Dilemma in strongly correlated materials: Hund’s metal vs relativistic Mott insulator

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We point out the generic competition between the Hund’s coupling and the spin-orbit coupling in correlated materials, and this competition leads to an electronic dilemma between the Hund’s metal and the relativistic Mott insulators. Hund’s metals refer to the fate of the would-be insulators where the Hund’s coupling suppresses the correlation and drives the systems into correlated metals. Relativistic Mott insulators refer to the fate of the would-be metals where the relativistic spin-orbit coupling enhances the correlation and drives the systems into Mott insulators. These contradictory trends are naturally present in many correlated materials. We study the competition between Hund’s coupling and spin-orbit coupling in correlated materials and explore the interplay and the balance from these two contradictory trends. The system can become a spin-orbit-coupled Hund’s metal or a Hund’s assisted relativistic Mott insulator. Our observation could find a broad application and relevance to many correlated materials with multiple orbitals.

Correlated quantum materials provide a rich platform to explore different competing interactions. The simplest one would be the competition between the electron kinetic energy and the Coulomb interactions between the electrons. This is captured by the well-known Hubbard model [1]. For the integer electron filling per site, a strong on-site electron interaction would directly convert the system from a metal into a Mott insulator with the formation of local moments [2–4]. This “big” parent picture is decorated in many different ways when extra interactions are included or emerge as the subleading effects. This includes, for example, the residual interactions and the magnetic ground states of the Mott insulators, the nature of the metallic bands, the band structure topology [5, 6], the nature of Mott transition [7–9], the orbital selectivity of Mott transition [10–12], etc. Two interesting decorations, Hund’s metal [13, 14] and relativistic Mott insulator [15, 16] that are discussed in this work, are from the Hund’s coupling and from the spin-orbit coupling, respectively. They are two interesting ideas that emerge in the theory of correlated electron materials over the past decade.

The Hund’s metal is concerned with how the correlated metallic regime is enhanced by the presence of the Hund’s coupling on multiply degenerate $d$-orbitals for many transition metal compounds [13, 14]. Most often, it was argued that, the Hund’s coupling effectively reduces the electron correlation by harnessing the interaction energy gain in the spin sector. Thus, the metallic regime is significantly expanded compared to the case without the Hund’s coupling. The relativistic Mott insulator is a concept about the role of strong spin-orbit coupling in correlated materials with multiple $4d/5d$ orbitals or bands [15]. It can sometimes be relevant for systems with $3d$ electrons when the spin-orbit coupling becomes active. The strong spin-orbit coupling twists the electron motions as it hops on the lattice and reduces the bandwidth of the electrons. Another description of this effect is that, the spin-orbit coupling breaks the whole bands into multiple spin-orbital-entangled subbands with much narrower bandwidths. As a result, the electron correlation is enhanced. The system becomes a spin-orbit-coupled Mott insulator. If there is no spin-orbit coupling, the system would be a correlated metal. It is the spin-orbit coupling that assists the electron correlation and drives the Mott insulating behaviors. As the spin-orbit coupling is a relativistic effect, the spin-orbit-coupled Mott insulator is often referred as the relativistic Mott insulator.

Having explained the underlying ideas of Hund’s metal and relativistic Mott insulator, one would immediately realize that, both of these two concepts are dealing with the correlated materials with multiple orbitals, but they have rather opposite tendencies. Thus, the correlated materials would fall into a dilemma between Hund’s metal and relativistic Mott insulator. The purpose of this work is to point out this dilemma through a specific example. The specific example in this work is simply adopted to explain the universal physics behind, and should not be interpreted as a localized specifics without any generalization. One should really extract the general message delivered by our specific example. We further design a calculation formalism to study the spin-orbit coupling, the Hund’s coupling and Mott physics in correlated materials, and hope to capture these competing effects at least in a crude manner.

We start with an extended Hubbard model with multiple orbitals and the two-fold degenerate $t_{2g}$ orbitals.

