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Letter

Multipole representation for anisotropic Coulomb interactions

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Multipole representation is proposed for the anisotropic Coulomb interactions in solids. Any local interactions can be expressed as the product of two multipole operators, and the interaction parameters are systematically classified based on the point group symmetry. The form of the multipole interactions are restricted not only by the symmetry and Hermiticity but also by the spatial structure of the interaction, which is closely related to the presence or absence of the odd-rank multipoles. As an exemplary demonstration, the screened Coulomb interaction for $\mathrm{SrVO_3}$ is considered, where only a few parameters are necessary for its description. By comparing it with the unscreened version, the totally symmetric A_1 representation is found to be strongly suppressed, but the A_1 component still gives a dominant contribution for the anisotropic part of the interaction. The anisotropic interactions are also applied to the localized two f-electron wave functions, which give the same-order contribution as the one-body level splitting estimated by the band structure calculation.

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The interactions among electrons cause a variety of intriguing phenomena in strongly correlated systems. In solids, the tight-binding model is frequently used for the description of the correlated electronic states, where the Coulomb interactions can be expressed in the second quantized Hamiltonian by the product of the four creation/annihilation operators of electrons. Although for most cases the Hubbard model with the on-site interaction is enough for the phenomena of interest, the multiorbital nature is still necessary to be considered for almost all the strongly correlated materials including d-electron systems such as iron-based superconductors, heavy-electron materials, and molecular-based conductors [1].

Usually, the Coulomb interaction is considered as the one in the spherical limit, where only a few parameters are necessary [2]. In the presence of the multiorbital effects in solids, however, a discontinuous point group symmetry at the correlated site complicates the spatial structure of the interaction. The Coulomb interaction in solids has been systematically studied for a spherical interaction with cubic crystalline field [3,4]. The more general interactions have also been considered by Bünemann and Gebhard, where the interaction parameters are classified based on point group symmetries [5]. In the present paper, we propose a simple representation in terms of multipole operator which was originally introduced for a description of the local degrees of freedom of f electrons [6–15] in terms of total angular momentum J. The multipoles have also been used for analyzing the two-particle Green functions [16], and applied to the other systems beyond the scope of f electrons [17–20]. As demonstrated in this paper, the multipole representation makes it simpler to consider the anisotropic Coulomb interactions in solids with discrete point group symmetries. There are even-rank and odd-rank multipoles as classified by the time-reversal symmetry (TRS), and we show that the spatial structure of the interaction is closely connected to the presence or absence of the odd-rank multipoles in the interaction. The proposed scheme is used for analyzing the complicated effective interactions in the first-principles calculations [21–29]. While the multipole representation for the classical Coulomb potential has been proposed [30], our scheme with a second quantization form can be used in the quantum mechanical level.

The local Coulomb interaction in solids is written in a general form as

$$\mathscr{H}_{\mathrm{C}} = \frac{1}{2}\int \mathrm{d}\boldsymbol{r}_1 \mathrm{d}\boldsymbol{r}_2 \mathrm{d}\boldsymbol{r}_3 \mathrm{d}\boldsymbol{r}_4 \times U(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_3,\boldsymbol{r}_4) : n(\boldsymbol{r}_1,\boldsymbol{r}_3)n(\boldsymbol{r}_2,\boldsymbol{r}_4) :, \qquad (1)$$

where $n(\boldsymbol{r},\boldsymbol{r}') = \sum_\sigma \psi_\sigma^\dagger(\boldsymbol{r})\psi_\sigma(\boldsymbol{r}')$ with the annihilation operator $\psi_\sigma(\boldsymbol{r})$ of electrons with spin σ $(=\uparrow,\downarrow)$. The colon (:) symbol makes the expression normal ordering, i.e., the creation operators are placed left and annihilation operators right with the consideration of anticommutation relation [3]. For specific principal and azimuthal quantum numbers (n, ℓ) of localized electrons at an atom, it is expressed as

$$\mathscr{H}_{\mathrm{C}} = \frac{1}{2}\sum_{m_1m_2m_3m_4} U_{m_1m_2m_3m_4} : n_{m_1m_3}n_{m_2m_4} :, \qquad (2)$$

where the density operator is defined by $n_{mm'} = \sum_\sigma c_{m\sigma}^\dagger c_{m'\sigma}$ with the electron annihilation operator $c_{m\sigma}$ of a magnetic quantum number m.

