

Orbital-dependent electronic masses in Ce heavy-fermion materials studied via Gutzwiller density-functional theory

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A series of cerium-based heavy fermion materials is studied using a combination of local density functional theory and the many-body Gutzwiller approximation. Computed orbital-dependent electronic mass enhancement parameters are compared with available experimental data extracted from measured values of the Sommerfeld coefficient. Gutzwiller density functional theory is shown to remarkably follow the trends across a variety of Ce compounds and to give important insights into the phenomenon of orbital-selective mass renormalization which in turn allows for a better understanding of a wide spread of data.

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I. INTRODUCTION

Heavy-fermion materials pose one of the greatest challenges in condensed matter physics. Their low-temperature linear specific heat coefficient can be up to 1000 times larger than the value expected from the free-electron theory; their magnetic moments can be screened by the Kondo effect and their electrical resistivity is frequently divergent but sometimes superconductivity can emerge at low temperatures [1].

Theoretical calculations based on density functional theory (DFT) in its popular local density approximation (LDA) [2] fail to reproduce strongly renormalized electronic masses in heavy-fermion materials due to improper treatment of many-body-correlation effects. Consider, for example, a well-known class of cerium heavy-fermion materials, such as two famous phases (α and γ) of cerium itself [3–5], so-called Ce-115s systems: CeXIn₅ ($X = \text{Co, Rh, Ir}$) [6–8] and numerous Ce-122 compounds [9–21]: CeX₂Si₂ ($X = \text{Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Ag}$). Table I gives evaluated via LDA densities of states specific heat coefficients γ for these materials as compared with experiments, where in many cases a factor of 2–20 error exists in underestimating γ while, in some cases, such, as CeCo₂Si₂, an overestimation occurs. It is the purpose of this work to show that a better treatment of electronic correlations via the recently introduced Gutzwiller density functional theory [22–25] can correct most of these errors and uncover exactly which orbitals become heavy, as is also illustrated in Table I.

The physics of the electronic mass enhancement is controlled by a low-frequency behavior of the local electronic self-energy which can be encoded in the simple Taylor-like form

$$\Sigma_{\alpha}(\omega) = \Sigma_{\alpha}(0) + (z_{\alpha}^{-1} - 1)\omega + \dots, \quad (1)$$

where we assume, for simplicity, the existence of some crystal-field representation $|\alpha\rangle$ that diagonalizes the self-energy matrix in a spin-orbital space of the localized f electrons. This expression suggests two main effects that may occur when correlations are brought into consideration on top of a band theory such as LDA: first the crystal-field correction to a local f -electron level is controlled by $\Sigma_{\alpha}(0)$ and, second, the actual band narrowing is controlled by a quasiparticle residue

parameter z_{α} . Both effects would affect our comparisons of γ in Table I.

Recently, advanced many-body approaches based on combinations of density functional and dynamical mean-field theory (DMFT) have been implemented [26] to study heavy-fermion systems [27,28] where self-consistent solutions of either Anderson- or Kondo-impurity problems have been done using most accurate continuous-time quantum Monte Carlo method [29,30]. While DMFT deals with full frequency dependent self-energy and is a lot more computationally demanding than traditional LDA, it taught us an important lesson in the so-called orbital selectivity in the Mott transition problem, i.e., when crystal-field dependent self-energies can reduce the effective degeneracy of the impurity. This affects the proximity of the quasiparticle residue z to become equal to zero when the ratio between the Hubbard U and the bandwidth W changes.

A recently introduced Gutzwiller density functional theory (GDFT) and the so-called LDA + G method [22–25] is a simplified variational approach that relies on the Gutzwiller approximation initially introduced to study itinerant ferromagnetism of the one-band Hubbard model [31], and later extended to multiband systems [32,33]. In this method, the atomic configurations of correlated orbitals are treated by adjusting their weights using a variational procedure. This leads to renormalized energy bands and mass enhancements for the electrons. The approximation was extensively studied in the limit of infinite dimensions [34,35] and was shown to be equivalent to a slave-boson mean-field theory [36] for both single-band and multiband models [37–39]. In LDA + G, the Gutzwiller-type trial wave function $\hat{P}|0\rangle$ is adopted with $|0\rangle$ and \hat{P} being the LDA ground state and the Gutzwiller projector, respectively. After further application of the Gutzwiller approximation, an effective Hamiltonian describing the dynamics of quasiparticles was obtained as $H_{\text{eff}} = \hat{P}H_{\text{LDA}}\hat{P}$, which contains two important features discussed above: the modification of the crystal and spin-orbital fields and the quasiparticle weight z_{α} . Thus, the method exactly casts the effect encoded into the low-frequency behavior of $\Sigma(\omega)$ [Eq. (1)].

The benchmark of the LDA + G scheme was demonstrated in Ref. [23]. For a nonmagnetic correlated metal SrVO₃, it

TABLE I. Comparison between calculated using LDA density of states and experimentally extracted specific heat coefficients γ and the extracted quasiparticle residues $z_{\text{expt}} = \gamma_{\text{LDA}}/\gamma_{\text{expt}}$ for a number of cerium-based heavy-fermion compounds considered in this work. The last columns show the predictions of γ and z using the LDA + G method with the values of $U = 4$ and 5 eV as well as the reference to the specific orbital degeneracy of the $j = 5/2$ manifold that exhibits the strongest enhancement.