![FIG. 1. (a) The hopping between the $t_{2g}$ orbitals from neighboring sites on the square lattice. (b) The energy splitting of three-fold degenerate $t_{2g}$ orbitals and the two-fold degenerate $e_g$ orbitals.](image-url)
orbitals on a square lattice (see Fig. 1). We consider the octa-
hedral crystal field environment for the transition metal ions.
The two-fold degenerate $e_g$ orbitals are higher in the energy
than the three-fold degenerate $t_{2g}$ orbitals. We assume that,
the crystal field separation $\Delta$ between the $e_g$ and the $t_{2g}$ or-
bitals is very high so that we can safely neglect the presence of
the upper $e_g$ orbitals. We restrict ourselves to the $t_{2g}$ orbitals.
This is sufficient for revealing the physics that was previously
advocated. The extended Hubbard model on the $t_{2g}$ manifold is
written as

$$
\mathcal{H} = \sum_{\langle ij \rangle} \sum_{m,n} \sum_{\alpha} \sum_{\mu} \xi_{im}^{\alpha} c_{m\alpha}^{\dagger} c_{j\alpha} + \sum_{i} \sum_{m,n} \sum_{\alpha} \frac{\lambda}{2} c_{m\alpha^{\dagger}} c_{m\alpha} c_{n\alpha} c_{n\alpha^{\dagger}} + \sum_{i} U \sum_{m} c_{im}^{\dagger} c_{im} c_{im}^{\dagger} c_{im} + \sum_{i} \left[ U' \sum_{m\neq n} c_{im}^{\dagger} c_{im} c_{im}^{\dagger} c_{im} + \frac{J'}{2} \sum_{m\neq n} c_{im}^{\dagger} c_{im} c_{im}^{\dagger} c_{im} \right] + \cdots,
$$

where $m, n = 1, 2, 3$ are the orbital indices corresponding to
the $yz, xz, xy$ orbitals for the $t_{2g}$ orbitals, $\alpha, \beta = \uparrow, \downarrow$ are the spin
indices, $\mu = x, y, z$ refers to the component for the spin
and orbital angular momenta, and the spin indices are automatic-
lly summed. The operator $c_{im\alpha}^{\dagger}$ ($c_{im\alpha}$) creates (annihilates)
an electron on the $m$ orbital with the spin quantum $\alpha$. On
the first line of Eq. (1), the first term describes the electron
hopping between different orbitals and the neighboring sites
on the lattice, the second term describes the atomic spin-orbit
coupling, and the third term is the intra-orbital Coulomb in-
teraction. On the second line of Eq. (1), the first term is the
inter-orbital interaction, the second term is the Hund’s cou-
pling, the third term describes the electron pair hopping, and
“…” refers to the extra interactions and effects that are not
considered here. Owing to the lattice symmetries and the or-
bital orientations, only the nearest-neighbor intra-orbital hop-
ping is non-vanishing and is set to $t$ (see Fig. 1). The inter-
actions in Eq. (1) are the standard Kanamori interactions. We
will take the atomic limit with $J = J'$ and $U' = U - 2J$ in our
calculation, and the interactions can then be described as $U-
interaction and $J$-interactions. This Hamiltonian is par-
cularly relevant for e.g. V$^{4+}$ ions with 3$d^1$ electron config-
urations in Sr$_2$VO$_4$ [17–20], Mo$^{4+}$ ions with 4$d^2$ electron config-
urations in Sr$_2$MoO$_4$ [21], Re$^{4+}$ ions with 5$d^1$ electron config-
urations in Sr$_2$ReO$_4$, Ru$^{4+}$ ions with 4$d^5$ electron config-
urations in Sr$_2$RuO$_4$ [22] and Cs$_2$RuO$_4$ [23–25], even Ir$^{4+}$ ion
with 5$d^5$ electron configurations in Sr$_2$IrO$_4$ after twisting the
hoppings [26], and other transition metal compounds. In
this work, we analyze the $d^2$ and $d^5$ configurations for the illus-
tration.

