Theoretical study of $J_{\text{eff}} = 1/2$ Mott insulator in Ir oxides: a strong spin-orbit coupling vs local electron correlations

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Abstract. The variational cluster approximation (VCA) is used to study the nature of a novel Mott insulator induced by a strong spin-orbit coupling along with moderate electron interactions for a two-dimensional three-band Hubbard model, consisting of the $t_{2g}$ manifold of 5$d$ orbitals, with five electrons (i.e., one hole) per site. To extend the variational space, we introduce a generalized local Kramer’s doublet as a variational state for VCA. We find that the Hund’s coupling induces magnetic anisotropy and stabilizes in-plane antiferromagnetic order constructed by the Kramer’s doublet. By carefully analyzing the expectation values of the generalized Kramer’s double operator, we also find that the local hole state in the Mott insulator is comparable to the Kramer’s doublet with $J_{\text{eff}} = 1/2$.

Sr$_2$IrO$_4$ [1], a 5$d$ transition metal oxide, has attracted much attention because of its novel Mott insulating behavior as well as a possible candidate for a superconductor similar to cuprates [2]. Sr$_2$IrO$_4$ is in a layered perovskite structure of K$_2$NiF$_4$ type [3], and each Ir ion is surrounded by six oxygens forming an ochahedral cage. Because of the large crystalline electrostatic field, Ir$^{4+}$ is the low-spin state with $(t_{2g})^5$ electron configuration. An inelastic neutron scattering experiment has found that the ground state of Sr$_2$IrO$_4$ is antiferromagnetically ordered with weak ferromagnetic moment caused by rotating the ochahedral cage [3, 4]. Recent x-ray scattering experiments have revealed that the ground state is close to a $J_{\text{eff}} = 1/2$ Mott insulator [5]. Here, $J_{\text{eff}} = |S - L|$ is an effective total angular momentum defined in the $t_{2g}$ manifold with the spin $S$ and the orbital angular $L$ momenta. The creation operator of $J_{\text{eff}} = 1/2$ state is given by $(s_{\sigma}c_{y\sigma} + c_{y\bar{\sigma}}^\dagger + i s_{\sigma}c_{x\bar{\sigma}}^\dagger)/\sqrt{3}$, where $c_{x\sigma}$ is a creation operator with spin $\sigma (=\uparrow, \downarrow)$ and orbital $\alpha (=xy, yz, zx)$, and with $s_{\sigma} = +1 (-1)$ for $\sigma = \uparrow (\downarrow)$ ($\bar{\sigma}$ indicates the opposite spin of $\sigma$). It should be emphasized that, although the $J_{\text{eff}} = 1/2$ state corresponds to the ground state in the atomic limit with a finite spin-orbit coupling, the realization of a $J_{\text{eff}} = 1/2$ Mott insulator in actual materials is highly non trivial. Therefore, we study the three-band Hubbard model with a spin-orbit coupling $\lambda$ [6] by using variational cluster approximation [7] (VCA) based on the self-energy functional theory [8] (SFT), which can treat electron correlations and spin-orbit coupling on the same footing.

We consider the following effective three-band Hubbard model, consisting of three $d$ orbitals...
(d_{xy}, d_{yz}, and d_{zx}), on the square lattice

\begin{equation}
H = H_{\text{kin}} + H_{\text{so}} + H_{\text{int}}
\end{equation}

\begin{align*}
H_{\text{kin}} &= \sum_{k\sigma} \epsilon_{k} c_{k\sigma}^{\dagger} c_{k\sigma}, \\
H_{\text{so}} &= \lambda \sum_{r} \mathbf{\ell}_{r} \cdot \mathbf{s}_{r}, \\
H_{\text{int}} &= U \sum_{\alpha} n_{r\alpha} \ldots + \frac{U'}{2} \sum_{\alpha \neq \beta} n_{r\alpha} n_{r\beta} + \frac{1}{2} \sum_{\alpha \neq \beta} n_{r\alpha} n_{r\beta} + 1/2 \left( U' - J \right) \sum_{r} \sum_{\alpha \neq \beta} n_{r\alpha} n_{r\beta} - J \sum_{r} \sum_{\alpha \neq \beta} c_{r\alpha}^{\dagger} c_{r\beta} c_{r\beta}^{\dagger} + J' \sum_{r} \sum_{\alpha \neq \beta} c_{r\alpha}^{\dagger} c_{r\beta}^{\dagger} c_{r\beta} c_{r\beta}^{\dagger}.
\end{align*}

