerable f-f hopping and f-c mixing, which is indispensable within the DFT-based calculation.

3. Quasi-particle band states & Multipole fluctuations

3.1 Renormalized tight-binding model

As mentioned in Sec. 2, the f electron state obtained here is fully itinerant and differs from the expected situation in the real material as $n^f(\Gamma_8) \sim 1$. In this section, we examine the change of the electronic states and its multipole fluctuations from the itinerant f band state to the localized f state when $n^f(\Gamma_8) = 1$ in the realistic CeB₆ bandstructures. For this purpose, we introduce a renormalization factor $Z_{mf}^m$ which is explicitly derived from the Fermi-liquid (FL) theory,³⁹ where the many-body correlation effect of the local f-f Coulomb interaction is introduced through the self-energy $\Sigma_{mf}(k, \varepsilon)$ which is almost local $\Sigma_{mf}(k, \varepsilon) = \Sigma_{mf}(\varepsilon)$ and can be expanded around $\varepsilon = 0$ by the following form,

$$\Sigma_{mf}(\varepsilon) = \Delta \varepsilon_m^f + \left( 1 - \frac{1}{Z_{mf}} \right) \varepsilon - i \gamma_m \varepsilon^2 + O(\varepsilon^3), \quad (3)$$

$$\Delta \varepsilon_m^f = \text{Re} \Sigma_{mf}^m(0), \quad Z_{mf}^m = \left( 1 - \frac{d}{d \varepsilon} \text{Re} \Sigma_{mf}^m(\varepsilon) \big|_{\varepsilon = 0} \right)^{-1}, \quad (4)$$

where $Z_{mf}^m$ corresponds to an inverse mass-enhancement $m/m'^*$, and $\Delta \varepsilon_m^f$ and $\gamma_m$ are a shift of the f energy-level and a damping rate of the quasi-particles respectively. Hence in the itinerant quasi-particle picture, our original model of $H_{TB}$ is renormalized by $Z_{mf}^m$ and $\Delta \varepsilon_m^f$, yielding the renormalized tight-binding model $H_{RTB}$ as explicitly given by,

$$H_{RTB} = \sum_{ij} \sum_{nm} \tilde{h}_{ij}^{ff} f^\dagger_{im} f^\dagger_{jm} + \sum_{ij} \sum_{\ell \ell'} \tilde{h}_{ij}^{cc} c^\dagger_{i\ell} c_{i\ell'} + \sum_{ij} \sum_{ml} \left( \tilde{V}_{im,jf} f^\dagger_{im} c_{\ell} + h.c. \right) \quad (5)$$

where the renormalized f-f (f-c) matrix elements $\tilde{h}_{ij}^{ff}$ ($\tilde{V}_{im,jf}$) are written as,

$$\tilde{h}_{ij}^{ff} = \begin{cases} \varepsilon_i^f + \Delta \varepsilon_{ij}^f, & i = j, \quad m = m' \varepsilon_i^f + \Delta \varepsilon_{ij}^f, & i \neq j \end{cases}, \quad (6)$$

$$\tilde{V}_{im,jf} = \sqrt{Z_{mf}^m} V_{im,jf} \quad (7)$$

where $\varepsilon_i^f$ is a f energy-level of the CEF state $m$, where $h_{ij}^{f'f''}$ is dropped for simplicity as $Z_{mf}^m = Z_f$ and $\Delta \varepsilon_{ij}^f = \Delta \varepsilon_f$, where $\Delta \varepsilon_f$ is set to $\Delta \varepsilon_f = 0.27$ eV so as to satisfy $n^f(\Gamma_8) = 1$ and $n^c = 21$ at $Z_f = 0$. Hence the f-f (f-c) hopping elements are renormalized by $Z_f (\sqrt{Z_f})$. Throughout the calculation, we determine a chemical potential $\mu$ so as to satisfy $n_{tot} = n^f + n^c = 22$ with $64^3 k$-meshes in the entire BZ.
3.2 Renormalized electronic states