Both the Hund’s metal and the relativistic Mott insulator
are more or less concerned with the Mott transition. To tackle
with the Mott transition of Eq. (1), we implement a slave-rotor
formulation of the electron operator [27], express the electron
operator as $C_{im\alpha} = e^{-i\phi} f_{im\alpha}$, then decouple the extended Hub-
bard model into the spin sector and the charge sector. This ap-
proximation automatically assumes the spin-charge separation
by placing the electron charge on the bosonic rotor and placing
the spin and the orbital on the fermionic spinor. Often, this
approach was used to describe the quantum spin liquid that is
proximate to Mott transition [28]. Magnetic orders can be in-
corporated into this formalism by including the $J$-interactions
onto the spinon sector and properly decoupling the interaction
according to the orderings [29]. The latter approach attributes
the Mott localization to the $U$-interaction, and the magnetic
orders to the $J$-interactions, even though both interactions to-
gether give rise to the magnetism in many cases. Here we are
not interested in addressing the nature of the ground state for
the Mott regime, but to understand the variation of the Mott
transition. Thus, we simply assume the Mott side is a spin
liquid and explore the fate of Mott transition in the presence
of extra couplings.

For our purpose, we first take away the $J$-interactions, and
decouple the extended Hubbard model into the spin sector
$\mathcal{H}_f$ and the the charge sector $\mathcal{H}_0$ with

$$
\mathcal{H}_f = \sum_{\langle ij \rangle} \sum_{m,n} \sum_{\alpha} \sum_{\mu} \xi_{ij}^{mn} f_{im\alpha}^{\dagger} f_{jn\alpha} + \sum_{i} \sum_{m,n} \sum_{\alpha} \frac{\lambda}{2} f_{im\alpha}^{\dagger} c_{im\alpha} c_{im\alpha}^{\dagger} f_{im\alpha} + \sum_{i} h_i f_{im\alpha}^{\dagger} f_{im\alpha},
$$

$$
\mathcal{H}_0 = \sum_{\langle ij \rangle} \langle \chi_{ij} e^{i\sum_{\alpha} \phi_{\alpha} f_{im\alpha}^{\dagger} f_{jm\alpha}} + h.c. \rangle + \sum_{i} \left( \frac{U}{2} \sum_{\alpha} l_{\alpha}^{2} + h \sum_{\alpha} l_{\alpha} \right),
$$

where $\xi_{ij}^{mn} = \langle e^{i\sum_{\alpha} \phi_{\alpha} f_{im\alpha}^{\dagger} f_{jm\alpha}} \rangle$, $h_i$ is a La-
grangian multiplier for each site to enforce the Hilbert space
constraint such that $\sum_{m,n} \sum_{\alpha} f_{im\alpha}^{\dagger} f_{im\alpha} = \bar{n} = \bar{L}_{\alpha}$, and $\bar{L}_{\alpha}$ is an
angular momentum variable conjugate to the U(1) phase $\theta$. Here
$\bar{n}$ is the electron occupation number per site, and $\bar{n} = 2
(3)$ for the $d^2$ ($d^3$) electron configuration. As the translation
symmetry is preserved throughout, one expects that $h_i$ has no
site-dependence and $h_i \equiv h$. Because $\langle \sum_{\alpha} l_{\alpha} \rangle = 0$, so we ex-
pect $h = 0$ in the self-consistent mean-field calculation. More-
over, due to the translation symmetry and the lattice rotation,
$\chi_{ij} \equiv \chi$. The charge sector model, $\mathcal{H}_0$, behaves more like a
boson Hubbard model at integer fillings, and we solve it with
a coherent state path integral formulation. After integrating
out the angular momentum $\bar{L}$, we obtain