Here, $c_{r\alpha}$ (or $c_{r\sigma}^{\dagger}$) is the annihilation (creation) operator of an electron with spin $\sigma$ ($\sigma = \uparrow, \downarrow$) and orbital $\alpha$ ($\alpha = xy, yz, \text{and} zx$) at site $r$. $c_{k\sigma}$ is the Fourier transform of $c_{r\sigma}$. $\epsilon_{k}$ is the dispersion of orbital $\alpha$, $\epsilon_{k}^{xy} = -2t_{1} \cos k_{x} \cos k_{y} - 4t_{2} \cos k_{x} \cos k_{y} - 2t_{3} \cos 2k_{x} + \cos 2k_{y} + \Delta$, $\epsilon_{k}^{yz} = -2t_{4} \cos k_{y} - 2t_{5} \cos k_{y} - 2t_{6} \cos k_{y}$, where $t_{1}, t_{2}, t_{3}$ correspond to the hopping integrals for $d_{xy}$ orbitals located at the nearest, next nearest, and third nearest neighbor sites, respectively. $t_{4}$ and $t_{5}$ are the nearest neighbor hopping integrals for $d_{yz}$ ($d_{zx}$) orbital in $y$ ($x$) and $x$ ($y$) directions, respectively. $\Delta$ is an energy level difference between $d_{xy}$ orbital and the other orbitals ($d_{yz}$ and $d_{zx}$), which is induced by an additional tetragonal crystalline electrostatic field. $H_{\text{so}}$ is the spin-orbit coupling term represented in the following matrix form

\begin{equation}
2\mathbf{\ell}_{r} \cdot \mathbf{s}_{r} = \sum_{\alpha} \left( \begin{array}{ccc}
\epsilon_{rxy} & \epsilon_{ryz} & \epsilon_{rzx} \\
\epsilon_{rxy} & \epsilon_{ryz} & \epsilon_{rzx} \\
\epsilon_{rxy} & \epsilon_{ryz} & \epsilon_{rzx}
\end{array} \right) \left( \begin{array}{ccc}
-s_{\sigma} & -i & 0 \\
-s_{\sigma} & 0 & i s_{\sigma} \\
i s_{\sigma} & -i & 0
\end{array} \right) \left( \begin{array}{c}
c_{rxy\sigma} \\
c_{ryz\sigma} \\
c_{rzx\sigma}
\end{array} \right),
\end{equation}

where $s_{\sigma} = +1$ ($-1$) for $\sigma = \uparrow$ ($\downarrow$). Finally, we introduce, for the local Coulomb interactions, the intra-orbital ($U$) and the inter-orbital ($U'$) Coulomb interactions, the Hund’s coupling $J$, and the pair-hopping $J'$, with $U = U' + 2J$ and $J = J'$ [9]. The number of electrons per site $N_{e}$ is set to be 5. We use a set of parameters ($t_{1}, t_{2}, t_{3}, t_{4}, t_{5}, \Delta$) = (0.36, 0.18, 0.09, 0.37, 0.06, 0.37, –0.36) eV [6], which can well reproduce the dispersion calculated by local density approximation with spin-orbit coupling [10].

We adopt the VCA method [7] based on the SFT [8]. The SFT introduces a reference Hamiltonian $H'$ with the same two-body interactions as the original Hamiltonian $H$ but with a different one-body part, and $H'$ may be solved numerically on a finite cluster. A approximate grand potential for $H$ is given in a functional form by $\Omega(\mathbf{t}') = \Omega' - \text{Tr} \ln(-\hat{G}_{0}^{-1} + \hat{\Sigma}(\mathbf{t}')) + \text{Tr} \ln(-\hat{G}^{-1}(\mathbf{t}'))$, where $\mathbf{t}'$ is the parameter set of one-body part in $H'$. $\Omega'$, $\hat{\Sigma}(\mathbf{t}')$, and $\hat{G}(\mathbf{t}')$ are the ground potential, self-energy, and Green’s function of the reference system $H'$, respectively. $\hat{G}_{0}$ is the non-interacting Green’s function of $H$. The variational condition $\partial \Omega(\mathbf{t}')/\partial \mathbf{t}' = 0$ determines an appropriate reference system $H'$, which describes the original system $H$ approximately.