	$N_{\text{LDA}}(0)$ St./(Ry cell)	γ_{LDA} mJ/(mol K ²)	γ_{expt} mJ/(mol K ²)	Ref. to γ_{expt}	z_{expt}	$\gamma_{\text{LDA+G}}$ ($U = 4$ eV)	$\gamma_{\text{LDA+G}}$ ($U = 5$ eV)	$z_{\text{LDA+G}}$ ($U = 4$ eV)	$z_{\text{LDA+G}}$ ($U = 5$ eV)	Orbital $j = 5/2$
α Ce	36	6.2	13	[3]	0.48			0.55	0.33	Γ_7, Γ_8
γ Ce	49	8.5						0.14	0.11	Γ_7, Γ_8
115s:										
CeCoIn ₅	150	26.0	290	[6]	0.09	70	104	0.20	0.13	$\Gamma_6, 2\Gamma_7$
CeRhIn ₅	156	27.2	420	[7]	0.065	75	120	0.19	0.12	$\Gamma_6, 2\Gamma_7$
CeIrIn ₅	165	28.6	720	[8]	0.04	79	210	0.14	0.07	$\Gamma_6, 2\Gamma_7$
122s:										
CeMn ₂ Si ₂	184	31.9	47	[9]	0.68	52	84	0.51	0.43	$\Gamma_6, 2\Gamma_7$
CeFe ₂ Si ₂	85	14.7	22	[10]	0.67	21	24	0.53	0.47	$\Gamma_6, 2\Gamma_7$
CeCo ₂ Si ₂	110	19.0	10	[11]	1.9?	35	43	0.47	0.36	$\Gamma_6, 2\Gamma_7$
CeNi ₂ Si ₂	115	19.9	33	[12]	0.60	27	29	0.47	0.43	$\Gamma_6, 2\Gamma_7$
CeCu ₂ Si ₂	64	11.1	1000	[13–15]	0.01	430	∞	0.10	0	Γ_6
CeRu ₂ Si ₂	103	17.8	350	[16–18]	0.05	70	190	0.33	0.13	$2\Gamma_7$
CeRh ₂ Si ₂	106	18.3	130	[19]	0.14	35	120	0.36	0.14	$2\Gamma_7$
CePd ₃ Si ₂	100	17.3	65–110	[20,21]	0.15–0.26	170	∞	0.045	0	Γ_7
CeAg ₂ Si ₂	205	35.5				140	430	0.10	0.06	Γ_6

produced narrower bands and larger effective masses than those found in standard LDA. Also the method was able to get a photoemission peak missed in the LDA calculation. These improvements are very close to the experimental results. Later, in Ref. [40], a complex phase diagram of Na_xCoO₃ was correctly reproduced. Recently, this method has been successfully applied to FeAs-based superconductors [25,41] and to Ce metal [42,43].

In this work we address the physics of cerium heavy-fermion materials via the use of the LDA + G approach. A great spread in the extracted values of mass enhancements data shown in Table I together with some unphysical values of $z > 1$ prompts us that in many classes of real compounds both the orbital selectivity encoded via the shifts $\Sigma_\alpha(0)$ on top of the LDA as well as the quasiparticle residues play an important role and have to be treated on the same footing. It therefore represents a stringent test of a many-body electronic-structure method such as LDA + G to heavy-fermion materials. In particular, in its recent application to elemental cerium [43], it has been shown that the spin-orbital splitting of the f level is renormalized by correlations and pushes energies of the $J = 7/2$ manifold up relative to $J = 5/2$ states. This resulted in lowering the degeneracy from 14 to 6 and in a greater mass enhancement of $J = 5/2$ manifold as compared to a non-spin-orbit-coupled calculation.

The paper is organized as follows: In Sec. II, the LDA + G method is described. The results for several typical families of heavy-fermion materials are presented in Sec. III. Finally, Sec. IV is the conclusion.

II. THE LDA + G METHOD

The Gutzwiller density functional theory and the LDA + G approximation have been described previously [22–25]. Here we merely summarize the equations of the method that we

implement using a linear muffin-tin orbital formalism [44] that includes both the full potential terms [45] and relativistic spin-orbit-coupling operator variationally [44].

A. Gutzwiller approximation

We first illustrate the method using a general multi-orbital Hubbard model. The Hamiltonian is

$$H = H_0 + H_{\text{int}} = \sum_{ij, \alpha\beta} t_{ij}^{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta} + \sum_i \sum_{\alpha\alpha'\beta\beta'} U_i^{\alpha\beta\alpha'\beta'} c_{i\alpha}^\dagger c_{i\alpha'}^\dagger c_{i\beta} c_{i\beta'}, \quad (2)$$

where $\alpha, \beta = 1, \dots, 2N$ are the spin-orbital indexes of the localized orbital, N is the number of orbitals, e.g., 7 for the f orbital. The first term is a tight-binding Hamiltonian which can be extracted from the LDA calculation. The second term represents the on-site Coulomb repulsion, $U_{\alpha\beta\alpha'\beta'} = \langle \alpha\alpha' | \frac{e^2}{r} | \beta\beta' \rangle$, although in actual calculations we assume it to be diagonal: $U_{\alpha\beta\alpha'\beta'} = U \delta_{\alpha\beta} \delta_{\alpha'\beta'}$.

In the atomic limit, for the localized orbital there are $2N$ different states which can be either occupied or empty. Therefore there is a total of 2^{2N} configurations $|\Gamma\rangle$. All these configurations form a complete basis and the density-density interaction is diagonal in this configuration space. It is obvious that these configurations should not be equally weighted. In the Gutzwiller method, we adjust the weight of each configuration. Therefore, it is convenient to construct projection operators that project onto a specific configuration Γ at site i :

$$\hat{m}_{i\Gamma} = |i\Gamma\rangle\langle i\Gamma|. \quad (3)$$

When the interaction term is absent, the ground state is the Hartree uncorrelated wave function (HWF) $|\Psi_0\rangle$ which is a Slater determinant of the single-particle states. When the

interaction is switched on, this wave function is no longer a good approximation. In the Gutzwiller method, we project the wave function onto a Gutzwiller wave function (GWF) $|\Psi_G\rangle$ by adjusting the weight of each configuration through variational parameters $\lambda_{i\Gamma}$ ($0 \leq \lambda_{i\Gamma} \leq 1$):

$$|\Psi_G\rangle = \hat{\mathcal{P}}|\Psi_0\rangle = \prod_i \hat{P}_i|\Psi_0\rangle, \quad (4)$$

where

$$\hat{P}_i = \sum_{\Gamma} \lambda_{i\Gamma} \hat{m}_{i\Gamma}. \quad (5)$$

Notice that when all $\lambda_{i\Gamma} = 1$, the GWF is going back to HWF. At the same time, setting $\lambda_{i\Gamma} = 0$ removes configuration Γ at site i . Therefore, a perfectly localized atomic state of site i is described by all $\lambda_{i\Gamma} = 0$ except for one, and in this way, the Gutzwiller wave function captures both the itinerant and localized behavior of the system.