Figure 2 (a) shows the $Z_f$-dependence of $f$ electron number per CEF eigenstates $n^f(\Gamma)$ with $T = 0.002$ eV, where $\Gamma = \Gamma_0^{5/2}, \Gamma_1^{5/2}, \Gamma_1^{7/2}, \Gamma_2^{7/2}$ and $\Gamma_3^{7/2}$, and $Z_f = 1$ ($Z_f = 0$) corresponds to the DFT-band (localized $f$) limit. With decreasing $Z_f$, $n^f(\Gamma)$ increases and finally becomes $n^f(\Gamma_3^{7/2}) = 1$ when $Z_f = 0$ while $n^f(\Gamma_1^{7/2})$ and all other $n^f(\Gamma_i^{7/2})$ decrease and reach zero at $Z_f = 0$. The change of $n^f(\Gamma)$ is rapidly for $Z_f < 0.1$ where the effective mass-enhancement reaches $m^*/m \gtrsim 10$. The $f$ electron magnetization $m_{spin} + m_{orb}$ as a function of $Z_f$ is also plotted in Fig. 2 (b) together with its spin, orbital and $J_z$-components $m_{spin}, m_{orb}$ and $m_{Jz}$, respectively, where the magnetic field is applied along the $z$-direction with $h = \mu_B H = 0.004$ eV. The Zeeman Hamiltonian is given by $H_Z = (\sigma^z + \Gamma) h$, and $m_{spin}$ and $m_{orb}$ are explicitly written as,

$$m_{spin} = -\mu_B \frac{1}{N} \sum_{k\sigma} \sum_{\mu \nu} \langle \sigma^z \rangle_{n\mu \nu} \epsilon_{k \mu \nu} u_{k \mu \nu}^\dagger f(\epsilon_{k \mu})$$

$$m_{orb} = -\mu_B \frac{1}{N} \sum_{k\sigma} \sum_{\mu \nu} \langle \sigma^z \rangle_{n\mu \nu} \epsilon_{k \mu \nu} u_{k \mu \nu}^\dagger f(\epsilon_{k \mu})$$

where $\sigma^z$ is a $z$-component of spin Pauli (orbital angular momentum) matrix for $m$-basis and $f(x)$ is the Fermi distribution function $f(x) = \frac{1}{\exp(x - \mu) + 1}$. With decreasing $Z_f$, $m_{rot}$ increases and finally reaches the saturated value of the $\Gamma_8$ state as $1.5\mu_B$ together with an opposite sign between $m_{orb}$ and $m_{spin}$ due to the SOC effect.

The $T$-dependence of the magnetization $m_{rot}$ and inverse magnetization $h/m_{rot}$ for several values of $Z_f$ are plotted in Fig. 2 (c) and (d) respectively. For $Z_f = 1 \sim 0.3$ the weak $T$-dependence of $m_{rot}$ is observed as a Pauli paramagnetic behavior of the itinerant $f$ electron, while for $Z_f < 0.1 m_{rot}$ increases with decreasing $T$, exhibiting the Curie paramagnetic behavior of the localized $f$ electron $m_{rot}/h \sim 1/T$, which is more clearly observed in the inverse magnetization $h/m_{rot}$ with a linear $T$-dependence. In such situations for $Z_f = 0.1 \sim 0.01$, the electronic state is similar to the purely localized $f$ electron on a single Ce-ion usually analyzed in the experiments. However in this study the $f$-$c$ mixings are still finite and the quasi-particle hybridization bands are formed with the wide-bandwidth $c$ band dispersion having the ellipsoidal FS observed ARPES of CeB$_6$.

Next we check such renormalized bandstructures for several values of $Z_f$ as shown in Fig. 3 (a)-(d) together with the DFT-bandstructure of CeB$_6$ [Fig. 3 (a)-(c)] and the LaB$_6$ GGA+$U$ band without $f$ weights [Fig. 3 (d)]. From $Z_f = 0.5$ [Fig. 3 (a)] to $Z_f = 0.1$ [Fig. 3 (b)], the whole bandstructures are still close to the DFT-band of CeB$_6$ but their $f$ bandwidths become narrow gradually, exhibiting a separation between the lower $J = 5/2$ bands and higher $J = 7/2$ bands. In Fig. 3 (d) with $Z_f = 0.01$ corresponding to $m^*/m \sim 100$, the almost flattened $J = 7/2$ bands, and $\Gamma_7$ and $\Gamma_5$ bands of $J = 5/2$ are clearly observed around $E_F$, and they slightly hybridize with the wide-bandwidth $c$ bands expanding from the X point in the BZ. Interestingly, the $c$ band dispersion with $Z_f = 0.01$ (red) is almost overlapping the LaB$_6$ band with the GGA+$U$ (black) as shown in Fig. 3 (d) except for the highly-flattened $f$ bands, resulting in the formation of almost the same FS of LaB$_6$. Hence the $c$ bands of CeB$_6$ with almost localized $f$ electron state coincides that of LaB$_6$ without the La-$f$ contribution, and then their FS is also almost the same as that of LaB$_6$ as shown in Fig. 1 (d). These results strongly support the localized $f$ electron picture for CeB$_6$ and then the approach based on the periodic Anderson model and its perturbation w. r. t. the $f$-$c$ mixing is expected to give a good starting point for treating this system.

