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Enhancement of spin-orbit interaction by competition between Hund’s coupling and electron hopping

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Abstract. We study the two-site system of three \( t_{2g} \) orbitals and examine the possibility to effectively enhance the spin-orbit interaction (SOI) by electron correlation. The ground state of this two-site system is calculated by exactly diagonalizing the Hamiltonian, and the effective on-site and inter-atomic SOI's are calculated. Changing the number of electrons in the system, we found the conditions for the enhancement of the effective SOI's. The conditions are the intermediate Hund’s coupling and the filling of four or five electrons.

1. Introduction
The spin-orbit interaction (SOI) is an emergence of relativistic effect in solids, which couples the spin and orbital degrees of freedom. It causes novel effects such as the anomalous Hall effect [1], spin Hall effect [2], and topological insulators [3, 4]. In addition, the SOI in materials is intensively studied for applications in the field of spintronics. To realize these novel effects, relatively strong SOI is necessary. For example, in compounds consisting of 5\( d \) elements, the magnitudes of the SOI and the Hubbard \( U \) are comparable, both of which are of the order of 0.5 eV. However the main players at the moment are rare heavy elements like Bi, Hg, and Pt, and thus it would be an obstacle for applications. Therefore, it is desirable that the relatively strong SOI is realized with lighter and abundant elements. When we focus on \( d \) electron systems, the SOI is weaker when we go up in the periodic table, from 5\( d \) to 4\( d \) and 3\( d \), while electron correlation is stronger [5]. We utilize this fact to reach the goal of the effectively enhanced SOI by the competition between electron correlation and electron hopping.

For localized electrons, the Hund’s rule [6] determines the ground state multiplet, which is described by the Russell-Saunders term symbol. In solids, strong electron correlation localizes electrons, and the local picture with the Hund’s rule well describes the ground state. In the opposite limit where electrons are itinerant, extended Bloch waves are the good description for electronic states. Electron correlation could be taken into account by the perturbation theory.

As a first step toward many-body problems, we have considered a two-site model of \( t_{2g} \) orbitals including electron correlation, electron hopping, and the SOI, and examined how the Hund’s rule is modified in the presence of electron hopping [7]. The effective intra- and inter-atomic SOI’s are calculated by exactly solving the eigenstates numerically. There we find the situation in which the SOI is effectively enhanced by the Hund’s coupling \( J \) in the intermediate spin case for the filling of 2 or 2.5 electrons per atom.
2. Model

We consider a two-site model of $t_{2g}$ orbitals where the Hamiltonian is described in the basis of atomic orbitals $|\psi_{ima}\rangle$ with $i$ being an atomic site, $m$ an orbital of an atom, and $\sigma$ a spin of an electron. The Hamiltonian is written as [8]

$$
\hat{H} = \sum_{i,j=1,2} \hat{H}_{ij}^{(t)} + \sum_{i=1,2} \hat{H}_{i}^{(\text{correlation})} + \sum_{i=1,2} \hat{H}_{i}^{(\text{SO})},
$$

where

$$
\hat{H}_{ij}^{(t)} = \sum_{\alpha \in \{\sigma, \sigma'\}} \sum_{m'm'} t_{im,jm'} d_{im\sigma}^{\dagger} d_{jm'\sigma},
$$

$$
\hat{H}_{i}^{(\text{correlation})} = \frac{1}{2}(U - 3J)\hat{N}_{i}(\hat{N}_{i} - 1) - 2J\hat{S}_{i}^{2} - \frac{3}{2}J\hat{L}_{i}^{2} + \frac{5}{2}J\hat{N}_{i},
$$

$$
\hat{H}_{i}^{(\text{SO})} = \zeta \sum_{\alpha \in \{\sigma, \sigma'\}} \sum_{m'm'} \sum_{\sigma\sigma'} d_{im\sigma}(l_{a})_{mm'}(s_{\alpha})_{\sigma\sigma'}d_{im'\sigma'},
$$