It is a difficult task to evaluate GWF. However, within the Gutzwiller method, we can map any operator \hat{O} acting on the GWF to a corresponding effective \hat{A}^G which acts on the HWF:

$$\langle \Psi_G | \hat{A} | \Psi_G \rangle = \langle \Psi_0 | \hat{\mathcal{P}}^\dagger \hat{A} \hat{\mathcal{P}} | \Psi_0 \rangle = \langle \Psi_0 | \hat{A}^G | \Psi_0 \rangle, \quad (6)$$

where

$$\hat{A}^G = \hat{\mathcal{P}}^\dagger \hat{A} \hat{\mathcal{P}}. \quad (7)$$

Specifically, when the operator \hat{A} is a single-particle operator, e.g., $\hat{A} = \sum_{ij,\alpha\beta} A_{ij}^{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta}$ where $A_{ij}^{\alpha\beta} = \langle i\alpha | \hat{A} | j\beta \rangle$, the Gutzwiller effective operator can be written as

$$\begin{aligned} \hat{A}^G &= \sum_{ij,\alpha\beta} \sqrt{z_{i\alpha}} A_{ij}^{\alpha\beta} \sqrt{z_{j\beta}} c_{i\alpha}^\dagger c_{j\beta} \\ &+ \sum_{i,\alpha} A_{ii}^{\alpha\alpha} (1 - z_{i\alpha}) c_{i\alpha}^\dagger c_{i\alpha}, \end{aligned} \quad (8)$$

where $z_{i\alpha}$ are the orbital-dependent quasiparticle residues: $0 \leq z_{i\alpha} \leq 1$. These are determined by the configuration weights:

$$\sqrt{z_{i\alpha}} = \sum_{\Gamma'} \frac{\sqrt{m_{i\Gamma'} m_{i\Gamma}} |\langle i\Gamma' | c_{i\alpha}^\dagger | i\Gamma \rangle|}{\sqrt{n_{i\alpha} (1 - n_{i\alpha})}}, \quad (9)$$

where $m_{i\Gamma} = \langle \Psi_G | \hat{m}_{i\Gamma} | \Psi_G \rangle$ and $n_{i\alpha}$ are the occupation numbers for the orbitals. Although, generally, the z s should be considered as matrices over orbital index, here we neglect their off-diagonal elements for simplicity.

B. Combination with local density approximation

Similar to the idea of the LDA + U or LDA + DMFT methods, we add the interaction term on top of the LDA calculation. The Hamiltonian is given by

$$H = H_{\text{LDA}} + H_{\text{int}} - H_{\text{DC}}, \quad (10)$$

where H_{LDA} is the LDA Hamiltonian, which has the same form as H_0 in Eq. (2), H_{int} is the on-site interaction term for the set of correlated orbitals, such as the f orbitals of heavy-fermion materials considered in this work. Since the LDA calculation has already included the Coulomb interaction in some averaged level, we need to subtract the double-counting term H_{DC} from LDA. Various forms of H_{DC} will be discussed later.

The Kohn–Sham approach uses a noninteracting system as a reference which keeps the same ground-state density as the interacting one. However, now our ground state is the GWF instead of the HWF. Therefore, we need to transform the Hamiltonian into the effective one in the $|\Psi_0\rangle$ basis. Since the Hamiltonian H_{LDA} is a single-particle operator, following Ref. [24] we obtain

$$\begin{aligned} H_{\text{LDA}}^G &= \langle \Psi_G | H_{\text{LDA}} | \Psi_G \rangle \\ &= \left(\sum_{\alpha i} \sqrt{z_{i\alpha}} |\phi_{\alpha i}\rangle \langle \phi_{\alpha i}| + 1 - \sum_{\alpha i} |\phi_{\alpha i}\rangle \langle \phi_{\alpha i}| \right) \\ &\quad \times H_{\text{LDA}} \left(\sum_{\beta j} \sqrt{z_{j\beta}} |\phi_{\beta j}\rangle \langle \phi_{\beta j}| + 1 - \sum_{\beta j} |\phi_{\beta j}\rangle \langle \phi_{\beta j}| \right) \\ &\quad + \sum_{\alpha i} (1 - z_{i\alpha}) |\phi_{\alpha i}\rangle \langle \phi_{\alpha i}| H_{\text{LDA}} |\phi_{\alpha i}\rangle \langle \phi_{\alpha i}|, \end{aligned}$$

where $|\phi_{\alpha i}\rangle$ represents a complete basis set of the correlated orbitals, and where we omit site index i from the quasiparticle residues $z_{i\alpha}$ due to lattice periodicity.

The interaction term acting on the GWF produces

$$H_{\text{int}}^G = \langle \Psi_G | H_{\text{int}} | \Psi_G \rangle = \sum_{i\Gamma} E_{\Gamma} m_{\Gamma}. \quad (11)$$

The expectation value of the total Hamiltonian gives us the total energy as a functional of the density ρ and configurational weights m_{Γ} :

$$E(\rho, \{m_{\Gamma}\}) = \langle \Psi_0 | H_{\text{LDA}}^G | \Psi_0 \rangle + \sum_{\Gamma} E_{\Gamma} m_{\Gamma} - E_{\text{DC}}. \quad (12)$$

A minimization similar to LDA is now performed. Representing density in terms of the Kohn–Sham states produces the equations for the quasiparticles:

$$\begin{aligned} \frac{\partial E(\rho, \{m_{\Gamma}\})}{\partial \langle \psi_{nk} |} &= \left(H_{\text{LDA}}^G + \sum_{\alpha} \left[\frac{\partial E}{\partial z_{\alpha}} \frac{\partial z_{\alpha}}{\partial n_{\alpha}} - \frac{\partial E_{\text{DC}}}{\partial n_{\alpha}} \right] |\phi_{\alpha}\rangle \langle \phi_{\alpha}| \right) \\ &\quad \times |\psi_{nk}\rangle = \epsilon_{nk} |\psi_{nk}\rangle, \end{aligned} \quad (13)$$

$$\frac{\partial E(\rho)}{\partial m_{\Gamma}} = \sum_{\alpha} \frac{\partial E}{\partial z_{\alpha}} \frac{\partial z_{\alpha}}{\partial m_{\Gamma}} + E_{\Gamma} = 0. \quad (14)$$