3.3 Multipole fluctuations in the quasi-particle bands

Before going to the calculation of the RKKY interaction, we examine the multipole fluctuations under the renormalized $f$ bands on CeB$_6$ by calculating the multipole susceptibility $X_{Of}(q)$ with the multipole operator $O_f$ shown in Table I and the wavevector $q$ which is given by,

$$X_{O_f}(q) = \sum_{m_{1}m_{2}m_{3}m_{4}} O_{m_{1}m_{2}}^{\Gamma} O_{m_{3}m_{4}}^{\Gamma} X_{m_{1}m_{2}m_{3}m_{4}}(q)$$

$$X_{m_{1}m_{2}m_{3}m_{4}}(q) = \frac{1}{N} \sum_{k\mu\nu} u_{k\mu\nu} u_{k\mu\nu}^{\dagger} q_{\mu\nu}^{\dagger} m_{k}$$

where $O_f = \sum_{mm'} O_{mm'}^{\Gamma} f_{m'}^{\Gamma} f_{mm'}^{\Gamma}$ and $O_{mm'}^{\Gamma}$ is the normalized $4 \times 4$ matrix element of $O_f$ in $\Gamma_8$ subspace, and $X_{m_{1}m_{2}m_{3}m_{4}}(q)$ is the irreducible $f$ electron susceptibility which depends on the distribution of $f$ states in the bandstructures through the renormalized $f$-$c$ mixing $j_{z}^f N_{f,m_{1}m_{2}}$. The $q$-dependence of $X_{O_f}(q)$ for each multipole moment with $Z_f = 1$ and $\Delta f = 0 \, eV$ corresponding to the DFT band limit as shown in Fig. 1 (a) (e) and (f). The obtained $q$-dependence is considerably weak and the explicit values of $X_{O_f}(q)$ fall within the only small range $X_{O_f}(q) \sim 4 \sim 5 \, eV^{-1}$ for all multipole moments and wavevectors $q$. Among them the $j_{z}^f$ magnetic multipole $\sigma^z$ susceptibility, where $\sigma^x$ and $\sigma^y$ are degenerate with $\sigma^z$, is barely large for the incommensurate wavevector around $q = (\pi, \pi, \pi)$, while the...
The weak-$q$ does not become large for the AFQ wavevector $\Gamma_3$, quadrupole $O_3$ and the $\Gamma_{2u}$ octupole $T_{2u}$ susceptibilities does not become large for the AFQ wavevector $q = (\pi, \pi, \pi)$. The weak-$q$ dependence of $\chi_{O_3}(q)$ becomes more notable for the almost localized $f$ case with $Z_f = 0.01$ and $\Delta E_f = 0.27$ eV as shown in Fig. 4 (b), where $\sigma^2$ becomes also maximum but its wavevector shifts to $q = (0, 0, 0)$ as shown in the inset of Fig. 4 (b).

In such a situation, the actual value of $\chi_{O_3}(q)$ becomes huge, where the extremely narrow $\Gamma_3$ bands are located in the very near and just above $E_F$ with tiny $f$-$c$ mixing, and then the hybridized band $\varepsilon_{kk}$ is highly degenerate for wide-range of the BZ, giving rise to the sizable enhancement of the Lindhard function of in Eq. (11). As far as such $q$-independent $\chi_{O_3}(q)$, it is difficult to describe the development of the $(\pi, \pi, \pi)$-AFQ mode with $(O_{2x}, O_{2y}, O_{2z})$ by the perturbation of the $f$-$f$ Coulomb interaction such as the random phase approximation (RPA) and its extensions.

