Variational Monte Carlo study of two-dimensional strong spin-orbit coupling system: Novel Mott insulating state in Ir oxide

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Abstract. Motivated by the novel 5$d$ Mott insulator Sr$_2$IrO$_4$, we have studied the three-orbital Hubbard model with a spin-orbit coupling (SOC) term in a two-dimensional square lattice. We have found the transition from a paramagnetic metal to an antiferromagnetic (AF) insulator at $U = U_{MI}$. A large SOC, which is characteristic of 5$d$ electron systems, greatly reduces $U_{MI}$ and even small $U$ can lead the system into the Mott insulator. Moreover, the Hund’s coupling induces the anisotropy and stabilizes the in-plane AF order. These mechanisms are considered to be valid for Sr$_2$IrO$_4$.

1. Introduction
Recently, the layered 5$d$ transition metal oxide Sr$_2$IrO$_4$ has attracted much attention as a candidate of a novel Mott insulator. In Sr$_2$IrO$_4$, $t_{2g}$ and $e_g$ orbitals are well separated by the large crystalline electrostatic field and the lower $t_{2g}$ orbital is filled with five electrons, $(t_{2g})^5$. In spite of the large band width and small on-site Coulomb interaction $U$, Sr$_2$IrO$_4$ is an insulator with a canted antiferromagnetic (AF) order. It is proposed that the strong spin-orbit coupling (SOC), which is characteristic of 5$d$ electron systems, is responsible for the insulating mechanism and even small $U$ can lead the system into a Mott insulator [1]. The proposed scenario is as follows. The SOC splits the $t_{2g}$ orbital into a $J_{\text{eff}} = 1/2$ state (twofold degenerate) and a $J_{\text{eff}} = 3/2$ state (fourfold degenerate). $J_{\text{eff}}$ denotes the “effective” total angular momentum that is represented by $-L + S$ [1]. When the SOC is large enough, the lower $J_{\text{eff}} = 3/2$ state is fully filled and the upper $J_{\text{eff}} = 1/2$ state becomes an active half-filled energy band. The band width of this half-filled band is much smaller than the original one without the SOC and even small $U$ can lead the system into a Mott insulator. The picture of “$J_{\text{eff}} = 1/2$ Mott insulator” is supported by several experiments such as resonant x-ray scattering [2], angle resolved photoemission spectroscopy [1], and optical conductivity [1]. This novel Mott insulator has a quite different character with usual 3$d$ Mott insulator where the effect of the SOC is only perturbative. However, the detailed theoretical study is still lacking except for first-principle calculations [1, 3]. With these facts in mind, we study the possibility of novel Mott insulating state and magnetic order induced by the strong SOC. The ground state properties of three-orbital Hubbard model with a SOC term are investigated with variational Monte Carlo method. We show that the large SOC works
cooperatively with $U$ and leads the system into a novel Mott insulating state with an in-plane AF order.

2. Model and method

We study the following three-orbital Hubbard model in a two-dimensional square lattice,

$$
H = \sum_{k,\sigma} \varepsilon_{k}(\mathbf{k})c_{k,\sigma}^{\dagger}c_{k,\sigma} + 2\lambda \sum_{i} \mathbf{L}_{i} \cdot \mathbf{S}_{i} + U \sum_{i,\alpha} n_{i\alpha} n_{i\bar{\alpha}} + \sum_{i,\alpha<\beta,\sigma} \left[ U_{\alpha\sigma} n_{i\alpha\sigma} n_{i\beta\bar{\sigma}} + (U_{\beta\bar{\sigma}} - J)n_{i\alpha\sigma} n_{i\beta\bar{\sigma}} \right] + J \sum_{i,\alpha<\beta,\sigma} \left( c_{i\alpha\sigma}^{\dagger} c_{i\beta\bar{\sigma}}^{\dagger} c_{i\beta\sigma} + c_{i\alpha\sigma}^{\dagger} c_{i\beta\bar{\sigma}}^{\dagger} c_{i\beta\bar{\sigma}} + H.c. \right). \tag{1}
$$

The indices $\alpha$ and $\beta$ represent the three $t_{2g}$ orbitals: $yz$ (1), $zx$ (2), and $xy$ (3). The first term is the band energy and the second term is the SOC. They can be written in the matrix form and shown later. The third term is the intraorbital Coulomb interaction and the fourth term is the interorbital Coulomb interactions including $z$-component of Hund’s coupling. The last term is the spin-flip and pair-hopping interactions.