Recalling the self-energy linear expansion [Eq. (1)], we see from Eq. (13) that the effective Hamiltonian to be diagonalized has the following form

$$H^G = H_{\text{LDA}}^G + \sum_{\alpha} [\Sigma_{\alpha}(0) - V_{\text{DC},\alpha} z_{\alpha}] |\phi_{\alpha}\rangle \langle \phi_{\alpha}|, \quad (15)$$

where $\Sigma_{\alpha}(0)$ and $V_{\text{DC},\alpha}$ are directly associated with various total-energy derivatives that appeared in Eq. (13).

C. Gutzwiller projected Hamiltonian

It is convenient to represent all matrices in the space of the Bloch eigenvalues $\epsilon_{\mathbf{k}j}$ and wave functions $|\mathbf{k}j\rangle$ that are obtained from the LDA calculation. The Gutzwiller

Hamiltonian to be diagonalized is given by

$$\begin{aligned} \langle \mathbf{k}j' | H^G | \mathbf{k}j \rangle = & \sum_{j''} \epsilon_{\mathbf{k}''j''} \left(\sum_{\alpha} \sqrt{z_{\alpha}} \langle \mathbf{k}j' | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j'' \rangle + \delta_{j'j''} - \sum_{\alpha} \langle \mathbf{k}j' | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j'' \rangle \right) \\ & \times \left(\sum_{\beta} \sqrt{z_{\beta}} \langle \mathbf{k}j'' | \phi_{\beta} \rangle \langle \phi_{\beta} | \mathbf{k}j \rangle + \delta_{j''j} - \sum_{\beta} \langle \mathbf{k}j'' | \phi_{\beta} \rangle \langle \phi_{\beta} | \mathbf{k}j \rangle \right) \\ & + \sum_{\alpha} (1 - z_{\alpha}) \langle \mathbf{k}j' | \phi_{\alpha} \rangle \epsilon_{\alpha} \langle \phi_{\alpha} | \mathbf{k}j \rangle + \sum_{\alpha} (\Sigma_{\alpha}(0) - V_{DC,\alpha}) z_{\alpha} \langle \mathbf{k}j' | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j \rangle, \end{aligned} \quad (16)$$

where a subset of correlated orbitals $|\phi_{\alpha}\rangle$ is introduced. Their levels are given by

$$\epsilon_{\alpha} = \sum_{\mathbf{k}''j''} \epsilon_{\mathbf{k}''j''} \langle \phi_{\alpha} | \mathbf{k}''j'' \rangle \langle \mathbf{k}''j'' | \phi_{\alpha} \rangle. \quad (17)$$

The quasiparticle residues z_{α} and the level shifts $\Sigma_{\alpha}(0)$ are obtained using the Gutzwiller procedure [24]. The double-counting potential $V_{DC,\alpha}$ corrects for the fact that the LDA already accounts for some of the correlation effects in a

mean-field manner. The eigenvalue problem

$$\sum_j (\langle \mathbf{k}j' | H^G | \mathbf{k}j \rangle - \delta_{j'j} E_{\mathbf{k}n}) B_j^{\mathbf{k}n} = 0 \quad (18)$$

produces renormalized energy bands $E_{\mathbf{k}n}$ and wave functions $\sum_j B_j^{\mathbf{k}n} |\mathbf{k}j\rangle$ of the quasiparticles.

D. Charge density

In order to find a new density, we calculate a Gutzwiller density matrix operator in the LDA representation:

$$\begin{aligned} \rho_{j'j}^{G\mathbf{k}} = & \sum_{j''} f_{\mathbf{k}j''} \left(\sum_{\alpha} \sqrt{z_{\alpha}} \langle \mathbf{k}j' | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j'' \rangle + \langle \mathbf{k}j' | \mathbf{k}j'' \rangle - \sum_{\alpha} \langle \mathbf{k}j' | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j'' \rangle \right) \\ & \times \left(\sum_{\beta} \sqrt{z_{\beta}} \langle \mathbf{k}j'' | \phi_{\beta} \rangle \langle \phi_{\beta} | \mathbf{k}j \rangle + \langle \mathbf{k}j'' | \mathbf{k}j \rangle - \sum_{\beta} \langle \mathbf{k}j'' | \phi_{\beta} \rangle \langle \phi_{\beta} | \mathbf{k}j \rangle \right) + \sum_{\alpha} \langle \mathbf{k}j' | \phi_{\alpha} \rangle (1 - z_{\alpha}) \rho_{\alpha} \langle \phi_{\alpha} | \mathbf{k}j \rangle, \end{aligned} \quad (19)$$

where

$$\rho_{\alpha} = \sum_{\mathbf{k}j} f_{\mathbf{k}j} \langle \mathbf{k}j | \phi_{\alpha} \rangle \langle \phi_{\alpha} | \mathbf{k}j \rangle. \quad (20)$$

Diagonalizing it produces new occupation numbers $n_{\mathbf{k}\lambda}$

$$\sum_j (\rho_{j'j}^{G\mathbf{k}} - \delta_{j'j} n_{\mathbf{k}\lambda}) C_j^{\mathbf{k}\lambda} = 0, \quad (21)$$

so that the density of quasiparticles in real space is given by

$$\rho^G(\mathbf{r}) = \sum_{\mathbf{k}\lambda} n_{\mathbf{k}\lambda} \left(\sum_{j'} C_{j'}^{\mathbf{k}\lambda} | \mathbf{k}j' \rangle \right) \left(\sum_j C_j^{\mathbf{k}\lambda*} \langle \mathbf{k}j | \right). \quad (22)$$