4. RKKY Interaction of CeB$_6$

4.1 Derivation of RKKY Hamiltonian

Here we consider the RKKY interaction between the multipole moments of $\Gamma_3$ quartet. For this purpose, we eliminate the $f$ energy-levels but use the $f$-$c$ mixing of the original Wannier model. The $c$ bandstructure for the calculation of RKKY couplings is shown in Fig. 5 (a) and (b), which is almost the same as that of LaB$_6$ as mentioned in Sec. 3 and is compared to the strongly renormalized quasi-particles case with $Z_f = 0.01$ as shown in Fig. 5 (c) and (d). During the calculation, $\mu$ is determined so as to keep $n_{tot} = n^r = 21$ and $T$ is set to $T = 0.005$ eV.

The multi-orbital Kondo lattice Hamiltonian for the present model is given by,

$$
H_{MKL} = \sum_{k\ell} h_{\ell\ell}^{c*}(k)c_{\ell\ell}^{c}e_{kk'}^c + \sum_{i} \sum_{\text{num}} \sum_{kk'} \sum_{\ell\ell'} J_{\text{i}}^{k\ell\ell'} f_{\text{i}m}^{k\ell} f_{\text{i}m}^{k\ell'} c_{\ell\ell'}^{c} c_{\ell\ell'}^{c},
$$

where $m$ represents 4-states in $\Gamma_3$ quartet $|m\rangle = |1\rangle \sim |4\rangle$ with an degenerate energy-level $e_{kk'}^c$, which are given with the $J$-base of $J = 5/2 \ |JM\rangle$ explicitly as, $|1\rangle = -\sqrt{\frac{5}{147}} |1\rangle - \sqrt{\frac{5}{147}} |\ell\rangle$, $|2\rangle = |+\frac{1}{2}\rangle$, $|3\rangle = |\ell\rangle$ and $|4\rangle = \sqrt{\frac{5}{147}} |-\frac{1}{2}\rangle + \sqrt{\frac{5}{147}} |\ell\rangle$. The c-c matrix element $h_{\ell\ell}^{c*}(k)$ includes the c-orbital energy $e_{kk'}^c$ for $\ell = \ell'$ and the c-c hopping $t_{\ell\ell'}^{c*}(k)$ for $\ell \neq \ell'$. The first term is rewritten by the c band eigenstate $c_{kk'} = \sum_{\ell} u_{k\ell}^{c} c_{\ell\ell}^{c}$ with the eigenenergy $\varepsilon_{kk'}$ and eigenvector $u_{k\ell}^{c*}$.

The Kondo coupling $J_{\text{i}}^{k\ell\ell'}$ in the second term consists of the $f^0$- and $f^2$-intermediate process. In this paper, we take simple two assumptions for $J_{\text{i}}^{k\ell\ell'}$: (1) only $f^0$-process is considered and the contribution of $f^2$-process is same as that of $f^0$-process and, (2) the scattered c orbital energies are fixed to $\mu$, namely $e_{kk'}^c, e_{kk'}^c \rightarrow \mu$. Then the Kondo coupling
where the prefactor 2 comes from the assumption (1) and \( V_{km\ell} \) is the \( k \)-represented \( f \)-c mixing element in Eq. (2).

The RKKY Hamiltonian can be obtained from the second-order perturbation w. r. t. the second term of \( H_{MKL} \), together with the thermal average for the \( c \) states. The final form is given by,

\[
H_{\text{RKKY}} = -\sum_{\langle ij \rangle} \sum_{m_1m_2m_3m_4} V_{km_1j} V_{km_2i} e^{-i(k-k')R_i} \langle f_{jm_1}^\dagger f_{jm_2}^\dagger f_{jm_3} f_{jm_4} \rangle, \tag{14}
\]

where \( V_{km_1j} \) is the RKKY coupling between the states \( m_1, m_2 \) at the unit-cell \( R_j \) and the states \( m_3, m_4 \) at \( R_i \) and \( \langle ij \rangle \) represents a summation for the intercell vectors \( R_{ij} = R_i - R_j \). The key quantity \( K_{m_1m_2m_3m_4}(q) \) is given by,