For a spherically symmetric case, it is well known that the above matrix element can be expressed by the Slater-Condon parameters, which are denoted as F^k where k is a rank [2].

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TABLE I. List of multipoles for each rank classified by the irreducible representations under the cubic O_h point group symmetry. See Ref. [12] for the concrete form of the multipoles.

Rank of multipoles	Type of multipoles (Γ)
$k = 0$	A_1
$k = 1$	T_1
$k = 2$	$E + T_2$
$k = 3$	$A_2 + T_1 + T_2$
$k = 4$	$A_1 + E + T_1 + T_2$
$k = 5$	$E + T_1^{\mathrm{a}} + T_1^{\mathrm{b}} + T_2$
$k = 6$	$A_1 + A_2 + E + T_1 + T_2^{\mathrm{a}} + T_2^{\mathrm{b}}$

Owing to the symmetry of the Gaunt coefficient, the odd k terms can be set as zero and only $\ell + 1$ parameters are needed ($k = 0, 2, \ldots, 2\ell$). In solids, on the other hand, the continuous symmetry does not exist, and therefore the matrix elements have much complicated structure as seen in, e.g., Ref. [28].

The central idea of this paper is to rewrite the Coulomb interaction in terms of multipole operators, which are defined by $M_\xi = \sum_{mm'\sigma} c^\dagger_{m\sigma} O^\xi_{mm'} c_{m'\sigma}$ where ξ is the index for the multipole and $\hat{O}^\xi$ is a Hermitian $(2\ell+1)\times(2\ell+1)$ matrix. These are constructed through the combination of angular momentum operators [12]. The number of the full set of matrices is also $(2\ell+1)^2$. Hence the series of the matrices is regarded as complete, and any matrix can be expanded by these matrices. More specifically, the matrices satisfy the relations $\mathrm{Tr}\, \hat{O}^\xi \hat{O}^{\xi'} = (2\ell+1)\delta_{\xi\xi'}$ and $\sum_\xi O^\xi_{m_1 m_2} O^\xi_{m_3 m_4} = (2\ell+1)\delta_{m_1 m_4}\delta_{m_2 m_3}$. These relations can be understood by considering the most simple situation with 2×2 matrices, where the three Pauli matrices and identity matrix are involved. We can transform the density operator $n_{mm'}$ into the multipole operators, and the interaction is written as

$$\mathscr{H}_\mathrm{C} = \frac{1}{2}\sum_{\xi\xi'} I(\xi,\xi') : M_\xi M_{\xi'} :, \tag{3}$$

where the relation $I(\xi,\xi') = I(\xi',\xi) = I^*(\xi,\xi')$ holds. Starting from Eq. (2), we can perform the multipole expansion uniquely with a given set of matrices. This is the most general expression for the local Coulomb interaction in the absence of the spin-orbit coupling in the interaction terms [31].

In solids, the multipole matrices are classified by the rank, which corresponds to the angular momentum of multipoles, and also by the irreducible representation under a given point group [12]. The complete set of the multipoles are explicitly given in the literature [12,20,32], and here we utilize them for description of interactions. The types of the multipoles are obtained by considering the irreducible decomposition for each rank [4,33], and we summarize the results in Table I for the cubic point group. The index for multipoles is then written as $\xi = (k, \Gamma, \alpha)$, where Γ identifies the type listed in Table I and α distinguishes the degenerate components belonging to Γ. Note that Γ is implicitly dependent on k (see Table I), and also α dependent on Γ.