The band energy and SOC are written in the matrix form,

$$
H_{0} = \sum_{k,\sigma} \left( \begin{array}{ccc}
\varepsilon_{1}(\mathbf{k}) & i\sigma\lambda & -\sigma\lambda \\
-i\sigma\lambda & \varepsilon_{2}(\mathbf{k}) & i\lambda \\
-\sigma\lambda & -i\lambda & \varepsilon_{3}(\mathbf{k})
\end{array} \right)
\left( \begin{array}{c}
c_{k,1}\sigma \\
c_{k,2}\sigma \\
c_{k,3}\bar{\sigma}
\end{array} \right).
$$

We can see that the SOC, $\lambda$, mixes the up- and down-spin states and the quasiparticles with pseudospins are newly formed. The total angular momentum $\mathbf{J}$ becomes a good quantum number and the orbital and spin angular momentum are no longer conserved quantities.

First, we construct the non-interacting tight-binding energy band to reproduce the first-principle band calculation. In Sr$_2$IrO$_4$, there is a large hybridization between $xy$ and $x^2 - y^2$ orbitals originated from the rotation of IrO$_6$ octahedra and the band structure becomes quite different from the one without rotation. Figure 1 shows the energy bands with and without the rotation of octahedra and SOC. To take account of the effect of rotation, we introduce next-nearest and third-nearest hopping integrals in $xy$ orbital. The chemical potential $\mu_3$ is also introduced to take account of the tetragonal splitting. Then the energy dispersion of $xy$ orbital is $\varepsilon_{3}(\mathbf{k}) = 2t_{1}(\cos k_{x} + \cos k_{y}) + 4t_{2}\cos k_{x}\cos k_{y} + 2t_{3}(\cos 2k_{x} + \cos 2k_{y}) + \mu_3$. $yz$ and $zx$ orbitals have almost one-dimensional character, $\varepsilon_{1}(\mathbf{k}) = 2t_{5}\cos k_{x} + 2t_{4}\cos k_{y}$ and $\varepsilon_{2}(\mathbf{k}) = 2t_{4}\cos k_{x} + 2t_{5}\cos k_{y}$ ($t_{4} \gg t_{5}$). These tight-binding parameters are chosen as $(t_{1}, t_{2}, t_{3}, t_{4}, t_{5}, \mu_3, 2\lambda) = (1.0, 0.5, 0.25, 1.028, 0.167, -0.1, 1.028)$ and they well reproduce the first-principle band dispersion and Fermi surface [1]. In the following, we set $t_{1} = t$ as an energy unit.

Next, we consider the effect of Coulomb interaction. The following trial wave function is used in this study,

$$
|\Psi\rangle = P_{3}\rho_{G}|\Phi\rangle. \tag{3}
$$

$|\Phi\rangle$ is the one-body part and obtained by diagonalizing $H_{0}$ with renormalized tight-binding parameters, $H_{0}(\vec{t}_{1}, \vec{\mu}_{3}, \vec{\lambda}_{\alpha\beta})$. Note that by the effect of Coulomb interaction and tetragonal splitting, the “effective” SOC has orbital dependence: $\lambda \rightarrow \lambda_{\alpha\beta}$. To consider the magnetically ordered state, the AF order parameter is added to $H_{0}$. Here, we study the out-of-plane AF order (Fig. 2(a)) and in-plane AF order (Fig. 2(b)). Note that the arrows in Fig. 2 represent the pseudospins, not the original spins. Since the rotational symmetry in pseudospin
Non-interacting tight-binding energy band (a) without rotation of octahedra and SOC, (b) with rotation and without SOC, and (c) with rotation and SOC.

space is broken by Hund’s coupling, these two states should have different energies. Then the matrix becomes $12 \times 12$ for the AF state (not shown here because of the limiting space).

$$P_G = \prod_{i, \gamma} [1 - (1 - g_\gamma) |\gamma_i\rangle \langle \gamma_i|]$$

is a Gutzwiller factor extended for the three-orbital system [4]. $i$ is a site index and $\gamma$ runs over possible spin configurations at each site. In the three-orbital system, the total number of the spin configuration at each site is $4^3 = 64$. We control the weight of each spin configuration by varying $g_\gamma$ from 0 to 1. It is a natural extension of Gutzwiller factor in the single-band model. The set of $\{g_\gamma\}$ is a variational parameter and optimized so as to give the energy minimum. In this study, we classify the possible 64 patterns into 12 groups by Coulomb interaction and therefore the number of variational parameter is 12.