\[
K_{m_1m_2m_3m_4}(q) = \frac{1}{N} \sum_q V_{km_1j} V_{km_2i} e^{iq(R_i-R_j)}, \tag{15}
\]

which consists of a square of the energy denominator \((\mu - \epsilon_{k\ell}^q)^2\), 4-product \( f \)-c mixings and the \( c \) band eigenvectors, and the Lindhard function with \( \epsilon_{ks} \). Thus it has \( 4^2 = 256 \) components of \( f \)-basis \( m_1, m_2, m_3, m_4 \) for each \( q \), and has to be summed for the \( f \) orbitals \([\ell_1, \ell_2, \ell_3, \ell_4]\) (604) and the band-index \( [s, s'] \) (60²). Then we introduce a \( f \)-c mixing matrix \( V_{km_1j}^\dagger V_{km_2i}^\dagger \) between \( m_1, m_2 \) via the \( c \) band state with \( k, s \) as follows,

\[
V_{km_1j}^\dagger V_{km_2i}^\dagger = \frac{1}{N} \sum_{kss'} V_{km_1j}^\dagger V_{km_2i}^\dagger e^{i(qk-k's')}, \tag{17}
\]

which includes whole information about the \( f \) state scattering between \( m_1, m_2 \) through the \( c \) state with \( k, s \), and has only \( 4^2 = 16 \) components of \( m_1, m_2 \) for each \( k, s \) with a summation for \([\ell, \ell']\) (60²). Hence once we calculate \( V_{km_1j}^\dagger V_{km_2i}^\dagger \), \( K_{m_1m_2m_3m_4}(q) \) can be easily obtained by using the following compact form,

\[
K_{m_1m_2m_3m_4}(q) = \frac{1}{N} \sum_{kss'} \sum_{m_1m_2m_3} V_{km_1j}^\dagger V_{km_2i}^\dagger e^{i(qk-k's')} (\mu - \epsilon_{k\ell}^q)^2 \epsilon_{ks} - \epsilon_{k+q,s'}, \tag{18}
\]

This expression helps us calculate all the contributions of the 60 \( f \) electron charge and/or orbital fluctuations to the RKKY multipole couplings.

In order to search the actual multipole ordering, we employ the mean-field (MF) approximation w. r. t. the multipole operator \( O_{q} \), resulting in the MF Hamiltonian as follows,

\[
H_{\text{RKKY}}^{\text{MF}} = -\sum_{\langle ij \rangle} \sum_{m_1m_2m_3m_4} K_{m_1m_2m_3m_4}(q) \langle f_{jm_1}^\dagger f_{jm_2}^\dagger f_{jm_3} f_{jm_4} \rangle, \tag{19}
\]

where the multipole coupling \( K_{m_1m_2m_3m_4}(q) = K_{m_1m_2m_3m_4}^{\text{MF}} \) and \( K_{m_1m_2m_3m_4}^{\text{MF}}(q) \) and the MF order parameter \( \delta O_{q} \) are given by,

\[
K_{m_1m_2m_3m_4}(q) = \sum_{m_1m_2m_3m_4} \langle f_{jm_1}^\dagger f_{jm_2}^\dagger f_{jm_3} f_{jm_4} \rangle \delta O_{m_1m_2m_3m_4}, \tag{20}
\]

\[
\delta O_{q} = \frac{1}{N} \sum_{\langle ij \rangle} \delta O_{ij}(R_i) e^{i(qR_i)} \epsilon_{qR_i}, \tag{21}
\]

where \( K_{m_1m_2m_3m_4}^{\text{MF}}(q) = (1/N) \sum_q K_{m_1m_2m_3m_4}(q) \) and \( \delta O_{q}(R_i) \) is MF multipole order parameter defined at the unit-cell vector \( R_i \). Then the MF multipole susceptibility \( \chi_{O_q}^{\text{MF}}(q) \) is written by,

\[
\chi_{O_q}^{\text{MF}}(q) = \frac{K_{m_1m_2m_3m_4}(q)}{1 - \chi_{O_q}(q) K_{m_1m_2m_3m_4}(q)}, \tag{22}
\]