Here, we explain the multipole matrices in detail for the cubic point group. Following the procedure given in Ref. [12], we construct the complete orthonormal basis for the matrices. Since the full list is shown in the literature [12,20,32], we do not list the full set of matrices but we only show a few of them. First of all, we define the angular momentum $(2\ell+1)\times(2\ell+1)$ matrices $\hat{L}_{x,y,z}$, and their combination leads to the complete set of matrices $\hat{O}^{k,\Gamma,\alpha}$. More concretely, we introduce the diagonal matrix $\hat{L}_z = \mathrm{diag}\,(\ell, \ell-1, \ldots, -\ell)$ by determining the quantization axis, and then the x and y components are constructed as they satisfy the commutation relations $[\hat{L}_\mu, \hat{L}_\nu] = \mathrm{i}\sum_\lambda \epsilon_{\mu\nu\lambda}\hat{L}_\lambda$, where $\epsilon_{\mu\nu\lambda}$ is the antisymmetric tensor. The trivial one is the rank 0 matrix

$$\hat{O}^{0,A_1} = \hat{1}, \tag{4}$$

where we have omitted the index α for the one-dimensional representations. The rank 1 matrices are equivalent to the angular momentum matrix. Noting that the trace of squared matrix is normalized to $2\ell+1$, we obtain

$$\hat{O}^{1,T_1,1} = \sqrt{\frac{3}{\ell(\ell+1)}}\hat{L}_x, \tag{5a}$$

$$\hat{O}^{1,T_1,2} = \sqrt{\frac{3}{\ell(\ell+1)}}\hat{L}_y, \tag{5b}$$

$$\hat{O}^{1,T_1,3} = \sqrt{\frac{3}{\ell(\ell+1)}}\hat{L}_z. \tag{5c}$$

The rank $k = 2$ operators are made from the combinations of $\hat{L}_{x,y,z}$. We utilize the polynomials $x^2 - y^2$, $3z^2 - r^2$ ($r^2 = x^2 + y^2 + z^2$), xy, yz, zx for the rank 2 representation, and replace them by the angular momentum matrix to obtain the rank 2 matrices. In order to make it Hermitian, we symmetrize the expression and obtain

$$\hat{O}^{2,E,1} \propto 3\hat{L}_z^2 - \hat{\boldsymbol{L}}^2, \tag{6a}$$

$$\hat{O}^{2,E,2} \propto \hat{L}_x^2 - \hat{L}_y^2, \tag{6b}$$

$$\hat{O}^{2,T_2,1} \propto \hat{L}_x\hat{L}_y + \hat{L}_y\hat{L}_x, \tag{6c}$$

$$\hat{O}^{2,T_2,2} \propto \hat{L}_y\hat{L}_z + \hat{L}_z\hat{L}_y, \tag{6d}$$

$$\hat{O}^{2,T_2,3} \propto \hat{L}_z\hat{L}_x + \hat{L}_x\hat{L}_z. \tag{6e}$$

Note that we need to normalize the expressions. Repeating the same procedure for the higher order ranks, we obtain the complete set of the $(2\ell+1)^2$ matrices.

The multipole interaction now becomes

$$\mathscr{H}_\mathrm{C} = \frac{1}{2}\sum_{k\Gamma}\sum_{k'\Gamma'} I(k,k';\Gamma,\Gamma')\sum_\alpha : M_{k\Gamma\alpha}M_{k'\Gamma'\alpha} : . \tag{7}$$

Because of selection rules in the group theory, the multipoles with different irreducible representations do not interact [34]. However, this does not mean the interaction is diagonal with respect to Γ: for example, the interaction parameter $I(2, 6; T_2, T_2^{\mathrm{a}})$ can be finite since the two multipoles belong to the same irreducible representation T_2. This property is also checked by using the concrete expressions for multipoles given in Refs. [12,20]. While we restrict ourselves to a fixed ℓ case, the extension for the parity mixing between the different orbital angular momenta is also possible with using a suitable multipole basis [32].

The above multipole representation can be identified with the familiar Slater-Condon parameters F^k in the spherical

