Spin-orbital state induced by strong spin-orbit coupling

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Abstract.
To clarify a crucial role of a spin-orbit coupling in the emergence of novel spin-orbital states in 5d-electron compounds such as Sr$_2$IrO$_4$, we investigate ground state properties of a $t_{2g}$-orbital Hubbard model on a square lattice by Lanczos diagonalization. In the absence of the spin-orbit coupling, the ground state is spin singlet. When the spin-orbit coupling is strong enough, the ground state turns into a weak ferromagnetic state. The weak ferromagnetic state is a singlet state in terms of an effective total angular momentum. Regarding the orbital state, we find the so-called complex orbital state, in which real $xy$, $yz$, and $zx$ orbital states are mixed with complex coefficients.

1. Introduction
Novel quantum phenomenon driven by the interplay of strong spin-orbit coupling and Coulomb interaction has been a subject of significant study in the fields of condensed matter physics and materials science. Since spin and orbital degrees of freedom are entangled at a local level by the spin-orbit coupling, the total angular momentum provides a good description of ordered phases and fluctuation properties. On the other hand, we would envisage possible realization of devices with exotic functionalities that utilize cross correlation involving magnetic and electric degrees of freedom, in which magnetic behavior can be manipulated by an applied electric field, while electric behavior can be controlled by a magnetic field.

Recently, a 5d transition metal oxide Sr$_2$IrO$_4$, in which Ir$^{4+}$ ions have five electrons with a low-spin ($t_{2g}$)$^5$ state, has attracted growing attention as a candidate for a novel Mott insulator characterized by an effective total angular momentum $J_{\text{eff}} = \frac{1}{2}$, where $J_{\text{eff}} = -L + S$, rather than spin commonly observed in 3d Mott insulators [1, 2]. The spin-orbit coupling in the $t_{2g}$ manifold stabilizes the $J_{\text{eff}} = \frac{1}{2}$ state. The emergent behavior of the $J_{\text{eff}} = \frac{1}{2}$ Mott insulator has been explored in experiments such as angle resolved photoemission spectroscopy [1], optical conductivity [1, 3], resonant x-ray scattering [2], and resonant inelastic x-ray scattering [4]. An isostructural iridate Ba$_2$IrO$_4$ has also been reported as the $J_{\text{eff}} = \frac{1}{2}$ Mott insulator [5]. Theoretical efforts have been devoted to clarify the characteristics of the $J_{\text{eff}} = \frac{1}{2}$ Mott insulator on the basis of first-principles calculations [1, 6] and model calculations [7, 8, 9, 10].

The purpose of this paper is to gain an insight into the spin-orbital state under the spin-orbit coupling from a microscopic viewpoint. We investigate ground-state properties of a $t_{2g}$-orbital Hubbard model including the spin-orbit coupling by exploiting numerical techniques. We discuss a spin-orbit-induced ground-state transition from a spin singlet state to a singlet state in terms of the effective total angular momentum.
2. Model and numerical method

We consider triply degenerate $t_{2g}$ orbitals on a square lattice in the $xy$ plane, and the electron number per site is five, corresponding to the low-spin state of Ir$^{4+}$ ions. The $t_{2g}$-orbital Hubbard model is given by

$$H = \sum_{i,\sigma} t_{\sigma}^{\text{in}} (d_{i,\sigma}^\dagger d_{i+1,\sigma} + \text{h.c.}) + \lambda \sum_{i} \mathbf{S}_i \cdot \mathbf{S}_i + U \sum_{i,\sigma} \rho_{\uparrow\sigma} \rho_{\downarrow\sigma} + \frac{U'}{2} \sum_{i,\sigma,\sigma',\tau} \rho_{\tau\sigma} \rho_{\tau\sigma'}$$

$$+ \frac{J}{2} \sum_{i,\sigma,\sigma',\tau \neq \tau'} d_{i,\sigma}^\dagger d_{i,\sigma'}^\dagger d_{i+1,\sigma} d_{i+1,\sigma'},$$

(1)

where $d_{i,\sigma}$ is an annihilation operator for an electron with spin $\sigma (=\uparrow, \downarrow)$ in orbital $\tau (=xy, yz, zx)$ at site $i$, $\rho_{\tau\sigma} = d_{i,\sigma}^\dagger d_{i,\tau\sigma}$, and $\mathbf{S}_i$ and $\mathbf{L}_i$ represent spin and orbital angular momentum operators, respectively. The hopping amplitude is given by $t_{\sigma\tau}^{xy} = t_{xy}$, $t_{\sigma\tau}^{yz} = t_{yz}$, and $t_{\sigma\tau}^{zx} = t$, and zero for other combinations of orbitals. Hereafter, $t$ is taken as the energy unit. $\lambda$ denotes the spin-orbit coupling, $U$, $U'$, $J$, and $J'$ are intra-orbital Coulomb, inter-orbital Coulomb, exchange (Hund’s rule coupling), and pair-hopping interactions, respectively. We assume that $U=U'+J+J'$ due to the rotational symmetry in the local orbital space, and $J=J'$ due to the reality of the $xy$, $yz$, and $zx$ orbital functions.

