Variational cluster approximation study of Mott transition with strong spin-orbit coupling

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Abstract. Motivated by recent experiments on Sr\(_2\)IrO\(_4\), the ground state magnetic and electronic structures are studied theoretically for a two-dimensional three-band Hubbard model with strong spin-orbit coupling. To treat spin-orbit coupling, local Coulomb interactions, and band structure effects on the same footing, the variational cluster approximation based on the self-energy functional theory is employed. It is found that for a relatively large coupling region, the ground state is an anisotropic antiferromagnetic Mott insulator of an effective local angular momentum \(J_{\text{eff}} = 1/2\) with \(xy\) plane as an easy plane. This anisotropy is caused by the strong spin-orbit coupling along with the inter-orbital Hund’s coupling. The momentum resolved one-particle excitations are also studied for the Mott insulating phase. It is found that the low-energy one-particle excitations consist mostly of the \(J_{\text{eff}} = 1/2\) state, a direct evidence of a novel \(J_{\text{eff}} = 1/2\) Mott insulator.

1. Introduction

Recent experiments on Sr\(_2\)IrO\(_4\) [1] have revealed a novel Kramers doublet \(J_{\text{eff}} = 1/2\) Mott insulator induced by strong spin-orbit coupling (SOC) and local Coulomb interactions among Ir 5\(d\) electrons. Sr\(_2\)IrO\(_4\) is a layered perovskite structure of K\(_2\)NiF\(_4\) type [2], and each Ir ion is surrounded by six oxygens forming an octahedral cage. Because of the large crystalline electrostatic field, Ir\(^{4+}\) is the low-spin state with \((t_{2g})^5\) electron configuration. Sr\(_2\)IrO\(_4\) is thus very similar to a 4\(d\) counterpart of Sr\(_2\)RhO\(_4\) [3]. Because the local Coulomb repulsion (the band width) becomes smaller (larger) with increasing the period in the same transition metal group, it is expected that Sr\(_2\)IrO\(_4\) should be more metallic-like than Sr\(_2\)RhO\(_4\). However, the experimental observations have shown the opposite: while Sr\(_2\)RhO\(_4\) is a metal [4], Sr\(_2\)IrO\(_4\) is an insulator [1]. This seemingly counterintuitive behavior was understood [1] by noticing that the larger SOC (\(\lambda\)) of about 0.4 eV in Sr\(_2\)IrO\(_4\) further splits the sixfold degenerate \(t_{2g}\) states (including spins) into twofold degenerate \(J_{\text{eff}} = 1/2\) states and fourfold degenerate \(J_{\text{eff}} = 3/2\) states. Four of the five electrons in Ir\(^{4+}\) ion fully occupy the \(J_{\text{eff}} = 3/2\) states, and the remaining electron partially occupy one of the \(J_{\text{eff}} = 1/2\) states. This half-filled \(J_{\text{eff}} = 1/2\) states form a band with smaller band width than the original \(t_{2g}\) band without SOC, and thus it becomes a Mott insulator once even smaller on-site Coulomb interactions than that for 4\(d\) electrons are considered. This picture is consistent with several experimental observations including angle resolved photoemission spectroscopy [1], optical conductivity [1], and resonant x-ray scattering [5]. However, theoretically, only the first-principles band structure calculations
have been reported [1, 6], and a systematic study based on many-body correlated-electron theory is highly desired.

The purpose of this paper is to study theoretically effects of the strong SOC as well as the local Coulomb interactions, including the Hund’s coupling, on the ground state phase diagram and the low-lying one-particle excitations for Sr$_2$IrO$_4$. Here the variational cluster approximation based on the self-energy functional theory is used for a multi-orbital Hubbard model to treat the local static and dynamical fluctuations exactly. We found that the strong SOC along with the Hund’s coupling induces an anisotropic antiferromagnetic Mott insulator of $J_{\text{eff}} = 1/2$ with $xy$ plane as an easy plane. We also found that the low-energy one-particle excitations can be described mostly by quasi-particle excitations of $J_{\text{eff}} = 1/2$ characteristics.