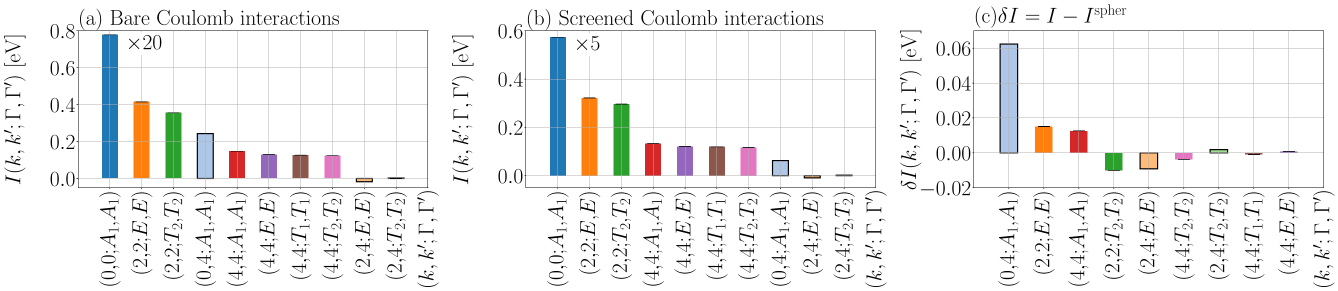

FIG. 1. Local Coulomb interaction parameters for $SrVO_3$. Multipole representation for (a) unscreened interaction and (b) screened interaction calculated by cRPA are shown. The value of $I(0,0;A_1,A_1)$ needs to be multiplied by 20 in (a) and 5 in (b). In (c), the deviation from the spherical limit $\delta I = I - I^{\mathrm{spher}}$ for the screened Coulomb interaction in (b) is plotted.

limit. In this case I is dependent only on the rank k of the multipoles, and there is a simple correspondence $I(k,k') \propto F^k \delta_{kk'}$ [34]. The Slater-Condon parametrization is frequently utilized in local density appoximation (LDA)+U and LDA+dynamical mean-field theory (DMFT) frameworks [35,36], but is not generally valid in solid state materials.

For a time-reversal symmetric system, the even- and odd-rank multipoles do not mix, since the odd-rank multipoles are odd under the time-reversal transformation. Actually, the appearance of even-rank or odd-rank multipoles are also closely related to the functional form of $U(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_3,\boldsymbol{r}_4)$. Usually, the interaction $U(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_1,\boldsymbol{r}_2)$ at two spatial points is considered. In this case, the interaction includes only the even-rank multipoles (See Supplemental Material (SM) A [34]), and hence it does not have the ability to describe the TRS breaking in interaction terms. The complexity enters when we consider the more general case $U(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_1,\boldsymbol{r}_4)$, which can be realized by considering the static three-point vertex correction to the Coulomb interaction. However, even in this case, we do not have odd-rank multipoles for the time-reversal symmetric system [34]. Thus, only the case of $U(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_3,\boldsymbol{r}_4)$ with static four-point vertex corrections induces the odd-rank multipole interactions. We note that, for a TRS broken system, the three-point function can have the odd-rank multipoles coupled to even-rank multipoles, but the two-point function does not have an ability to describe the TRS breaking. In this way, the appearance or disappearance of odd-rank multipoles is connected with the spatial structure of the interaction.

Below, we consider the three specific cases as a demonstration of multipole representation of the Coulomb interactions.

Application to p electrons. In order to have intuition for multipole interactions, let us first consider the p electrons ($\ell = 1$) with real wave function basis. The results are also applicable to the t_{2g} orbitals of d electrons [37] and t_{1u} orbitals of fulleride materials [38]. The interaction is usually parametrized as the Slater-Kanamori interaction [39–41]: $U_{\gamma\gamma\gamma\gamma} = U$, $U_{\gamma\gamma'\gamma\gamma'} = U'$, $U_{\gamma\gamma'\gamma'\gamma} = U_{\gamma\gamma\gamma'\gamma'} = J$ $(\gamma \neq \gamma')$ where γ represents $p_{x,y,z}$ orbitals instead of the magnetic angular momentum m $(= 0, \pm 1)$. We note that this Kanamori parametrization is not valid in the case with full d or f orbitals. The relations to the multipole representation are identified as $I(0,0;A_1,A_1) = \frac{1}{3}(U + 2U')$, $I(2,2;E,E) = \frac{1}{3}(U - U')$, $I(2,2;T_2,T_2) = \frac{2}{3}J$ and $I(1,1;T_1,T_1) = 0$. The cubic symmetry is reflected in the difference between $I(2,2;E,E)$ and $I(2,2;T_2,T_2)$, and in the spherically symmetric case we have the relation $I(2,2;E,E) = I(2,2;T_2,T_2)$ identical to the well-known condition $U' = U - 2J$. We emphasize that only in this spherical case can the Slater-Condon parametrization ($F^{0,2}$) be used. The odd-rank multipole with $\Gamma = T_1$ is absent in this case, which is related to the fact that the Slater-Kanamori parametrization is based on the interactions at two spatial points.