which is enhanced towards the multipole ordering instability for the ordering moment \( O_{q} \) and wavevector \( q \), and finally diverges at a critical point of the multipole ordering transition temperature \( T = T_{O_q} \) where \( \chi_{O_q}^{\text{MF}}(q) \) reaches unity. The \( q \)-dependence of \( \chi_{O_q}(q) \) is weak as shown in Sec. 3, and then the sign and maximum value of \( \chi_{O_q}^{\text{MF}}(q) \) determines the multipole ordering moment and wavevector for any given \( T \). Hereafter we set \( \mu - \epsilon_{k\ell}^q = 1 \text{ eV} \) for simplicity, since this factor is independent of \( q \) and \( m \), and hence does not affect the ordering type and wavevector, whose effect is discussed in Sec. 5.

4.2 \( q \)-dependence of RKKY coupling \( \chi_{O_q}(q) \)

The obtained RKKY multipole couplings \( \chi_{O_q}(q) \) for several multipole moments along the high symmetry line in the BZ are plotted as shown in Fig. 6 (a)-(d), where the positive (negative) coupling for a certain multipole \( O_{q} \) and wavevector \( q \) enhances (suppresses) the corresponding multipole fluctuation as explained in Eq. (22), and its positive maximum value gives a leading multipole ordering mode. The obtained results for the leading multipole ordering modes upto the 10th largest coupling are summarized in Table I.

The couplings of the \( \Gamma_5 \) quadrupoles \( (O_{zzz}, O_{zz}O_{zz}) \) become largest among all moments and \( q \), which perfectly corresponds to the AFQ ordering of CeB\(_6\). In addition, \( \Gamma_{2a} \) octupole \( T_{3z} \) coupling is quite large and comparable to the \( \Gamma_5 \) quadrupoles with the same wavevector as shown in Fig. 6 (a) but slightly small within the present calculation accuracy as shown in Table II, which seems to be the same value from the previous discussions\(^{36,37}\) where \( O_{xyz} \) and \( T_{3z} \) have almost same matrix elements and yield the similar fluctuations in phase I. Furthermore the quadrupoles \( (O_{x}, O_{zz}O_{z}) \) and octupole \( T_{3z} \) couplings also take a substantial peak for \( q = (0,0,0) \) as shown in Fig. 6 (a) and (d) and correspond to the elastic softening of \( C_{4z}^{15-18} \).

The next largest coupling is the \( \Gamma_{5a} \) octupole \( \zeta^z \) at \( q = (\pi,0,0) \) [Fig. 6 (a) & (d)] which is degenerate for \( \zeta^x \) \([\zeta^y]\) octupole at \( q = (0,\pi,0) \) \([0,0,\pi]\) due to the cubic symmetry. The role of the \( \Gamma_{5a} \) octupoles \( \zeta^x, \zeta^y, \zeta^z \) is also discussed for the phase IV observed in the La-doping system Ce\(_{2-x}\)La\(_x\)B\(_6\) with \( x < 0.36 - 0.66 \) where the \( q = (\pi,0,0) \) antiferro-octupolar (AFQ) ordering of \( (\zeta^x+\zeta^y+\zeta^z)/\sqrt{3} \) is considered to be a possible mode. In contrast, the present theory suggests the \( \Gamma_{5a} \) AFO with the domainic structure of \( \zeta^x, \zeta^y, \zeta^z \) and \( \zeta^y \) for \( q = (\pi,0,0), (0,\pi,0) \) and \( (0,0,\pi) \), respectively, and this point will be discussed in the next subsection.

In addition to this, the \( \Gamma_{3z} \) quadrupole \( O_{zz} \) coupling is quite large for \( q = (0,0,0) \) (not shown) and becomes similar value of the octupole coupling \( \zeta^z \) as shown in Table II, which is also degenerate for the rotated moments to the each principle-axis \( O_{zz} \) and \( O_{zz} \). This is namely the \( \Gamma_{3z} \) AFQ mode where the moment directions and wavevectors
are perpendicular such as the multipole moments of $O_{2z\rightarrow z}$, $O_{2z\rightarrow y}$ and $O_{2z\rightarrow x}$ with the corresponding wavevectors for $q = (\pi, 0, 0), (0, \pi, 0)$ and $(0, 0, \pi)$ respectively.