2. Model and Method

As mentioned above, the local electronic configuration of Ir$^{4+}$ ion in Sr$_2$IrO$_4$ is the low-spin ($t_{2g}$)$^5$ because of the large crystal field effect, which has also been estimated much larger than the SOC and the local Coulomb interactions [6]. Therefore, we use the following effective Hubbard model for Sr$_2$IrO$_4$, consisting of three orbitals ($d_{xy}$, $d_{yz}$, and $d_{zx}$), defined on the square lattice:

$$
H = H_t + H_U,
$$

$$
H_t = \sum_{i,\alpha} E_{\alpha} n_{i,\alpha,\sigma} + \sum_{k,\alpha,\sigma} \varepsilon_{\alpha}(k) c_{k\alpha\sigma}^{\dagger} c_{k\alpha\sigma} + 2\lambda \sum_{i} L_i \cdot S_i,
$$

$$
H_U = U \sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow} + \frac{1}{2}(U’ - J) \sum_{i,\alpha,\sigma} \sum_{\alpha \neq \beta} n_{i,\alpha,\sigma} n_{i,\beta,\sigma} + \frac{U’}{2} \sum_{\alpha \neq \beta} n_{i,\alpha,\sigma} n_{i,\beta,\sigma},
$$

$$
-J \sum_{i,\alpha \neq \beta} c_{i,\alpha,\uparrow}^{\dagger} c_{i,\beta,\alpha} c_{i,\beta,\downarrow} + J’ \sum_{i,\alpha \neq \beta} c_{i,\alpha,\uparrow}^{\dagger} c_{i,\beta,\alpha} c_{i,\beta,\downarrow} + J’ \sum_{i,\alpha \neq \beta} c_{i,\alpha,\downarrow}^{\dagger} c_{i,\beta,\alpha} c_{i,\beta,\uparrow}.
$$

Here, $c_{i,\alpha,\sigma}^{\dagger}$ represents the creation operator of an electron with spin $\sigma = (\uparrow, \downarrow)$ and orbital $\alpha = (xy, yz, zx)$ on site $i$, and $n_{i,\alpha,\sigma} = c_{i,\alpha,\sigma}^{\dagger} c_{i,\alpha,\sigma}$. $E_{\alpha}$ is on-site potential for orbital $\alpha$, and we set $E_{yz} = E_{zx} \neq E_{xy}$ reflecting the tetragonal symmetry of the system. The energy dispersions of $xy$, $yz$, and $zx$ orbital are $\varepsilon_{xy}(k) = -2t_1 \cos k_x + \cos k_y - t_2 \cos k_x \cos k_y - t_3 (\cos 2k_x + \cos 2k_y)$, $\varepsilon_{yz}(k) = -2t_4 \cos k_y + t_2 \cos k_x$, and $\varepsilon_{zx}(k) = -2t_4 \cos k_x - t_2 \cos k_y$ with $(t_2, t_3, t_4, t_5) = (0.5t_1, 0.25t_1, 1.028t_1, 0.167t_1)$ where $t_1 = 0.36$ eV, respectively. Restricting the Hilbert space to the $t_{2g}$ orbitals, the SOC term is written as

$$
2L_i \cdot S_i = \sum_{\alpha} \begin{pmatrix} c_{i,x,y,\sigma}^{\dagger} & c_{i,y,z,\sigma}^{\dagger} & c_{i,z,x,\sigma}^{\dagger} \\ -s & 0 & -i \\ -s & 0 & is \\ i & -is & 0 \end{pmatrix} \begin{pmatrix} c_{i,x,y,\bar{\sigma}} \\ c_{i,y,z,\bar{\sigma}} \\ c_{i,z,x,\bar{\sigma}} \end{pmatrix},
$$

where $s = +1$ ($-1$) for $\sigma = \uparrow$ ($\downarrow$). For the local Coulomb interactions, we introduce the intra-orbital ($U$) and the inter-orbital ($U’$) Coulomb interactions, the Hund’s coupling $J$, and the hopping $J’$, with $U = U’ + 2J$ and $J = J’$ [7]. The electron density is set to be 5 per site.

We adopt the variational cluster approximation (VCA) method [8] based on the self-energy functional theory (SFT) [9]. This method takes into account precisely effects of short-range static and dynamical correlations, and thus superior to simple mean field approximations. The method has been applied successfully to a wide range of strongly correlated electronic systems to study, e.g., competition of various phases in high-$T_c$ cuprates [10, 11] and in layered organic conductors [12]. 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 $(t’)$, and $H'$ may be solved numerically on a finite cluster. An approximate grand potential $\Omega |\Sigma(t')|$ of the original system $H$ is given in a functional form by $\Omega |\Sigma(t')| = \Omega’ - \text{Tr} \ln \left( -G_0^{-1} + \Sigma(t') \right) + \text{Tr} \ln \left( -G^{-1} \right)$,
where $\Omega'$, $\Sigma'(t')$, and $G'$ are the exact ground potential, the exact self-energy, and the exact Green function of the reference system $H'$, respectively. $G_0$ is the non-interacting Green function of the original system $H$. The condition $\partial \Omega [\Sigma(t')] / \partial t'|_{t'=t''} = 0$ determines an appropriate reference system which describes the original system approximately.