Application to d electrons. Next we consider the actual materials. For an exemplary demonstration, we take the concrete material $SrVO_3$. We calculate the electronic band structure of $SrVO_3$ from the LDA of the DFT [42,43] and construct the maximally localized Wannier function of the V $3d$ orbitals hybridized with the O $2p$ orbitals. We also perform the constrained random phase approximation (cRPA) technique to calculate the screened interaction for the Wannier functions, which reflects the screening effects in the cubic crystal [44,45]. The detail of the *ab initio* calculation is shown in SM B [34].

Figure 1 shows the multipole interactions for $3d$ electrons at the V site with O_h point group symmetry, where the values are listed in descending order with respect to the absolute values. We emphasize that only a few interaction parameters are needed to express the full interaction term in solids, even though the original Coulomb tensor has, naively, $(2\ell + 1)^4 = 625$ components. Since the numerical errors are in general included in the raw data [34], we symmetrize the interactions, but the error bars are invisible for our data. The bare Coulomb interactions are shown in (a). We can see, for instance, the difference between $I(k,k;E,E)$ and $I(k,k;T_2,T_2)$ which reflects the cubic symmetry. The screened interactions are also shown in Fig. 1(b), where the A_1 components are much suppressed, while the others remain almost unchanged. This is understood as follows: The electronic charge is responsible for the screening to reduce the Coulomb interaction, and the charge component is represented as totally symmetric representation A_1. Because of the cubic symmetry, the different ranks for A_1 are mixed and are much influenced by screening. Intuitively, the parameter $I(0,4;A_1,A_1)$ may be interpreted as that the spherical deformation of charge is accompanied by the cubic deformation in the solids with O_h symmetry.

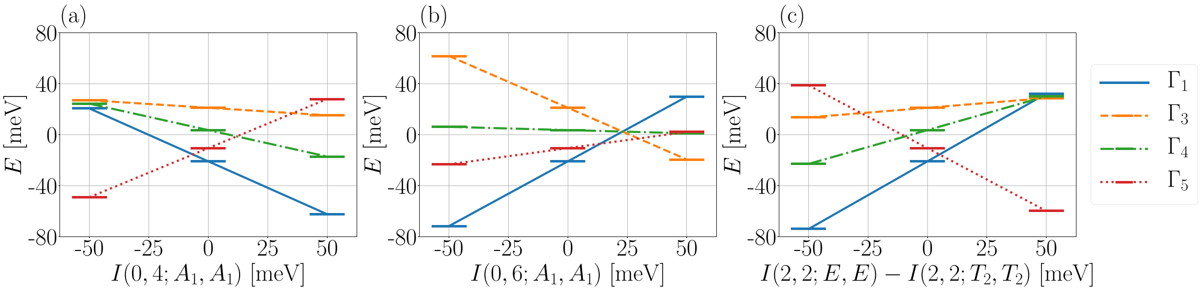

FIG. 2. Anisotropic interaction effects on the crystal field energy levels of f^2 wave function. The types of interaction parameters are (a) $I(0,4;A_1,A_1)$, (b) $I(0,6;A_1,A_1)$, and (c) $I(2,2;E,E)-I(2,2;T_2,T_2)$. The level scheme of the local one-body potential calculated for UBe$_{13}$ is shown at $I=0$ and the deviation enters for $I\neq 0$.

We note that the odd-rank multipoles are absent in the cRPA results. This is consistent with the fact that the RPA calculation is performed for a fixed single wave vector $\boldsymbol{q}$ of the effective interaction where no three-point vertex correction is considered. The effective interaction then originates from a function of the two spatial points and the odd-rank multipoles do not appear as discussed before. Since the spatial inversion does not change the local interaction term for a fixed ℓ [34], the absence of the odd-rank multipoles indicates that the local cRPA interaction is always time-reversal and inversion symmetric.