$$P_{3c} = \exp \left[ -\sum_{i \neq j} v_{ij} n_i n_j \right]$$

is a charge Jastrow factor that controls the long-range charge correlation. The long-range charge correlation is known to be important for describing Mott transition [5, 6]. In this study, we assume that $v_{ij}$ depends only on the distance between charges, $v_{ij} = v(|r_i - r_j|)$. Since $i$ and $j$ run over all lattice sites, the number of variational parameter increases with the system size. For 10×10 square lattice, we consider up to 19th-neighbor correlation and therefore the number of variational parameter is 19.

The ground state energy and other physical quantities are calculated with the variational Monte Carlo method. The variational parameters mentioned above are simultaneously optimized by using the stochastic reconfiguration method [7]. This method makes it possible to optimize many parameters efficiently and stably. Once the trial wave function is determined, we can easily calculate physical quantities.

3. Numerical Results

Figure 2(c) shows the ground state phase diagram of the three-orbital Hubbard model in a two-dimensional 10×10 square lattice. The transition from the paramagnetic metal to the AF insulator occurs at $U = U_{\text{MI}} \sim 3.8t$. This value is much smaller than the one in a weak SOC case. When the AF order parameter is introduced, the degeneracy along $(\pi/2, \pi/2)-(\pi, 0)$ is lifted and the highest band is split away by the AF gap. As shown in Figs. 1(b) and 1(c), the SOC lifts up the two branches out of six energy bands and it makes it easier to fully open the AF gap. Therefore, the larger the SOC, the easier the system becomes an AF insulator. Indeed, our calculation shows that the $U_{\text{MI}}$ monotonically decreases with increasing the SOC, $U_{\text{MI}}/t = 8.8, 5.5, 3.8, 3.1$ for $2\lambda/t = 0.2, 0.6, 1.028, 1.4$, respectively. Moreover, as $U$ increases, the effective SOC also increases and they work cooperatively for insulating state. We expect
that this insulating mechanism is valid for Sr$_2$IrO$_4$. Although the Coulomb interaction $U$ is much smaller than the band width and naively the metallic state is expected, Sr$_2$IrO$_4$ is an insulator with a canted AF order. This fact can be explained if the SOC in Sr$_2$IrO$_4$, $\lambda_{Ir}$, is large enough to reduce $U_{MI}$ smaller than $U_{Ir}$. Indeed, $\lambda_{Ir}$ is estimated as 0.4-0.5eV, much larger than 3d and 4d electron systems, and our calculation indicates $U_{MI} \sim 1$eV. We expect that $U_{Ir} > U_{MI}$ is satisfied and the “spin-orbit induced” Mott insulator is realized in Sr$_2$IrO$_4$.

In the insulating region, the in-plane AF order (x-AFI, Fig. 2(b)) is stabilized. The out-of-plane AF order (z-AFI, Fig. 2(a)) has always higher energy than x-AFI and does not appear in the phase diagram. The origin of this energy difference is a Hund’s coupling. If there is no Hund’s coupling, the pseudospin SU(2) symmetry is preserved and z-AFI and x-AFI are energetically degenerated. However, the Hund’s coupling introduces the anisotropy and the in-plane magnetism (x-AFI) is more favored than the out-of-plane magnetism (z-AFI). This result is consistent with the study of effective strong SOC spin model by Jackeli and Khaliullin [8]. The magnetic x-ray diffraction experiment [2] also supports the in-plane magnetism in Sr$_2$IrO$_4$.

To explain the canted AF order observed in the experiment, more detailed band structure and the introduction of y component of the magnetic moment is necessary. It is an interesting future problem.

4. Summary

We have studied the three-orbital Hubbard model with a SOC term in a two-dimensional square lattice. We have shown that the spin-orbit induced AF insulator is realized for $U > U_{MI}$. With a large SOC, $U_{MI}$ is greatly reduced and even small $U$ can lead the system into the Mott insulator. The Hund’s coupling induces the anisotropy and stabilizes the in-plane AF order.

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