We numerically investigate ground-state properties of the $2 \times 2$ four-site system by Lanczos diagonalization. Because of the three orbitals, the number of bases per site is $4^3=64$, and the matrix dimension of the Hamiltonian becomes huge as the system size increases. In general, the matrix dimension is reduced by decomposing the Hilbert space into a block-diagonal form by using symmetries of the Hamiltonian. In the present case, the total number of electrons is a good quantum number. We cannot use the $z$ component of the total spin as a good quantum number, since the spin SU(2) symmetry is broken by the spin-orbit coupling. We have not utilized the lattice symmetry such as the translational symmetry. Thus the matrix dimension is 10,626 for four sites, while it grows to 377,348,994 for eight sites.

3. Results

First we briefly discuss the local electron configuration in the atomic limit. As shown in Fig. 1, the $t_{2g}$ level is split by the spin-orbit coupling into $j_{\text{eff}}=\frac{1}{2}$ doublet and $j_{\text{eff}}=\frac{3}{2}$ quartet, in which the eigenstates are characterized by the effective total angular momentum $j_{\text{eff}}$. The

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Local electron configuration and wavefunction of the $t_{2g}$ orbitals under the spin-orbit coupling. To visualize the wavefunction, we draw the surface defined by $r=\sqrt{\sum_\sigma |\psi(\theta, \phi, \sigma)|^2}$ in the polar coordinate with $\sigma$ being real spin, while the color denotes the weight of spin up and down states.}
\end{figure}
Figure 2. The magnitude of the sum total of angular momenta in the whole system at \( U' = 10 \) and \( J = 2 \). (a) Spin and orbital angular momenta. (b) Total and effective total angular momenta.

eigenstates are \( |\alpha\pm\rangle = \frac{1}{\sqrt{3}}|xy\pm\rangle \pm \frac{1}{\sqrt{3}}|yz\mp\rangle + \frac{1}{\sqrt{3}}|zx\mp\rangle \) with eigenenergy \( \lambda \) for the \( j_{\text{eff}} = \frac{1}{2} \) doublet, and \( |\beta\pm\rangle = \frac{1}{\sqrt{2}}|yz\pm\rangle \mp \frac{1}{\sqrt{2}}|zx\mp\rangle \) and \( |\gamma\pm\rangle = -\frac{1}{\sqrt{6}}|xy\pm\rangle \mp \frac{1}{\sqrt{6}}|zx\mp\rangle \) with eigenenergy \( -\frac{1}{2} \) for the \( j_{\text{eff}} = \frac{3}{2} \) quartet, where we introduce \( \beta \) and \( \gamma \) orbitals to distinguish two Kramers doublets in the \( j_{\text{eff}} = \frac{3}{2} \) quartet, and pseudospins to label two states in each Kramers doublet. Note that due to the entanglement of spin and orbital states, the wavefunction exhibits anisotropic charge and spin distributions. Since we have five electrons for an Ir\( ^{4+} \) ion, the lower \( j_{\text{eff}} = \frac{3}{2} \) quartet is fully occupied, while the upper \( j_{\text{eff}} = \frac{1}{2} \) doublet is half-filled.

Now we move on to the Lanczos results. In Fig. 2(a), we present the magnitude of sum total of spin and orbital angular momenta in the whole system, defined by \( \langle (\sum_i S_i)^2 \rangle = S_{\text{tot}}(S_{\text{tot}} + 1) \) and \( \langle (\sum_i L_i)^2 \rangle = L_{\text{tot}}(L_{\text{tot}} + 1) \), respectively. At \( \lambda = 0 \), \( S_{\text{tot}} \) is found to be zero, indicating a spin singlet ground state. As \( \lambda \) increases, \( S_{\text{tot}} \) gradually increases and a sudden change occurs at a transition point, above which the ground state turns to a weak ferromagnetic state. Note that the induced spin moment is reduced from the maximum value \( \frac{1}{2} \times 4 = 2 \). We also find an abrupt change for \( L_{\text{tot}} \), implying that the orbital configuration is reorganized. Figure 2(b) represents the magnitude of sum total of total and effective total angular momenta in the whole system, defined by \( \langle (\sum_i J_i)^2 \rangle = J_{\text{tot}}(J_{\text{tot}} + 1) \) and \( \langle (\sum_i J_{\text{eff},i})^2 \rangle = J_{\text{eff, tot}}(J_{\text{eff, tot}} + 1) \), respectively. \( J_{\text{tot}} \) is enhanced in the weak ferromagnetic state. In contrast, \( J_{\text{eff, tot}} \) decreases and approaches zero in the limit of large \( \lambda \). Thus the weak ferromagnetic state is regarded as a singlet state in terms of the effective total angular momentum.