The $\Gamma_{4u}$ magnetic multipole couplings of $(\sigma^x, \sigma^y, \sigma^z)$ and $(\eta^x, \eta^y, \eta^z)$ are plotted in Fig. 4 (d) and their maximum values in $q$-space are smaller than that of the quadrupole and octupole couplings as shown in Table II, where the $\Gamma_{4u}$ maximum peak values are less than half of the first leading peak value of the $\Gamma_{5g}$-$c(\pi, \pi, \pi)$. The RKKY ordering vectors for phase III, $\mathbf{Q}_1$ and $\mathbf{Q}_2$, the couplings of the magnetic multipoles $(\sigma^x, \sigma^y, \sigma^z)$ have small peaks as shown in Fig. 4 (e) and they shall be enhanced and dominant only when the system enters into phase II, which is not discussed in the present paper.

As usually discussed in the itinerant $f$ electron picture with the multi-orbital Hubbard model,\textsuperscript{67–69} the weak coupling theory like the RPA and its extensions yields largely enhanced magnetic multipole (spin) fluctuations which become always larger than the nonmagnetic multipole (orbital) fluctuations like $\Gamma_{5g}$ multipoles here. As for CeB$_6$, the $f$ electron itself is already localized at each Ce site and the remained magnetic and nonmagnetic multipole moments interact with the RKKY intersite couplings, where the magnetic multipole coupling does not necessarily dominate over the nonmagnetic one, since the dominant RKKY coupling is determined by the detail of the $f$-$c$ mixing and the mediating $c$ electron charge and/or orbital fluctuations.

Here we note the $c$ electron charge and orbital fluctuations and their contribution to the coupling $\mathbf{R}_{\Omega_{c}}(q)$. By changing summation for the orbital-set in Eq. (16) and the band-index in Eq. (18), we have obtained that both effects of the Ce-$d_{z^2}$ orbitals of $d_{2z^2-r^2}$ and $d_{3z^2-r^2}$ distributed in band10 and band11 and the charge fluctuation of B$_6$-molecule having a maximum at $q = (\pi, \pi, \pi)$ and large values along R-M line play significant roles for the $\Gamma_{5g}$ AFQ mode. In particular, we observe a non-negligible contribution from the 23th-band which does not have FS but is very close to $E_F$ along the $\Gamma$-M direction as shown in Fig. 5 (b). The explicit results and further analysis of such $c$ electron contributions to the multipole couplings will be presented in elsewhere.

4.3 $R_{ij}$ dependence of RKKY coupling $K_{\Omega_{c}}(\mathbf{R}_{ij})$

In general, the RKKY interaction is known to have long-range and oscillating features discussed in the early studies.\textsuperscript{35,36} For the multipole ordering of CeB$_6$, however, the coupling is limited only in the nearest neighbor terms in the previous studies.\textsuperscript{32–39} In contrast, the present formalism provides the real space dependent couplings $K_{\Omega_{c}}(\mathbf{R}_{ij})$ which is explicitly written as

$$K_{\Omega_{c}}(\mathbf{R}_{ij}) = \sum_{m_1m_2m_3m_4} C_{m_1m_2}^{F} C_{m_3m_4}^{F} K_{m_1m_2m_3m_4}(\mathbf{R}_{ij}),$$

(23)

where $K_{m_1m_2m_3m_4}(\mathbf{R}_{ij})$ is given in Eq. (15).

Figure 7 shows the site-dependence of the RKKY multipole couplings $K_{\Omega_{c}}(\mathbf{R}_{ij})$ with the intersite vector $\mathbf{R}_{ij}$ up to 20-th neighbor sites as shown in Table III, where the positive (negative) sign corresponds to the ferro (antiferro) coupling for each neighboring site.

As shown in Fig. 7 (a), the $\Gamma_{1g}^{(1)}$ magnetic multipole $(\sigma^x, \sigma^y, \sigma^z)$ couplings exhibit several sign changes with a few site-intervals and degeneracy due to the symmetry of paramagnetic phase for all $n$-th neighbors, where $\sigma^x$, $\sigma^y$ and $\sigma^z$ corresponds to the $x$, $y$- and $z$-moment direction respectively.

