Effect of Hund’s coupling on nonlocal correlations: a cluster DMFT study

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We study the spatial correlation effects in multiorbital systems, especially in a paramagnetic metallic state subject to Hund’s coupling. We apply a cluster extension of the dynamical mean-field theory (DMFT) to the three-orbital Hubbard model away from half filling, where the previous DMFT studies revealed that local correlation effects caused by Hund’s coupling bring about unusual strongly-correlated metallic behaviors. We find that Hund’s coupling significantly affects the nonlocal correlations, too; it strongly modulate the electron distributions in the momentum space so as to make some momentum region almost half-filled and strongly correlated. It leads to an anomalous electronic state distinct both from the Fermi liquid and the Mott insulator. We identify the mechanism with the intersite ferromagnetic correlations induced by Hund’s coupling.

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Introduction. Hund’s coupling is known to play essential roles in various electronic properties of strongly correlated materials such as transition-metal oxides. It aligns the electronic spins at each atomic site to maximize the total spin. In recent years, its substantial role in paramagnetic metals has also become recognized: Haule and Kotliar found, in their study on iron-based superconductors, that it is Hund’s coupling $J$ rather than the Hubbard $U$ that induces the strong correlation effects in these materials \[1\]. They coined a term “Hund’s metal” for this state. For degenerate $N$-orbital systems at integer fillings (except for the filling $n = 1, N$, and $2N - 1$), $J$ has more complicated effects: While $J$ increases the effective mass of quasiparticles for small $U$, it allows the quasiparticles to survive up to a rather large $U$ (“Janus-faced” behavior). This behavior has been discussed in relation with diverse metallic properties of perovskite-type transition-metal oxides \[2, 3\]. More insights were obtained by Werner et al., who observed a frozen local-spin moment in the metallic state of the three-orbital Hubbard model away from half filling (“spin-freezing” behavior) \[4\]. It was also pointed out that Hund’s coupling suppresses orbital correlations (“band decoupler”) and leads to the orbital-selective Mott transition \[3, 6\] in nondegenerate multiorbital models \[3, 8\].

These exotic behaviors have been revealed by the dynamical mean-field theory (DMFT) \[5\] applied to the multiorbital Hubbard models. The DMFT studies show that these behaviors originate from the local correlation effects. However, in real materials, nonlocal correlation effects, which are neglected in the DMFT, may also play essential roles. To study these, cluster extensions of the DMFT (cDMFT) \[10\] have been developed. In fact, the cDMFT applied to the single-orbital Hubbard model has unveiled significant effects of the spatial correlations, which induce the differentiation of the electronic structure in the momentum space, especially in the vicinity of the Mott metal-insulator transition \[10\].

Nevertheless, the effect of Hund’s coupling on the spatial correlations is an open issue. A notable exception is the work by Kita et al., who found an anomalous metal with pseudogap in a two-orbital system by means of the cDMFT combined with the non-crossing approximation \[11\]. However, a study with a more serious treatment of the correlation effects, as well as a study on the three-orbital systems which are relevant to many real transition-metal oxides, are still missing. This is because the cDMFT (with numerically exact solvers) has been computationally too expensive to apply to the multiorbital models. Especially, if one employs the quantum Monte Carlo (QMC) method to solve the multiorbital impurity problem of the cDMFT, it has been a challenge to preserve the spin-rotational symmetry. Recently, we developed an efficient sample-update algorithm \[12\] for the continuous-time QMC (CT-QMC) method \[13\] based on the interaction expansion \[14, 15\]. The development enabled us the cDMFT calculations for the multiorbital Hubbard models within a reasonable computational time \[12\].

In this Letter, we investigate the spatial correlation effects in the presence of Hund’s coupling for the degenerate three-orbital Hubbard model at the electron filling $n = 2$, where the previous single-site DMFT studies predicted the exotic behaviors (such as the "Janus-faced" behavior \[3\] and the “spin-freezing” behavior \[4\]) induced by Hund’s coupling. We find that Hund’s coupling induces a drastic filling rearrangement in the momentum space. It occurs so as to fix some momentum region at nearly half filling, and thereby makes the the region conspicuously strongly correlated, leading to an anomalous metal. We elucidate that this is caused by an intersite ferromagnetic correlations triggered by the interplay be-
patches for transition oxides with simple model for, for example, layered transition-metal between Hund’s coupling and the electron transfers.