E. Incompleteness of basis

To see the importance of the issue, let us examine a shift of the LDA eigenstates $\epsilon_{\mathbf{k}j}$ by an arbitrary value x . We obtain the new Gutzwiller Hamiltonian as the old one plus the correction

$$\langle \mathbf{k}j' | \tilde{H}^G | \mathbf{k}j \rangle = \langle \mathbf{k}j' | H^G | \mathbf{k}j \rangle + x o_{j'j}(\mathbf{k}), \quad (23)$$

where $o_{j'j}(\mathbf{k})$ is a matrix that can be proved to be equal to $\delta_{j'j}$ only under the assumption that the LDA wave functions form a mathematically complete basis set, i.e.,

$$\sum_{j''} \langle \phi_{\alpha} | \mathbf{k}j'' \rangle \langle \mathbf{k}j'' | \phi_{\beta} \rangle = \delta_{\alpha\beta}. \quad (24)$$

Unfortunately, modern electronic-structure methods deal with finite basis sets, and the last relationship is only approximately satisfied. As a result, different choices of the energy zero for the LDA eigenvalues $\epsilon_{\mathbf{k}j}$ may lead to slightly different output, although in our application to well-localized cerium $4f$ electrons, this introduces only minor noise in our calculated results. In the following, we always assume that the LDA eigenvalues are measured with respect to the Fermi energy which is the only physically relevant energy in this problem.

F. Double-counting potential

As one sees from Eq. (16) the actual self-energy correction used in the LDA + G calculation is $\Sigma_{\alpha}(0) - V_{DC,\alpha}$. Frequently, a so-called LDA + U version [46] of the double counting potential $V_{DC,\alpha}$ is used, that for our case is just an orbital-independent energy shift given by

$$V_{DC}^{\text{LDA}+U} = U(dn_f - 1/2), \quad (25)$$

where n_f is the average number of f electrons which at the Ce f shell is close to unity. As one sees, this correction has just an overall level shift by $U/2$ and does not modify the Gutzwiller extracted spin-orbit and crystal fields encoded in the α dependence of $\Sigma_{\alpha}(0)$. Unfortunately, it is not exactly clear whether the overall level shift of Ce f electrons has a physical effect, since the standard LDA + U double counting

was introduced in connection to the Hartree–Fock value of the self-energy which is the value at infinite frequency $\Sigma(\infty)$. It therefore may not be suited for correcting the low-energy physics of heavy-fermion systems.

As a result, in this work we adopt a different strategy in order to elucidate the physics of orbital selectivity in Ce heavy-fermion compounds: our calculations are first performed without $\Sigma_\alpha(0)$ correction assuming that the double-counting potential

$$V_{\text{DC},\alpha}^{(1)} = \Sigma_\alpha(0). \quad (26)$$

It has an important justification that the LDA calculated Fermi surfaces do not acquire any modifications as was evident from some heavy-fermion uranium compounds [47]. Second, we introduce the crystal-field-averaged double counting

$$V_{\text{DC}}^{(2)} = \frac{1}{N} \sum_{\alpha} \Sigma_{\alpha}(0), \quad (27)$$

which keeps the average position of the f level intact but allows for its crystal-field modifications found self-consistently via the LDA + G procedure. Since both $V_{\text{DC},\alpha}^{(1)}$ and $V_{\text{DC},\alpha}^{(2)}$ rely on Gutzwiller extracted $\Sigma_{\alpha}(0)$, which itself is obtained from the LDA + G functional minimization procedure, the entire method is still variational and allows an accurate estimate of the total energy. Comparing calculations with two types of double counting, important conclusions can be drawn about exactly which orbitals of a given heavy-fermion system play a major role in its electronic mass enhancement.

III. RESULTS AND DISCUSSION

We are interested in calculating the mass enhancement parameters of cerium f electrons which in a simple single-band theory would be given by the ratio of m^*/m_{LDA} . A common approach to extract this data is to compare the values of the Sommerfeld coefficient γ evaluated using the LDA density of states at the Fermi level $N_{\text{LDA}}(0)$:

$$\gamma_{\text{LDA}} = \frac{\pi^2}{3} k_B N_{\text{LDA}}(0), \quad (28)$$

with the measured electronic specific heat which, according to Fermi-liquid theory, behaves at low temperatures as γT . However, some care should be taken when adopting this procedure. First, densities of states of real systems include multiband features and contributions from both heavy and light electrons. Second, LDA densities of states assume some crystal-field effects which should in general be supplemented by many-body corrections encoded in $\Sigma_{\alpha}(0)$. Therefore not only band narrowing but also level shifts are expected to occur in real life on top of LDA. Third, many of the materials discussed in our work undergo either antiferromagnetic or superconducting transition before reaching $T \rightarrow 0$ limit. We quote the data for γ_{expt} in Table I using the data for their lowest temperature paramagnetic phases.

There are several other ways to access this information that we are going to use in this work. Optical spectroscopy experiments can provide access to effective masses but those are frequency dependent. Angle-resolved photoemission spectroscopy (ARPES) experiments measure directly one-electron

spectral functions

$$A(\mathbf{k},\omega) = \frac{|\text{Im}\Sigma(\mathbf{k},\omega)|}{[\omega - \text{Re}\Sigma(\mathbf{k},\omega)]^2 + \text{Im}\Sigma^2(\mathbf{k},\omega)}. \quad (29)$$

Here $\text{Re}\Sigma$ is the real part and $\text{Im}\Sigma$ is the imaginary part of the electronic self-energy. Under the assumption of the locality of self-energy, the quasiparticle residues

$$z = \left(1 - \frac{\partial \text{Re}\Sigma(\omega)}{\partial \omega}\right)^{-1} \quad (30)$$

can be extracted by comparing ARPES spectra against calculated LDA energy bands.

The de-Haas—van-Alphen (dHvA) effect is another powerful experimental technique which measures the properties of the Fermi surface under applied magnetic field [48]. LDA calculations can identify each cyclotron orbit seen by the dHvA experiment and find corresponding effective masses. However, complexity in the shapes of three-dimensional (3D) Fermi surfaces in real systems also makes this method not perfect.