The result of the screened Coulomb interactions shown in Fig. 1(b) can be represented as the sum of the spherically symmetric part $I^{\rm spher}$ plus its deviation, which is suitable for examining the contribution from the discrete cubic symmetry. The spherical part of the interaction is extracted as $I^{\rm spher}(k)=\frac{1}{2k+1}\sum_\Gamma d_\Gamma I(k,k;\Gamma,\Gamma)$ where $d_\Gamma=\sum_\alpha 1$ is the number of degeneracy. This is used for the definition of the cubic deviation $\delta I=I-I^{\rm spher}$ shown in Fig. 1(c). Here the dominant component is $I(0,4;A_1,A_1)$ as compared to the others, even though it has been much reduced by the screening effect. The second largest one is the rank-2 with non-A_1 representation, and the other interaction values are basically decreased as the rank increases. The present results thus indicate that the dominant contributions for the anisotropic part are given by the A_1 component plus rank-2 component.

Whereas the data in Fig. 1 are sufficiently accurate, the more complex materials may produce the larger numerical errors. The quality of data can be improved by using the symmetries, and if the interaction originates from spatial two-point functions as in cRPA, the odd-rank multipoles can also be dropped, which is not a requirement of symmetry.

In this way, the screening behavior in solids becomes clearer with multipole representation. It is interesting to explore the interaction parameter structures in a wide range of materials with cRPA and the more accurate technique beyond cRPA.

Application to f electrons. We further apply the anisotropic multipole interactions to the localized f-electron wave function under the cubic crystalline field [52]. We consider the two f-electron (f^2) wave functions realized in Pr- and U-based materials, for which the interaction effects are relevant. By considering the spherical part of the Coulomb interaction and the spin-orbital coupling (Hund's rule), we obtain the ground state $J=4$ multiplet $|M\rangle$ ($M\in[-J,J]$) (SM C [34]). We now demonstrate that the anisotropic interaction affects the wave function and modifies the energy level structure. We take UBe$_{13}$ as an example, which shows the robust non-Fermi-liquid behavior and unconventional superconductivity, and the multichannel Kondo effects have been suspected as their possible origins [46,47]. In the multichannel Kondo effect scenario, the realization of non-Kramers Γ_3 doublet ground state is a necessary condition for a robust non-Fermi liquid. In our previous works, we focus on the fact that the conduction electrons of UBe$_{13}$ can be seen as compensated metal and propose possible scenario for the unconventional superconductivity [48,49]. On the other hand, there is another possibility that the competition between Kondo singlet and crystalline field Γ_1 singlet leads also to the non-Fermi liquids [50,51]. With these backgrounds, we consider the effect of the anisotropic multipole interaction on the f^2 wave functions and see what kind of the ground state is favored by multipole interactions.

For the estimation of the local one-body level splitting, we calculate the band structures of UBe$_{13}$ and find the local on-site potentials for f electrons [34]. Since Fig. 1 shows that the dominant contribution is the A_1 type involving the rank 0 component, we consider the multipole interactions $I(0,4;A_1,A_1)$, $I(0,6;A_1,A_1)$, and also the second largest one $I(2,2;E,E)-I(2,2;T_2,T_2)$ in Fig. 1(c). The magnitude of the typical values are estimated as $I(0,4;A_1,A_1)/I(0,0;A_1,A_1)\simeq 0.02$ from Figs. 1(b) and 1(c). Assuming that $I(0,0;A_1,A_1)$ $(=F^0)$ is nearly 2.5 eV for a U atom [53], we consider the range $|I|<50$ meV. The results are shown in Fig. 2, where the anisotropic interactions are included by the first-order perturbation theory [34]. The crystalline field Γ_1 singlet is the ground state without multipole interactions, and once the anisotropy is introduced, we find Γ_1, Γ_3, and Γ_5 ground states depending on the parameters. Thus the cubic anisotropy of the interaction can substantially modify the crystal field structure determined by the non-correlated parts.

This scheme is applicable to any types of material, and hence our results show that we need to be careful about the anisotropic interaction effects when one determines the multiple f-electron wave functions from a microscopic point of view [54]. For the one-body part of the Hamiltonian, the energy spectrum is expected to be accurately described by the first-principles band structure calculations, but the interaction effect considered in this paper is a correlation effect which is not included in the band-structure calculations. We note that this is not true for the f^1 in Ce and its hole analog, f^{13} in Yb, since the interaction effects for the localized crystal field levels are irrelevant.