In investigating broken symmetry phases, VCA treats the short-range correlations exactly within a cluster, without introducing any factorization of the interaction terms, yet the obtained Green function is still defined on the infinite lattice. The only approximation comes from the limited forms of the self-energy functional, e.g., the size of clusters and the number of variational parameters, on which the variational principle is applied. To discuss the symmetry-breaking long-range ordered states in VCA, we introduce suitably chosen fictitious Weiss fields in the set of variational parameters $t'$ [8, 14, 15]. The Weiss field term associated with the magnetic order considered in this study is expressed as $H'_{h_z} (\alpha = x, z)$ where $H'_{h_z} = h'_{z} \sum_{i,\sigma} e^{i\mathbf{q} \cdot \mathbf{R}_i} \gamma_{i,\sigma}^{\dagger} \gamma_{i,\sigma}$. Here $\gamma_{i,\sigma} = (c_{i,xy,\sigma} - sc_{i,yz,\sigma} - isc_{i,zx,\sigma})/\sqrt{3}$ is the annihilation operator of the $J_{\text{eff}} = 1/2$ state at site $i$. $\mathbf{q} = (\pi, \pi)$ [antiferromagnetic (AF) order], and $\mathbf{R}_i$ represents the position of site $i$. The Weiss fields $H'_{h_x}$ and $H'_{h_y}$ correspond to the out-of-plane and in-plane magnetic ordering, respectively. The reference system used here consists of 2 sites and the variational parameters $t'$ are the Weiss field $h'_{x}$ ($h'_{z}$) and the chemical potential $\mu'$.

### 3. Numerical Results

The results for $\Omega [\Sigma(h'_{\alpha})] - \Omega [\Sigma(0)]$ with $U/t_1 = 4$ and several values of $J/U$ are shown in Fig.1. It is found that the minimum of the grand potential appears at a finite value of $h'_{\alpha}$, indicating that the ground state is AF Mott insulating. It is also interesting to notice that the grand potential for the in-plane AF ordering ($h'_{x}$) is smaller than that for the out-of-plane AF ordering ($h'_{z}$), and the energy difference becomes zero with $J/U \rightarrow 0$. This strongly suggests that the Hund’s coupling is the origin of the magnetic anisotropy. Our finding is consistent with the recent theoretical study [13] and also in qualitative agreement with experiments [2].

![Figure 1](image_url) (Color online) The grand potential $\Omega [\Sigma(h'_{\alpha})] - \Omega [\Sigma(0)]$ in the Mott insulating phase for several values of $J/U$ with $U/t_1 = 4$, $\lambda/t_1 = 0.514$, and $(E_{xy} - E_{yz})/t_1 = -1.0$.

We then calculate the one-particle excitation spectra [14], and the results are shown in Fig.2. It is clearly seen that the unoccupied states are well separated from the occupied states, forming a well-defined upper Hubbard band. Projecting the spectral weight onto the $J_{\text{eff}} = 1/2$ state ($\gamma_{i,\sigma}$ and $\gamma_{i,\sigma}^{\dagger}$), we find that most of the low-energy one-particle excitations, especially the upper Hubbard band, is described by quasi-particles with $J_{\text{eff}} = 1/2$ characteristics, except for electron
removal excitation spectra at k's around (0, 0). These results support the picture of a novel $J_{\text{eff}} = 1/2$ Mott insulator mentioned in Introduction.

Figure 2. (Color online) Momentum resolved one-particle excitation spectra $A(k, \omega)$ for $U/t_1 = 3.5, J/U = 0.1, \lambda/t_1 = 0.514, \text{and } (E_{xy} - E_{yz})/t_1 = -1.0 \text{ with } t_1 = 0.36 (\text{eV})$. For comparison, projected one-particle excitation spectra onto $J_{\text{eff}} = 1/2$ states are also shown in dashed lines. Here $\Gamma$, $X$, and $M$ correspond to $q = (0, 0)$, $(\pi, \pi)$, and $(\pi, 0)$, respectively, in antiferromagnetic Brillouin zone. A vertical line at $\omega = 0$ corresponds to Fermi energy.

4. Summary
We applied the VCA to study the ground state magnetic and electronic structures of the three-band Hubbard model with the strong SOC for Sr$_2$IrO$_4$. We found that the Hund’s coupling and the strong SOC induce an anisotropic AF Mott insulator with $xy$ plane as an easy plane. We also found that the low-energy one-particle excitations is described mostly by quasi-particles with $J_{\text{eff}} = 1/2$ characteristics.

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