It is remarkable that the LDA + G calculation returns the orbital-dependent quasiparticle residues z_{α} directly. In Table I, we are quoting these mass enhancement data and *not* those obtained via our calculated LDA + G densities of states.

A. α Ce and γ Ce

We first discuss our calculations for the cerium metal which is famous for its isostructural phase transition from its α to γ phase that is accompanied by a 15% volume expansion, and which has attracted great attention in the past [4,5,49–52] and current [43] literature. It is remarkable that our LDA + G calculation can correct most of the error in predicting the volume of the α phase for both types of the double countings that we explore in this work. Moreover, as Fig. 1 illustrates, it clearly shows a double-well type of behavior for the energy vs volume [with smaller (larger) minima corresponding to α (γ) phases] when using the crystal-field-averaged double counting $V_{\text{DC}}^{(2)}$ and the Hubbard U s within the range between 3.5 and 5.5 eV. This is in accord with the previous LDA + DMFT studies for cerium [50]. Similar behavior has been also found when studying $\alpha \rightarrow \delta$ transition in metallic plutonium [53].

The specific heat measurement of α Ce gives its Sommerfeld coefficient $\gamma \sim 13$ mJ/(mol K²) [3]. The optical spectroscopy experiment [54] estimates the effective mass to be $\sim 6m_e$ in α Ce and to be $\sim 20m_e$ in γ Ce, indicating the itinerant (localized) features of the α (γ) phases. However, the estimated optical effective masses are frequency dependent.

Figure 2 shows the dependence of the quasiparticle residues as a function of the Hubbard U for volumes corresponding to α and γ Ce where the left (right) plots represent our calculations with $V_{\text{DC}}^{(1)}$ ($V_{\text{DC}}^{(2)}$) type of double countings. It is clear when crystal or spin-orbital corrections are not taken into account (Fig. 2, left plot), the effective masses for various cerium orbitals are very similar in values and are not very strongly enhanced even for large values of U . (We use relativistic cubic harmonics representation where $j = 5/2$ level is split onto Γ_7, Γ_8 , and $j = 7/2$ onto $\Gamma_6, \Gamma_7, \Gamma_8$ states.) The situation changes dramatically when we account for the local self-energy correction (Fig. 2, right plot): a Coulomb renormalized spin-orbit splitting pushes Γ_7, Γ_8 states of $j = 5/2$ manifold

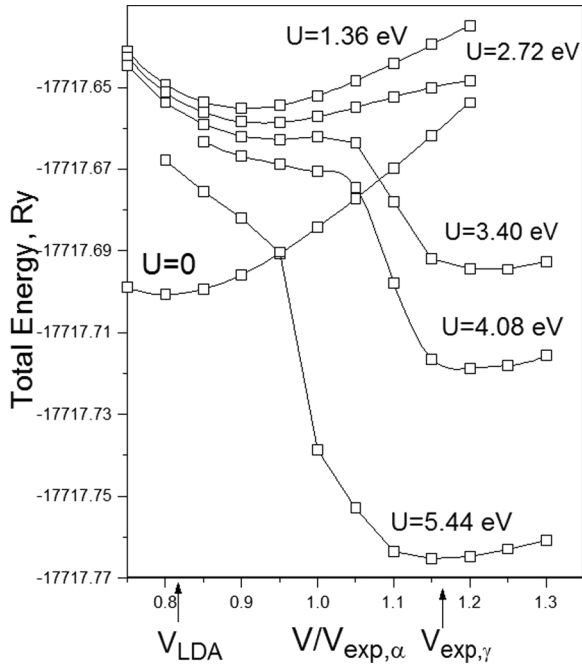


FIG. 1. Calculated total energy vs volume using the LDA + G method with several values of Hubbard U .

down and $\Gamma_6, \Gamma_7, \Gamma_8$ states of $j = 7/2$ manifold up. This results in redistributing the occupancies of the f electrons which now reside mainly at the $j = 5/2$ level. Thus, the effective degeneracy is 6 instead of 14 and the quasiparticle residues become much more sensitive to the values of U .

Actual comparison of the quasiparticle residues with experiment needs an accurate estimate of the Hubbard U for cerium f electrons which is typically around 5 eV, although, if the Hund's rule with $J \sim 1$ eV (important for the virtual f^2

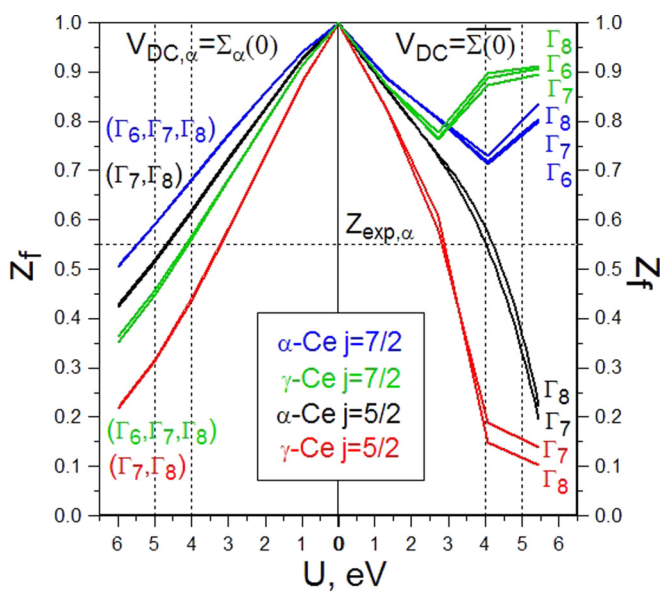


FIG. 2. (Color online) Calculated dependence of quasiparticle residues as a function of Hubbard U in α and γ phases of cerium metal using the LDA + G approach.

state) is taken into account, the effective interaction is reduced a little bit to $U - J$. Due to a great sensitivity of quasiparticle residues to this parameter, and for the purposes of our work, we simply show in Table I the range of z s that one can obtain for the values of U in the range between 4 and 5 eV. For α Ce we find them to be between 0.55 and 0.33 which are values close to their experimental estimates. This is also consistent with the result of the LDA + DMFT calculation [55]. While there is no specific heat data for the γ phase, optical measurements [54] show a factor of 3 to 4 enhanced masses as compared to the ones of the α phase, and our reduced values of z s are in accord with this trend.