To study the symmetry-broken long-range-ordered states in the VCA, we introduce suitably chosen fictitious Weiss fields in a set of variational parameters $\mathbf{t}'$. In this study, we introduce the Weiss field acting on a local Kramer’s doublet

\begin{equation}
H_{x}' = h_{x} \sum_{r\sigma} e^{iQ_{x} r a_{r\sigma}^{\dagger} a_{r\sigma}}, \quad H_{z}' = h_{z} \sum_{r\sigma} e^{iQ_{z} r s_{\sigma} a_{r\sigma}^{\dagger} a_{r\sigma}},
\end{equation}

with $Q = (\sigma, \pi)$. Here, $a_{r\sigma}^{\dagger}$ ($a_{r\sigma}$) is a creation (annihilation) operator of the local Kramer’s doublet defined as

\begin{equation}
a_{r\sigma}^{\dagger} = s_{\sigma} \cos \theta c_{rxy\sigma}^{\dagger} + \frac{\sin \theta}{\sqrt{2}} \left( c_{ryz\sigma}^{\dagger} + i s_{\sigma} c_{rzx\sigma}^{\dagger} \right).
\end{equation}
The Weiss field $H'_x$ and $H'_z$ correspond to the in-plane and out-of-plane antiferromagnetic (AF) ordering, respectively. The reference system used here consists of 4 sites and the variational parameters $t'$ are $h'_x$, $(h'_z)$, and $\theta'$, which are given in Eq. (3), in addition to chemical potential $\mu'$.

Figure 1 shows our results of the ground state energy per site $E = \Omega + \mu N_e$ as a function of $h'_x$ ($h'_z$) with different $\theta'$ for $U/t_1 = 4$ and $J/U = 0.15$. It is clearly seen in Figure 1 that the minimum of $E$ appears at a finite value of $h'_x$ ($h'_z$), indicating that the ground state is antiferromagnetically ordered. It is also interesting to notice that the ground state energy for the in-plane AF ordering ($h'_x$) is smaller than that for the out-of-plane AF ordering ($h'_z$). The difference $\Delta_h$ of the minimum energies between the optimized states with the in-plane and out-of-plane AF orderings is summarized in Table 1. It is observed in Table 1 that the energy difference $\Delta_h$ becomes zero with $J/U \rightarrow 0$. This strongly suggests that the Hund’s coupling is the origin of the magnetic anisotropy. This finding is indeed consistent with the earlier theoretical study from the strong coupling limit [11].

Next, we estimate the charge gap $\Delta_{c,x}$ ($\Delta_{c,z}$) for in-plane (out-of-plane) AF state by investigating the profile of chemical potential $\mu$ versus electron density $N_e$, i.e., searching a region where $-\partial^2 \Omega / \partial \mu^2 = 0$. Our results for $\Delta_{c,x}$ and $\Delta_{c,z}$ for different $J/U$ with $U/t_1 = 4$ are summarized in Table 1. One can see in Table 1 that $\Delta_{c,x}$ and $\Delta_{c,z}$ are comparable and are both finite for a range of $J/U$ studied. The fact that the charge gap is finite is the direct evidence that the ground state is in fact insulating. It is also intriguing to notice that the charge gap decreased monotonically with increasing $J$. The reason of this monotonically decreasing behavior of charge gap is because we set $U = U' + 2J$ and $J = J'$, and thus $U'$ simply decreases with increasing $J$. Indeed, in the atomic limit, the charge gap is given by

$$\Delta_e = U - \frac{J}{2} + 3\lambda - \sqrt{\frac{(2\lambda + 5J)^2}{4} + 8\lambda^2}, \quad (5)$$

and thus monotonically decreases with increasing $J$.