To summarize, we have proposed a systematic and simple way to express the Coulomb interaction in solids by using the multipole operators. The interaction parameters are restricted by the symmetries, and we have also identified that the spatial structure of the interaction functional form is closely connected to the presence or absence of odd-rank multipoles. The multipole representation can be utilized for examining the structure of the complicated cRPA interactions and for studying the crystal field ground states of the localized correlated electrons. Whereas we focus on the cubic crystal in the present paper, in principle, the formulation with multipoles can be applied to any local interactions including molecules and quasicrystals, and can also be generalized for the intersite interactions. The scheme can be further utilized to compactify the two-particle Green's functions in multiorbital systems, which shares a structure similar to the Coulomb interaction term.

Moreover, in recent years, the tight-binding models based on the first-principles calculation have been intensively used not only for the strongly correlated electrons but also for the topological systems. The effect of interaction is necessary for a quantitative description of electronic properties, where one must consider the complicated Coulomb interaction tensor in solids. Our technique is generic and useful in accessing the interaction parameters with discrete symmetries.

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[1] For a review, see, *Superconductivity*, edited by K.-H. Bennemann and J. B. Ketterson (Springer, Berlin, 2004).

[2] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1951).

[3] M. E. A. Coury, S. L. Dudarev, W. M. C. Foulkes, A. P. Horsfield, P.-W. Ma, and J. S. Spencer, Phys. Rev. B **93**, 075101 (2016).

[4] S. Sugano, Y. Tanabe, and H. Kamimura, *Multiplets of Transition-Metal Ions in Crystals* (Academic Press, New York, 1970).

[5] J. Bünemann and F. Gebhard, J. Phys.: Condens. Matter **29**, 165601 (2017).

[6] F. J. Ohkawa, J. Phys. Soc. Jpn. **52**, 3897 (1983).

[7] R. Shiina, H. Shiba, and P. Thalmeier, J. Phys. Soc. Jpn. **66**, 1741 (1997).

[8] Y. Kuramoto and H. Kusunose, J. Phys. Soc. Jpn. **69**, 671 (2000).

[9] P. Santini and G. Amoretti, Phys. Rev. Lett. **85**, 2188 (2000).

[10] A. Kiss and Y. Kuramoto, J. Phys. Soc. Jpn. **74**, 2530 (2005).

[11] T. Takimoto, J. Phys. Soc. Jpn. **75**, 034714 (2005).

[12] H. Kusunose, J. Phys. Soc. Jpn. **77**, 064710 (2008).

[13] For a review, see, Y. Kuramoto, H. Kusunose, and A. Kiss, J. Phys. Soc. Jpn. **78**, 072001 (2009).

[14] K. Haule and G. Kotliar, Nat. Phys. **5**, 796 (2009).

[15] H. Ikeda, M.-T. Suzuki, R. Arita, T. Takimoto, T. Shibauchi, and Y. Matsuda, Nat. Phys. **8**, 528 (2012).

[16] R. Tazai and H. Kontani, Phys. Rev. B **100**, 241103(R) (2019).

[17] K. I. Kugel and D. I. Khomskii, Sov. Phys. JETP Lett. **15**, 446 (1972).

[18] G. Khaliullin, Phys. Rev. Lett. **111**, 197201 (2013).

[19] M.-T. Suzuki, T. Koretsune, M. Ochi, and R. Arita, Phys. Rev. B **95**, 094406 (2017).

[20] S. Hayami, M. Yatsushiro, Y. Yanagi, and H. Kusunose, Phys. Rev. B **98**, 165110 (2018).

[21] For a review, see, M. Imada and T. Miyake, J. Phys. Soc. Jpn. **79**, 112001 (2010).

[22] T. Miyake, K. Nakamura, R. Arita, and M. Imada, J. Phys. Soc. Jpn. **79**, 044705 (2010).

[23] T. Misawa, K. Nakamura, and M. Imada, J. Phys. Soc. Jpn. **80**, 023704 (2011).

[24] T. Misawa, K. Nakamura, and M. Imada, Phys. Rev. Lett. **108**, 177007 (2012).

[25] T. Misawa and M. Imada, Nat. Commun. **5**, 5738 (2014).