Here $d$ ($d^{\dagger}$) is the electron annihilation (creation) operator. The matrix elements of the transfer matrix $t_{im,jm'}$ are given following the Slater-Koster tables [9]. The number operator $\hat{N}_{i}$, and the orbital and angular momentum operators $\hat{L}_{i}$, $\hat{S}_{i}$ are defined as follows: $\hat{N}_{i} = \sum_{\sigma} d_{im\sigma}^{\dagger} d_{im\sigma}$, $\hat{L}_{ia} = \sum_{\sigma\sigma'} \sum_{m'm'} d_{im\sigma}(l_{a})_{mm'}d_{im'\sigma}$, and $\hat{S}_{i\alpha} = \sum_{\sigma\sigma'} \sum_{m'm'} d_{im\sigma}(s_{\alpha})_{\sigma\sigma'}d_{im'\sigma}$, where the matrix elements are given by $(l_{a})_{mm'} = i\epsilon_{amm'}$ and $(s_{\alpha})_{\sigma\sigma'} = (\sigma_{\alpha})_{\sigma\sigma'}/2$ with $\sigma_{\alpha}$ ($\alpha = x, y, z$) being a Pauli matrix. (We set $h = 1$ throughout this paper.) We note that $d_{gyz}$, $d_{zxy}$, and $d_{xyz}$ orbitals correspond to $m = x, y,$ and $z$, respectively. The relation $U = U' + 2J$ is assumed here to ensure the spin and orbital rotational symmetry. $\zeta$ determines the strength of the SOI.

The effective on-site SOI $\bar{\lambda}_{i}$ and the inter-atomic SOI $\bar{\tau}_{SOmm'}$ are defined as follows:

$$
\bar{\lambda}_{i} = - \langle \psi_{0} | \sum_{\alpha \in \{\sigma, \sigma'\}} \sum_{mm'} \sum_{\sigma\sigma'} d_{im\sigma}(l_{a})_{mm'}(s_{\alpha})_{\sigma\sigma'}d_{im'\sigma'} | \psi_{0} \rangle,
$$

$$
\bar{\tau}_{SOmm'} = \frac{1}{2} \langle \psi_{0} | \sum_{\sigma\sigma'} d_{im\sigma}(l_{a})_{mm'}(s_{\alpha})_{\sigma\sigma'}d_{2m'\sigma'} + \text{H.c.} | \psi_{0} \rangle.
$$

$|\psi_{0}\rangle$ is the ground state, and if the ground states are degenerate, an average will be taken over all the degenerate ground states with equal weights. We note that the effective inter-atomic SOI $\bar{\tau}_{SOmm'}$ is nonzero only when the indices $\alpha, m, m'$ are aligned in cyclic order of $x, y, z$ because of $(l_{a})_{mm'} = i\epsilon_{amm'}$ for $t_{2g}$ orbitals.

We align the two sites along the $z$ axis. Electron hopping between the two sites forms molecular orbitals, and its magnitude varies with the form of bonding. We set $t_{yz,yz} = t_{xz,xz} = t$ for the $\pi$ bonds and $t_{xy,xy} = -t'$ for the $\delta$ bond. Since the overlap of the wave function is larger for the $\pi$ bonds than the $\delta$ bond, we choose $t' = 0.5t$, and the SOI $\zeta = 0.1t$ in the following analysis. The model is illustrated in Fig. 1.

This two-atom model accommodates electrons ranging from zero to twelve. Since the $n$-electron system ($n \leq 6$) is complement to the $(12 - n)$-electron system, we consider only the cases of six or less electrons. We show below the effective SOI’s by changing the strength of electron correlation and the number of electrons in the system. The results are obtained by exactly solving the eigenstates numerically.

3. Results

3.1. One electron

The effective SOI’s are independent of electron correlation: $\bar{\lambda} = 0.25$, $\bar{\tau}_{SOyz,xx} = 0.125$, and $\bar{\tau}_{SOzx,xy} = \bar{\tau}_{SOxy,yz} = 0$. 