Finally, let us study the nature of the insulating state obtained above. If we assume the local one hole state in the Mott insulator can be represented by the generalized local Kramer’s doublet defined in Eq. (4), the optimal $\theta = \theta_{opt}$ describing this local hole state should be defined by maximizing the hole density, i.e.,

$$\partial_{\theta} \sum_{\sigma} \langle a_{\sigma \gamma \sigma} a_{\sigma \gamma \sigma}^\dagger \rangle = 0 \quad (6)$$

where $\langle \cdot \cdot \rangle$ corresponds to average taken for the ground state. Note that in general $\theta_{opt}$ determined in Eq. (6) is different from $\theta'$ defined in Eq. (3). This is because the latter is simply a variational parameter introduced to optimize the self-energy, whereas the former corresponds
to a property of the ground state. The optimal values $\theta_{\text{opt}}$ obtained for different $J/U$ are summarized in Table 1. As seen in Table 1, $\theta_{\text{opt}}$ is found almost no $J/U$ dependent, and the value is about $\theta = 63.5^\circ \pm 0.5^\circ$. This value is very close to the one expected in the $J_{\text{eff}} = 1/2$ limit, i.e., $\theta = \arctan \sqrt{2} \sim 54.74^\circ$. The small deviation from the $J_{\text{eff}} = 1/2$ state is caused by slight suppression of $d_{xy}$ component. We should note that $\theta_{\text{opt}}$ is also related to the expectation values of the local spin $\langle S_x \rangle$ and local orbital $\langle L_x \rangle$ angular momenta [12]. For instance, the ratio of $\langle L_x \rangle / \langle S_x \rangle$ is given by $2\sqrt{2}\tan \theta$ if we assume that the ground state is described by $|\psi\rangle = \prod_i (a^{\dagger}_{i\uparrow} + e^{iQx}a^{\dagger}_{i\downarrow}) |0\rangle$. Although this assumption seems oversimplified, this relation is found to be almost satisfied in our calculation with $\langle L_x \rangle / \langle S_x \rangle \sim 5.7-6.0$. It is interesting to note that these values are rather consistent with recent experiments on Sr$_2$IrO$_4$, for which $\langle L \rangle / \langle S \rangle \sim 6$ has been observed. Here $\langle L \rangle$ and $\langle S \rangle$ are the corresponding angular momentum components parallel to the magnetic moment [13].

In summary, we have studied the three-band Hubbard model with the spin-orbit coupling using VCA for Sr$_2$IrO$_4$. We have found that the in-plane AF states, consisting of the pseudo-spin of a local Kramer’s doublet state, are stabilized by the Hund’s coupling. We have also shown that the obtained local Kramer’s doublet of the Mott insulating state is comparable to the one with $J_{\text{eff}} = 1/2$.

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Table 1. $J/U$ dependence of the charge gap $\Delta_{c,x}$ ($\Delta_{c,z}$) for the in-plane (out-of-plane) AF state, the difference $\Delta_h$ of the minimum energies between the optimized in-plane and out-of-plane AF states, the optimal $\theta_{\text{opt}}$ derived from Eq.(6), and the expectation values of $\langle S_x \rangle$ and $\langle L_x \rangle$ for the ground state. $U/t_1 = 4.0$ is used.

| $J/U$ | $\Delta_{c,x}/t_1$ | $\Delta_{c,z}/t_1$ | $\Delta_h/t_1$ | $\theta_{\text{opt}}$ | $\langle S_x \rangle$ | $\langle L_x \rangle$ |
|-------|-------------------|-------------------|----------------|------------------|----------------|----------------|
| 0.00  | 1.784             | 1.784             | 0.00000        | 64.475           | 0.0642         | 0.3835         |
| 0.05  | 1.486             | 1.464             | 0.00204        | 64.181           | 0.0634         | 0.3747         |
| 0.10  | 1.135             | 1.095             | 0.00464        | 63.823           | 0.0617         | 0.3604         |
| 0.15  | 0.752             | 0.704             | 0.00795        | 63.401           | 0.0588         | 0.3381         |
| 0.20  | 0.358             | 0.278             | 0.01218        | 62.933           | 0.0538         | 0.3042         |

\[ \theta_{\text{opt}} = \arctan \left( \frac{\Delta_h}{\Delta_{c,x}} \right) \]