[26] M. Hirayama, T. Misawa, T. Miyake, and M. Imada, J. Phys. Soc. Jpn. **84**, 093703 (2015).

[27] M. Hirayama, Y. Yamaji, T. Misawa, and M. Imada, Phys. Rev. B **98**, 134501 (2018).

[28] M. Hirayama, T. Misawa, T. Ohgoe, Y. Yamaji, and M. Imada, Phys. Rev. B **99**, 245155 (2019).

[29] T. Ohgoe, M. Hirayama, T. Misawa, K. Ido, Y. Yamaji, and M. Imada, Phys. Rev. B **101**, 045124 (2020).

[30] A. C. Simmonett, F. C. Pickard IV, and H. F. Schaefer III, J. Chem. Phys. **140**, 184101 (2014).

[31] The operator expansion is in principle possible also for the spin-orbital coupled interactions. See Supplemental Material D [34].

[32] H. Kusunose, R. Oiwa, and S. Hayami, J. Phys. Soc. Jpn. **89**, 104704 (2020).

[33] See, for example, M. S. Dresselhaus, G. Dresselhaus, and A. Jario, *Group Theory* (Springer, Berlin, 2008).

[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.104.L081108 for details on multipole representation, band-structure calculation and f-electron wave function, which includes Refs. [4,20,32,33,42,55–57].

[35] M. T. Czyzyk and G. A. Sawatzky, Phys. Rev. B **49**, 14211 (1994).

[36] M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, Phys. Rev. B **80**, 085101 (2009).

[37] For a review, see, A. Georges, L. de Medici, and J. Mravlje, Annu. Rev. Condens. Matter Phys. **4**, 137 (2013).

[38] For a review, see, Y. Nomura, S. Sakai, M. Capone, and R. Arita, J. Phys.: Condens. Matter **28**, 153001 (2016).

[39] J. C. Slater, *Quantum Theory of Atomic Structure* (McGraw-Hill, New York, 1960).

[40] J. Kanamori, Prog. Theor. Phys. **30**, 275 (1963).

[41] C. Castellani, C. R. Natoli, and J. Ranninger, Phys. Rev. B **18**, 4945 (1978).

[42] M. Methfessel, M. van Schilfgaarde, and R. A. Casali, in *Lecture Notes in Physics*, edited by H. Dreysse (Springer-Verlag, Berlin, 2000), Vol. 535.

[43] D. M. Ceperley and B. J. Alder, Phys. Rev. Lett. **45**, 566 (1980).

[44] F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S. Biermann, and A. I. Lichtenstein, Phys. Rev. B **70**, 195104 (2004).

[45] M. Hirayama, T. Miyake, and M. Imada, Phys. Rev. B **87**, 195144 (2013).

[46] D. L. Cox, Phys. Rev. Lett. **59**, 1240 (1987).

[47] For a review, see, D. L. Cox and A. Zawadowski, Adv. Phys. **47**, 599 (1998).

[48] S. Iimura, M. Hirayama, and S. Hoshino, Phys. Rev. B **100**, 094532 (2019).

[49] S. Iimura, M. Hirayama, and S. Hoshino, Phys. Rev. B **102**, 064505 (2020).

[50] S. Yotsuhashi, K. Miyake, and H. Kusunose, J. Phys. Soc. Jpn. **71**, 389 (2005).

[51] S. Nishiyama, H. Matsuura, and K. Miyake, J. Phys. Soc. Jpn. **79**, 104711 (2010).

[52] K. R. Lea, M. J. M. Leask, and W. P. Wolf, J. Phys. Chem. Solids **23**, 1381 (1962).

[53] D. van der Marel and G. A. Sawatzky, Phys. Rev. B **37**, 10674 (1988).

[54] H. Takahashi and T. Kasuya, J. Phys. C **18**, 2697 (1985).

[55] G. Kresse and J. Furthmüller, Phys. Rev. B **54**, 11169 (1996).

[56] Y. Wang, H. Weng, L. Fu, and X. Dai, Phys. Rev. Lett. **119**, 187203 (2017).

[57] S.-T. Tamura, S. Iimura, and S. Hoshino, Phys. Rev. B **102**, 024505 (2020).