$$
Z_0 = \int D\Phi D\Phi'
\exp\left[-\frac{1}{2} \int d\tau \sum_{i} \left( \frac{\partial_{\alpha} \Phi_{\alpha}^{\dagger}}{2U} + \langle \chi_{ij}^{\dagger} \Phi_{j}^{\dagger} + h.c. \rangle \right) \right].
$$
where we have replaced $e^{-i\Phi}$ with $\Phi$, and $|\Phi| = 1$. The unimodular condition on $\Phi$ can be imposed by introducing a Lagrangian multiplier, $\Lambda$. The excitation spectrum of charge boson $\Phi$ can be solved via a saddle point approximation and the uniformity requirement with $\Lambda = \Lambda$. We find that the $\Phi$ spectrum is given by $\Omega_{\Phi} = \sqrt{2U/\Lambda - 2|\phi|(|\cos k_x + \cos k_y|)}$ where the lattice constant is set to unity. When the gap of the spectrum vanishes, the charge boson $\Phi$ is condensed and the system goes from the Mott insulator to the metallic state. We find that the Mott localization occurs at $|U/\chi| = 4.84$. To obtain the actual critical $U/t$ for the Mott transition, one further requires the knowledge from the spinon sector to produce the parameter $\chi$, and this $\chi$ parameter depends on $\langle e^{i\theta} - e^{-i\theta} \rangle$ at the Mott transition. The latter quantity can then be directly computed from Eq. (4), and we find that $\langle e^{i\theta} - e^{-i\theta} \rangle = \sum_k \Theta_{\chi} / (8\chi N)$ for the nearest-neighbor bonds at the Mott transition where $N$ is the number of lattice sites. The parameter $\chi$ can be evaluated from solving the spinon Hamiltonian $\mathcal{H}_F$ with

$$\chi = \sum_{m,n} \sum_{i,j} \langle f^\dagger_{i\alpha} f_{j\beta} \rangle \sum_{m=2,3} \langle f^\dagger_{km} f_{km\alpha} \rangle \cos k_x$$

$$= -\frac{1}{N} \sum_k \sum_{m=2,3} |M(k)_{m,n}|^2 \Theta_\epsilon \cos k_x. \quad (5)$$

Here using the translation symmetry, we only need to consider the $\hat{x}$ direction bond. The spinon Hamiltonian is diagonalized by the canonical transformation $f_{km\alpha} = M(k)_{m,n\alpha} d_{n\alpha}$, where $d_{n\alpha}$ is the spinon eigenmode and the energy is given by $\epsilon_{n\alpha}(k)$ with the spinon Fermi energy $\epsilon_F$. For each $\lambda/t$, there is a corresponding $\chi$ parameter from the spinon sector, and thus a corresponding $(U/t)_c$ for the Mott transition. Thereby, we are able to construct the phase diagram in the $U/(t-\lambda)/t$ plane.

In Fig. 2(a) and Fig. 2(b), we depict the phase diagram for the $d^2$ and $d^3$ electron configurations, respectively. It is shown that, the critical Hubbard $U$-interaction for the Mott transition is gradually suppressed as the spin-orbit coupling is increased. It turns out out, both $d^2$ and $d^3$ fillings have qualitatively similar phase diagrams as expected. The spin-orbit coupling suppresses the electron bandwidth, and a weaker $U$-interaction would already drive the Mott transition [15].

On the other hand, the Hubbard $U$-interaction suppresses the bandwidth, which then enhances the effect of the spin-orbit coupling. These two interpretations provide a physical understanding of the phase diagram. In the right region of the Mott insulating phases, the system should be more appropriately quoted as a relativistic Mott insulator to reflect the strong spin-orbit coupling. Likewise, In the right region of the metallic phase, the system is better to be quoted as a spin-orbit-coupled metal [30].