To clarify how the orbital state changes as \( \lambda \) varies, we measure the charge density in each of the \( t_{2g} \) orbitals, \( n_x = \sum_x \langle \rho_x \rangle \), in different two basis sets. Note that due to the translational invariance, the charge density is equivalent in all sites. In Fig. 3(a), we plot the charge density in the real \( xy \), \( yz \), and \( zx \) orbitals. At \( \lambda = 0 \), we find that \( n_{xy} = 1.5 \) and \( n_{yz} = n_{zx} = 1.75 \), since the itinerancy of orbitals depends on the hopping direction and holes preferably occupy the \( xy \) orbitals rather than the \( yz \) and \( zx \) orbitals. As \( \lambda \) increases, \( n_{xy} \), \( n_{yz} \), and \( n_{zx} \) approach \( \frac{5}{7} \), indicating that \( xy \), \( yz \), and \( zx \) orbital states are mixed with equal weight. Note that \( n_{yz} \) and \( n_{zx} \) are equivalent irrespective of \( \lambda \). Transforming the basis set into the complex \( \alpha \), \( \beta \), and \( \gamma \) orbitals, we can see the characteristics of the orbital state from the viewpoint of the effective total angular momenta. As shown in Fig. 3(b), we observe a sharp change at the transition point, implying again the reorganization of the orbital configuration. For large \( \lambda \), the \( \alpha \) orbitals are singly occupied and relevant to the low-energy property, while the \( \beta \) and \( \gamma \) orbitals are fully
occupied, consistent with the local electron configuration in the atomic limit, as shown in Fig. 1. This clearly indicates the emergence of a complex orbital state, in which spin and orbital states are entangled with complex number coefficients.

4. Summary
We have studied ground-state properties of the $t_{2g}$-orbital Hubbard model with the spin-orbit coupling by Lanczos diagonalization. We have found that due to the spin-orbit coupling, the ground state changes from the spin singlet to the singlet in terms of the effective total angular momentum. The complex orbital state characterized by $J_{\text{eff}} = \frac{1}{2}$ emerges in the spin-orbit-induced state. To clarify novel magnetism in 5$d$ transition metal oxides such as Sr$_2$IrO$_4$, an important issue is to understand the excitation dynamics under the strong spin-orbit coupling, which we will discuss elsewhere in future.

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References
[1] Kim B J, Jun H, Moon S J, Kim J Y, Park B G, Leem C S, Yu J, Noh T W, Kim C, Oh S J, Park J H, Durairaj V, Cao G and Rotenberg E 2008 Phys. Rev. Lett. 101 076402
[2] Kim B J, Ohsumi H, Komesu T, Sakai S, Morita T, Takagi H and Arima T 2009 Science 323 1329
[3] Moon S J, Jin H, Choi W S, Lee J S, Seo S S A, Yu J, Cao G, Noh T W and Lee Y S 2009 Phys. Rev. B 80 195110
[4] Ishii K, Jarrige I, Yoshida M, Ikeuchi K, Mizuki J, Ohashi K, Takayama T, Matsuno J and Takagi H 2011 Phys. Rev. B 83 115121
[5] Okabe H, Isobe M, Takayama-Muromachi E, Koga A, Takeshita S, Hiraishi M, Miyazaki M, Kadono R, Miyake Y and Akimitsu J 2011 Phys. Rev. B 83 155118
[6] Jin H, Jeong H, Ozaki T and Yu J 2009 Phys. Rev. B 80 075112
[7] Jackeli G and Khaliullin G 2009 Phys. Rev. Lett. 102 017205
[8] Watanabe H, Shirakawa T and Yunoki S 2010 Phys. Rev. Lett. 105 216410
[9] Onishi H 2011 J. Phys. Soc. Jpn. 80 SA141
[10] Wang F and Senthil T 2011 Phys. Rev. Lett 106 136402