B. Ce-115s

We next discuss our applications to the so-called 115 series of cerium heavy-fermion compounds CeMIn_5 ($M = \text{Co, Rh, Ir}$). They have a tetragonal HoCoGa_5 -type structure which results in additional splitting of all Γ_8 quadruplets into Γ_6 and Γ_7 doublets. Despite having similar crystal structure and almost identical LDA electronic structures, these systems show very different properties, a fact which has attracted great interest. CeCoIn_5 is a heavy-fermion superconductor with critical temperature $T_c = 2.3$ K, highest known in Ce-based systems [6] and with a Sommerfeld coefficient $\gamma \cong 290$ mJ/(mol K²) measured just above T_c . CeIrIn_5 is also a superconductor with $T_c = 0.4$ K [8] with $\gamma = 720$ mJ/(mol K²) above T_c that is nearly temperature independent. CeRhIn_5 , on the other hand, is an antiferromagnet with Néel temperature $T_N = 3$ K but becomes a superconductor with $T_c = 2.1$ K above a critical pressure $P_c \sim 16$ kbar [7]. From the C/T data, there is a peak at $T_N = 3.8$ K, indicating the onset of magnetic ordering. In order to find the electronic specific heat, one needs to use isostructural, nonmagnetic LaRhIn_5 to subtract the lattice contribution to C . However, it is difficult to define precisely the electronic specific heat above T_N due to the peak at T_N . A simple entropy-balance construction gives a Sommerfeld coefficient $\gamma \geq 420$ mJ/(mol K²) for $T > T_N$.

Those different properties are considered as the result of the localized vs itinerant nature of the $4f$ electrons. The dHvA measurements for CeCoIn_5 have shown the effective cyclotron masses to be within the range from $9m_0$ to $20m_0$ which is consistent with the specific heat data [56,57]. The LDA calculation with a model of itinerant f electrons shows a reasonable agreement with the dHvA data [58] while complete localization of the f electrons is needed to get the agreement with the angle-resolved photoemission spectroscopy (ARPES) data [59]. The dHvA experiment has been performed also for the antiferromagnetic state of CeRhIn_5 [60,61]. Although an earlier LDA calculation with the itinerant model shows some agreement with the experimental data [61], the localized nature of the f electrons was confirmed by the dHvA measurements in $\text{Ce}_x\text{La}_{1-x}\text{RhIn}_5$ [62] and by comparing the Fermi surfaces between CeRhIn_5 and LaRhIn_5 [63]. With the application of pressure, the dramatic change of the Fermi surface was observed indicating the change from the localized antiferromagnetic state to the itinerant heavy-fermion state [64]. For CeIrIn_5 , there is some experimental controversy. The effective cyclotron mass m_c^* is observed in the range from $6.3m_e$ to $45m_e$, indicating a large enhancement [65]. LDA

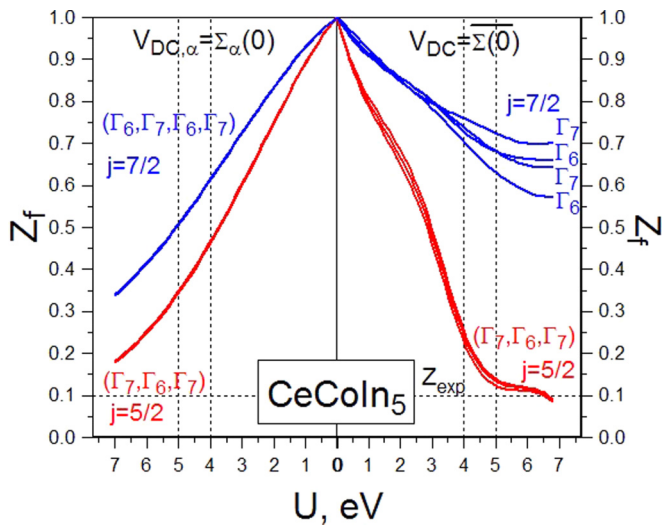


FIG. 3. (Color online) Calculated dependence of quasiparticle residues as a function of Hubbard U in CeCoIn_5 using the LDA + G approach.

with itinerant f electrons [58,65] explains well the geometry and the volume of the Fermi surface but the band masses are much smaller than the cyclotron masses. The photoemission spectrum is well described by the LDA + DMFT calculation [27] where the degree of itineracy in CeIrIn_5 is thought to be even larger than in CeCoIn_5 [66]. Also, this method shows the calculated effective masses to be of the same order as the experimental ones [67]. On the other hand, the ARPES study [68,69] shows that CeIrIn_5 and CeRhIn_5 have nearly localized $4f$ electrons.

The results of our paramagnetic LDA + G calculations for all three 115 compounds are presented in Table I. Similar to our calculation for cerium, we find that the calculated effective masses are only moderately enhanced ($z \sim 0.3$ to 0.5) if we do not account for the crystal-field or spin-orbit corrections to the self-energy on top of the LDA. When we perform self-consistent calculations including level shifts, much smaller values of the quasiparticle residues can be reached. Figure 3 illustrates this behavior for CeCoIn_5 . The situation here is similar to cerium where the spin-orbit coupling gets renormalized by correlations making the effective degeneracy of the f electrons equal 6. Actual values of the quasiparticle residues in Table I are given for U equals 4 and 5 eV: we see that the estimated z is the largest for CeCoIn_5 while the f electrons are more localized in CeRhIn_5 and CeIrIn_5 . The residual discrepancies can be attributed to the above-discussed uncertainties seen in experiments and also to the intrinsic error connected to the Gutzwiller procedure, as our prior studies of the performance of this method against quantum Monte Carlo have shown a 30% overestimation of z s [70].

C. Ce-122s

We finally discuss our applications to Ce 122 types of systems. By itself, RM_2X_2 is an enormous class of ternary intermetallic compounds with over a hundred members, where R is a rare-earth element, M denotes a transition metal ($3d$, $4d$, or $5d$), and X is either silicon or germanium.