2
3.2. Two and three electrons

For the cases of electrons ranging from two to six, the results are shown in Fig. 2. We change $J/t$ and $U'/t$ as free parameters with the condition $U = U' + 2J$. Note that the on-site Hubbard interaction $U$ is reduced by the Hund’s coupling $J$ to be $U_{\text{eff}} = U - 3J = U' - J$ [10]. The effective Hubbard interaction $U_{\text{eff}}$ becomes negative for $J > U'$, and thus this parameter region is unphysical.

The ground state of the system can be understood by focusing on the total spin $S$ of the two sites, although it is not a good quantum number in the presence of SOI [7]. For the two-electron system, the total spin can take either 0 or 1, which corresponds to low-spin or high-spin state, respectively. In the absence of SOI, the ground states at $J = 0$ are degenerate consisting of three $S = 0$ states and three $S = 1$ states, and infinitesimal Hund’s coupling $J$ lifts this degeneracy by lowering the energy of $S = 1$ states. If the SOI is turned on, the ground states gradually change with its width characterized by the strength of the SOI $\zeta$. Even in the presence of SOI, the ground state near $J = 0$ is approximated by a linear combination of $S = 0$ and $S = 1$ states (denoted L+H in Fig. 2), and it crosses over to the high-spin state (H in Fig. 2).

For the two-electron system, $\lambda$ and $\tilde{t}_{SOyz,xx}$ become smaller by electron correlation, whereas $\tilde{t}_{SOxx,xy} = t_{SOxy,yz}$ is very small. $\tilde{t}_{SOxx,xy}$ has tiny values because the $d_{xy}$ orbitals are almost empty since two electrons mainly form $\pi$-bonding orbitals.

For the three-electron system, there is discontinuity in the expectation values which separates the low-spin and high-spin states. The discontinuity is mainly dependent on $J/t$, and not on $U'/t$. It is because the Hund’s rule results from the Hund’s coupling $J$, not from $U$ or $U'$ [see Eq. (3)].

The ground state of the three-electron system can be understood basically in the same way as the two-electron case. Namely, there are two kinds of the ground states: the low-spin ($S = 1/2$) and high-spin ($S = 3/2$) states. In the low-spin state, the total spin has a small value and electrons are mobile between the two sites. On the other hand, the total spin has a large value and electrons are localized on a site in the high-spin state. The largest effective SOI’s are realized in the low-spin region, and electron correlation makes the effective SOI’s smaller. The effect of the SOI is to make spin and orbital degrees rotate. However, electron correlation aligns electron spins and quenches orbital degrees, which results in the small value of the effective SOI’s.
**Figure 2.** Results of the exact diagonalization. The number of electrons in the system varies from two to six. The effective on-site SOI $\lambda$ and inter-atomic SOI $\tilde{t}_{SO}^{x,z,y}$, $\tilde{t}_{SO}^{x,x,y}$, $\tilde{t}_{SO}^{y,y,z}$ are shown for each case. The ground state of the system is indicated in the figure: L means the low-spin state, I the intermediate-spin state, and H the high-spin state. Discontinuous change of the total spin is depicted by the solid lines. L+H in the results of two electrons means that the ground state is a linear combination of low-spin ($S=0$) and high-spin ($S=1$) states, and it is not dictated by a single spin value. Its width in the direction of $J/t$ is characterized by the SOI $\zeta/t$. The high-spin state in the four-electron case is further separated into three parts (Ha, Hb, and Hc) in the presence of SOI. In the shaded regions for $J > U'$, the effective Hubbard interaction $U_{eff}$ becomes negative, which is unphysical.
3.3. Four and five electrons
For the cases of four and five electrons, we can observe the enhancement of the effective SOI’s by electron correlation; i.e., the effective SOI’s become larger than those in the absence of electron correlation. For the four electron system, the effective on-site SOI $\lambda$ can be about six times larger than that of the noninteracting case, and for the five electron system, it can be about four times larger.