We now include the effect of the $J$-interactions. As we have previously explained, the proper treatment of the $J$-interactions requires the knowledge of the ground state on the Mott side. We do not intended to address the actual ground state on the Mott side, and thus, we tend to consider the $J$-interactions in a qualitative manner. In the strong Mott regime with decoupled atoms, things can be understood in both qualitatively and quantitatively. It is shown that [13, 14], the renormalized effective correlation $U_r$ can be obtained by calculating the energy cost for changing the valence charge of two neighboring ions from their original electron occupation, i.e. transferring one electron from one site to the other. When the electron occupation is not at half-filling (i.e. not occupying each orbital with one electron), the system can gain energies from the Hund’s coupling and the inter-orbital interaction, and the renormalized correlation is $U_r = U - 3J$. When the electron occupation is at half-filling, transferring electrons would automatically introduce the double electron occupation on a single orbital and thus increase the correlation energy. The renormalized correlation in this case is $U_r = U + 2J$ for our $d^2$ configuration. In Fig. 2(c) and Fig. 2(d), we depict the new phase diagrams after taking into account the renormalized correlation. In Fig. 2(c) for the $d^2$ configuration, the effective correlation is reduced by the Hund’s coupling, and thus a large region that was insulating in Fig. 2(a) becomes metallic. This region is nothing but Hund’s metal. On the right part inside this region, as the strong spin-orbit coupling is involved, it should be quoted as a spin-orbit-coupled Hund’s metal. It turns out that, the $5d$ compound BaOsO$_3$ was recently proposed as a spin-orbit-coupled Hund’s metal [31].
coupling first, then one arrives with a orbit coupling persists even in the strong Mott regime. For the assisted relativistic Mott insulator”. The interplay between the Hund’s coupling and the spin-orbit coupling persists even in the strong Mott regime. For the assisted relativistic Mott insulator. This is a Hund’s assisted Mott insulator.

The coupling have opposite effects on the electron correlation for the electron occupation off from the half filling of all orbitals. At the half filling, the spin-orbit coupling and the Hund’s coupling are found to enhance the electron correlation. In real materials, often the spin-orbit coupling in the systems with heavy ions should be seriously considered, and the Hund’s coupling is unavoidable almost for any material with electron correlations.

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Fig. 2(d), for the $d^3$ configuration, the effectively correlation is enhanced, and a large metallic region in Fig. 2(b) is converted into Mott insulators. This is a Hund’s assisted Mott insulator, and the right region of it should then be called a “Hund’s assisted relativistic Mott insulator”.

The parent state of the Fe-based superconductors is often a correlated metal. As this metal was interpreted as a Hund’s metal, the resulting superconductor was then proposed as a Hund’s superconductor [36, 37]. The spin-orbit coupling was recently invoked for the Fe-based superconductors [38], and various topological features such as Dirac band touching and majorana physics were proposed [39, 40]. It can be a good chance to include both Hund’s coupling and the spin-orbit coupling together in the future work.

To conclude, the spin-orbit coupling and the Hund’s coupling have opposite effects on the electron correlation for the electron occupation off from the half filling of all orbitals. At the half filling, the spin-orbit coupling and the Hund’s coupling are found to enhance the electron correlation. In real materials, often the spin-orbit coupling in the systems with heavy ions should be seriously considered, and the Hund’s coupling is unavoidable almost for any material with electron correlations.

Discussion.—In our illustrative study of the Hund’s coupling and spin-orbit coupling in the correlated materials, we only considered the $d^2$ and $d^3$ configurations, and the uncorrelated or weakly correlated regimes in our examples are all metallic. In reality, it could happen that the uncorrelated or weakly correlated regime is a band insulator. In that case, the topological aspect of the band structure should be considered. As the spin-orbit coupling is involved, whether the band insulator is a topological insulator or not is an interesting and relevant question. Just like the metallic behavior can be driven by the Hund’s coupling, the candidate topological band insulator if exists is an example of Hund’s topological insulator. Likewise, one could use Hund’s coupling to enlarge the region of other topological matter at the single-particle level.

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