After its first discovery of superconductivity with a transition temperature $T_c \simeq 0.5$ K in CeCu_2Si_2 [13], the interest in this family awakens, especially due to the interplay between antiferromagnetic and superconducting orders. Here we focus on the subclass $\text{Ce}M_2\text{Si}_2$ ($M = \text{Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Ag}$) where all members have body-centered tetragonal ThCr_2Si_2 -type structure with space group $I4/mmm$.

For the $M = 3d$ series, no magnetic order is found except for CeMn_2Si_2 where the Mn local moments order below 379 K [71]. For CeCu_2Si_2 , the electronic specific heat coefficient $\gamma \simeq 1000$ mJ/(mol K²) is the largest one among this family. On the other hand, CeFe_2Si_2 , CeCo_2Si_2 , and CeNi_2Si_2 are weak paramagnets with relatively small values of γ which are shown in Table I. These three compounds are also known as valence fluctuation systems.

For $M = 4d$ series, first, CeRu_2Si_2 is known as an archetypal Kondo lattice compound: it is a paramagnet with a relative large $\gamma \simeq 350$ mJ/(mol K²). The other three are antiferromagnets at low temperatures: CeRh_2Si_2 has the highest ordering temperature 36–39 K [72,73] while CePd_2Si_2 [21,72] and CeAg_2Si_2 [72,74] order antiferromagnetically at 8.5–10 K and 8–10 K, respectively. With the application of pressure, superconductivity was found in CeRh_2Si_2 [75] and in CePd_2Si_2 [76]. CeRu_2Si_2 is unique in this subclass since the superconductivity is not observed down to a few mK. This makes it the best target material for studying its heavy-fermion state. Interestingly, here a metamagnetic transition was found and extensively studied [77]. The cyclotron effective mass $m_c \sim 120m_e$ is observed in the dHvA experiment indicating the renormalized heavy-fermion state [78]. The electronic structure calculation using LDA with itinerant f -electron model qualitatively explains the dHvA data [79–81]. The metamagnetic transition was also studied by dHvA experiments [82–84] demonstrating that the f -electron character is changed from itinerant to localized across the metamagnetic transition. All members of this family of compounds were put into the Doniach (T_N vs J_K) phase diagram [85]. Later, using the LDA + DMFT scheme, the phase diagram was renewed [28].

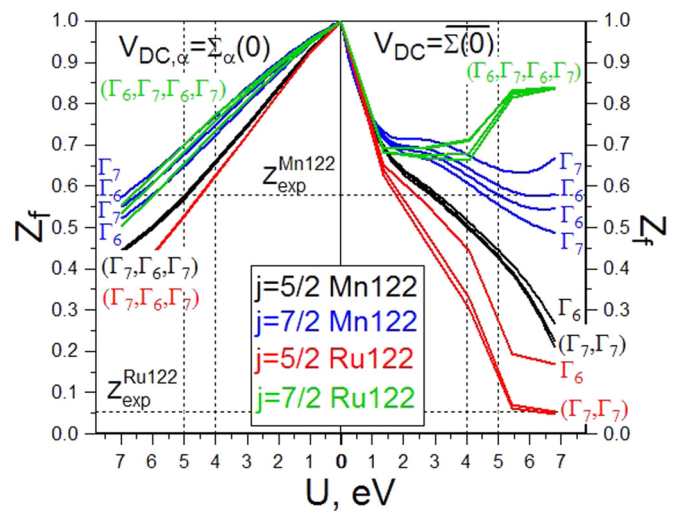


FIG. 4. (Color online) Calculated dependence of quasiparticle residues as a function of Hubbard U in CeMn_2Si_2 and CeRu_2Si_2 using the LDA + G approach.

Results of our applications to Ce 122s are presented in Table I. They assume a paramagnetic heavy-fermion state for all systems and the experimental γ s are extracted from the specific heat data measured above the temperatures of antiferromagnetic-superconducting transition. For the 122 systems with $3d$ elements, such as Mn, Fe, Co, and Ni, the LDA + G procedure returns only moderately enhanced electron masses which we find in agreement with experiment. An example of the dependence of z_α vs U is shown in Fig. 4 for Mn based 122, where the Coulomb interaction renormalizing spin-orbit splitting is essential to reduce the orbital degeneracy from 14 to 6 when using the crystal-field-averaged double counting, $V_{DC}^{(2)}$. This is similar to our findings in Ce and Ce-115 systems. Our calculation for Cu based 122 shows that its quasiparticle residues are very sensitive to the values of U above 4 eV. In fact, they begin to reach almost zero when U approaches 5 eV. It is also known experimentally that this system shows enormous mass enhancement just before it goes into the superconducting state.

The 122 compounds with $4d$ elements (Ru, Rh, Pd, Ag) exhibit strongly renormalized quasiparticle masses. Our LDA + G calculations listed in Table I correctly follow this trend where we see a strong reduction of z s as compared to our calculations with $3d$ elements. We also find another interesting effect: with increasing U the effective degeneracy of the f electrons acquires a reduction not only due to the Coulomb-assisted renormalization of spin-orbit splitting but also due to renormalization of the crystal fields: Fig. 4 shows

this behavior for Ru-122 where we see that the values of z become different for various crystal-field levels of the $j = 5/2$ manifold: one Γ_6 and two Γ_7 doublets. We find that a similar effect occurs in all other $4d$ types of Ce 122s and the last column of Table I lists our results showing which particular orbitals exhibit the strongest mass enhancement.

IV. CONCLUSION

In conclusion, using a recently proposed LDA + G approach we have studied quasiparticle mass renormalizations in several classes of Ce heavy-fermion compounds. We find that the calculation gives correct trends across various systems as compared to the measured Sommerfeld coefficient and reproduces the order of magnitude of the experimental value. We also uncover an interesting orbital dependency of the quasiparticle residues for each studied compound which provides an important physical insight to how correlations affect the effective degeneracy of cerium f electrons placed in various crystallographic environments.

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