Flipping a spin changes the total spin by one, and thus three types of ground states are realized in these systems: the low-spin, intermediate-spin, and high-spin states. For the four-electron system, the total spin is either $S = 0$, $S = 1$, or $S = 2$, while $S = 1/2$, $S = 3/2$, or $S = 5/2$ for the five-electron system. Realization of the intermediate-spin state is a key for the large effective SOI’s. To make the effective SOI’s larger, the degrees of freedom should be large for both spin and orbital angular momenta. In the low-spin region, the spin has a small value whereas the orbital has a large value. On the other hand, the spin is large and the orbital is small in the high-spin region. Both cases are not fitted for the SOI to play a major role since either spin or orbital degrees is quenched. However, in the intermediate-spin state, both the spin and orbital angular momenta have moderate values, which is an ideal stage for the enhancement of the effective SOI’s.

We also found a complex behavior in the four-electron system. The intermediate-spin state vanishes by increasing $U''/t$, and the SOI splits the high-spin state into three types (Ha, Hb, Hc in the Fig. 2). This behavior cannot be seen in the systems with different number of electrons. It might be related to the bad metallic behavior in the three-orbital model with two electrons per atom [11]. For the five-electron system, the ground state can again be explained by focusing on $J/t$. The low-spin and intermediate-spin states are smoothly connected in the five-electron system since the SOI smears the discontinuity between the two states. If the SOI is switched off, we can observe the discontinuity that separates the low-spin and intermediate-spin states.

3.4. Six electrons
Six electrons in this system corresponds to half-filling. The result of the half-filled system is completely different from those for the systems with other electron numbers. The total spin $S$ remains $S = 0$ even in the strong coupling region, whereas it is maximized in other cases. This is reasonable since the half-filled systems is described by the quantum antiferromagnetic Heisenberg model in the strong coupling limit. Therefore, the total spin is small regardless of electron correlation and the spins of the two site have antiferromagnetic correlation [7].

The effective SOI’s are not enhanced in the six-electron system, although there is a large difference in magnitudes of the on-site SOI $\lambda$ and the inter-atomic SOI $t_{SO}$. It results from the difference between the total spin and the local spin. The on-site SOI reflects the local spin, i.e., the spin of a site. On the other hand, the inter-atomic SOI is related to the total spin of the two sites. Electron correlation polarizes the spin on a site, but the antiferromagnetic spin correlation makes the total spin of the two sites small. Therefore the large local spin keeps the effective on-site SOI large, and the small total spin leads to the small effective inter-atomic SOI.

4. Discussion
We have considered the two-site model of $t_{2g}$ orbitals and have sought for the possibility of the enhanced effective SOI’s by electron correlation. The enhanced effective SOI’s are found for the cases of four and five electrons, i.e., 2 or 2.5 electrons per atom, where the intermediate-spin states are realized for the moderate electron correlation. Both the on-site and inter-atomic SOI’s become larger than those in the absence of electron correlation. In the intermediate spin states, both the spin and orbital angular momenta are not polarized and have moderate values. It indicates that the fluctuating or frustrating spin state is preferred for the effective SOI’s to be enhanced. In other cases, the ground state is either the low or high spin state,
and electron correlation makes the effective SOI’s smaller. Possible candidate materials for the scenario should have $t_{2g}^{2-2.5}$ configurations. Here we consider perovskite and layered-perovskite transition metal oxides as possible candidates, where $d$ orbitals are split into $t_{2g}$ and $e_g$ orbitals. LaVO$_3$ is a Mott insulator [12–15], where V$^{3+}$ has $t_{2g}^{2}$ configuration. Other candidate materials are La$_x$Sr$_{1-x}$CrO$_3$ ($0 \leq x \leq 0.5$) [16–18] with $t_{2g}^{2-2.5}$ configuration, LaSrVO$_4$ [19] with $t_{2g}$ configuration, and (La$_x$Sr$_{1-x}$)$_2$CrO$_4$ ($0 \leq x \leq 0.5$) [18] with $t_{2g}^{2-2.5}$ configuration. Our findings would be a useful guideline to find a material that holds the effectively strong SOI without using rare heavy elements.

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**References**