We also confirm that a monopole operator is defined as a unit matrix for $\Gamma_8$-basis is also degenerate with $(\sigma^x, \sigma^y, \sigma^z)$ possessing the same oscillating feature. The couplings of $I$ and
same value for 6 first neighbor sites

\[ \Gamma \]

pling together with the corresponding moment types, wavevectors, maximum

\[ O \]

ing AFQ mode show staggered and isotropic behaviors, where

\[ q \]

first neighbors

\[ x \]

with decreasing

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\[ \frac{1}{2} \]

with the corresponding wavevectors for

\[ T \]

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pling depending the coordination number for each site, since

\[ \Gamma \]

a La-substituted site has no multipole moment. Consequently,

\[ f \]

ff

\[ \sigma \]

ff

\[ R \]

ff

\[ \frac{1}{2} \]

ff

\[ \alpha \]

ff

\[ \Gamma \]

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roughly estimated by $\mu$ in Sec. 4, we take
obtained from the DFT-bandstructure calculation. As mentioned
since the $f$-process is fully one-body effect and directly ob-
tained from the DFT-bandstructure calculation. As mentioned
in Sec. 4, we take $\mu - \epsilon_{f}^{0} = 1$ eV, but the excitation en-
ergy from the $f^{0}$-stable to $f^{0}$-intermediate states in CeB$_{6}$ is
roughly estimated by $\mu - \epsilon_{f}^{0} = 2 - 4$ eV,$^{29,31}$ so that our
results of $\overline{K}_{0}$ obtained in Sec. 4 should be multiplied by a
single reduction factor $\frac{1}{T} \sim \frac{1}{10}$, which yields the same order of
the actual transition temperature of CeB$_{6}$ as a few K, for ex-
ample, the inter-quadrupole coupling value $K_{f}^{q} = 2.1$ K.$^{17}$

The explicit determination of the couplings and the trans-
formation temperatures needs the many-body energy differ-
cence between the ground and intermediate $f^{0}$ and $f^{0}$ states, where to
what extent the many-body effect from the $f^{0}$ and more mul-
tiple $f$ processes changes the present result is an important
question elucidating the multipole ordering system with dif-
ferent valence materials such as PrB$_{6}$ and NdB$_{6}$. The explicit
calculation of the coupling including the $f^{0}$-process and/or
more many-body contribution, and the whole phase diagram
in $T$-$H$ plane will be presented in the subsequent paper.

As a complementary approach to the present localized $f$
electron treatment, the dynamical mean field theory$^{30}$ en-
abling to take account of the full local correlation effect and
its extensions$^{71}$ including the intersite correlation could be
valid for directly describing the fully localized $f$ states start-
ing from the itinerant $f$ states and their multiple ordering
phenomena including superconductivity.$^{72}$ The application of
such many-body theory to the realistic materials and their
comparison with present theory are also the essential future
problems.

We would like to thank Y. Ōno and Y. Iizuka for valuable
comments and discussions.

| $n$-th neighbor sites and distances for the intersite vectors between Ce-Ce $R_{ij} = a (n_{1}e_{x} + n_{2}e_{y} + n_{3}e_{z})$ where $\{e_{x}, e_{y}, e_{z}\}$ are unit vectors along $x, y, z$-direction. | $\hat{R}_{ij}$/a |
|---|---|
| 1 | (1,0,0) |
| 2 | (1,1,0) |
| 3 | (1,1,1) |
| 4 | (2,0,0) |
| 5 | (2,1,0) |
| 6 | (2,1,1) |
| 7 | (2,2,0) |
| 8 | (2,2,1) |
| 9 | (3,0,0) |
| 10 | (3,1,0) |
| 11 | (3,1,1) |
| 12 | (2,2,2) |
| 13 | (3,2,0) |
| 14 | (3,2,1) |
| 15 | (4,0,0) |
| 16 | (4,1,0) |
| 17 | (3,2,2) |
| 18 | (4,1,1) |
| 19 | (3,3,0) |
| 20 | (3,3,1) |

Table III. The $n$-th neighbor sites and distances for the intersite vectors between Ce-Ce $R_{ij} = a (n_{1}e_{x} + n_{2}e_{y} + n_{3}e_{z})$ where $\{e_{x}, e_{y}, e_{z}\}$ are unit vectors along $x, y, z$-direction.

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