Model and method. We study the degenerate three-orbital Hubbard model on a square lattice, which is a simple model for, for example, layered transition-metal oxides with $t_{2g}$ electrons at the Fermi level. The Hamiltonian is given by

$$
\mathcal{H} = -t \sum_{\langle i, j \rangle, \sigma} \hat{e}_{i \sigma}^\dagger \hat{e}_{j \sigma} + \mu \sum_i \hat{n}_{i \uparrow} + U \sum_i \hat{n}_{i \uparrow} \hat{n}_{i \downarrow} + U' \sum_{i, \langle l, m \rangle, \sigma} \hat{n}_{i \uparrow} \hat{n}_{l \sigma} \hat{n}_{l \sigma} \hat{n}_{m \sigma} + J \sum_{i, \langle l, m \rangle, \sigma} \hat{c}_{i \sigma}^\dagger \hat{c}_{l \uparrow} \hat{c}_{l \downarrow} \hat{c}_{m \sigma}^\dagger + J \sum_{i, \langle l, m \rangle, \sigma} \hat{c}_{i \sigma}^\dagger \hat{c}_{m \sigma} \hat{c}_{l \uparrow} \hat{c}_{l \downarrow}, \tag{1}
$$

where $\hat{e}_{i \sigma}^\dagger$ ($\hat{e}_{i \sigma}$) creates (annihilates) an $\ell$th-orbital electron ($\ell = 1-3$) with spin $\sigma$ at site $i$ and $\hat{n}_{i \sigma} = \hat{c}_{i \sigma}^\dagger \hat{c}_{i \sigma}$. $t$ is a transfer integral to the neighboring sites, denoted by $\langle i, j \rangle$. $\mu$ is the chemical potential. For interaction parameters, we introduce intraorbital Coulomb repulsion $U$, interorbital Coulomb repulsion $U'$, and Hund’s coupling $J$ with the relation $U' = U - 2J$. The spin-exchange and pair-hopping terms (3rd line in Eq. 1) are necessary to keep the spin-SU(2) symmetry. Hereafter, we use $t = 1$ as a unit of energy. The electron density per site is set to be $n = 2$ (i.e., two electrons per site).

We analyze the model 1 with the dynamical cluster approximation (DCA) version of the cDMFT, 10,17,18 where we employ two momentum patches illustrated in Fig. 1(a). We call inner [outer] patch (0, 0) [$(\pi, \pi)$] patch. The DCA approximates the self-energy to be constant in the each patch; we call the one for (0, 0) [$(\pi, \pi)$] patch $\Sigma_{00}$ [$\Sigma_{\pi\pi}$]. Short-range spatial correlations are incorporated as the difference between $\Sigma_{00}$ and $\Sigma_{\pi\pi}$. The correlation effects are calculated through mapping the original lattice model 1 onto a two-site impurity model embedded in a self-consistently determined bath sites. The most time-consuming part of the computation is to solve the two-site impurity model, for which we employ an improved CTQMC method developed in Ref. 12. We restrict ourselves to paramagnetic and paraorbital solutions 19. The temperature $T$ is set to be $0.1t$ and correspondingly the inverse temperature $\beta = 10$. We mainly consider the case of $J = U/4$ and, just for comparison, we also show the results for $J = 0$.

Results and discussions. In Fig. 1(a), we show the Fermi surface for $n = 2$ in the non-interacting limit ($U = 0$). The corresponding non-interacting density of states and the Fermi level are shown in Fig. 1(b). We see that at $U = 0$, all the electrons reside in the (0, 0) patch and the $(\pi, \pi)$ patch is completely empty. On the other hand, in the atomic limit ($U \to \infty$), the fillings for $(0, 0)$ and $(\pi, \pi)$ patches should be the same because the momentum dependence vanishes. Although these two limits are trivial, it is highly nontrivial how they are connected as the interaction $U$ varies.

In Figs. 1(c-e), we show, in the following cases, how the filling of each orbital evolves as the Hubbard $U$ increases: (c) $J = U/4$ with [without] the spin-flip and pair-hopping terms, and (e) $J = 0$. In all the cases, the filling $n_{00}$ [$n_{\pi\pi}$] for $(0, 0)$ [$(\pi, \pi)$] patch is 2/3 [0] at $U = 0$. As the Hubbard $U$ [and accordingly Hund’s coupling $J$ in (c) and (d)] increases, $n_{00}$ [$n_{\pi\pi}$] gradually decreases (increases) in all the cases. However, there is a remarkable difference between $J > 0$ and $J = 0$ cases: For $J > 0$ the fillings show a “plateau” (where the filling has little dependence on $U$) for a range of large interactions, which is colored by green in the figures, while there is no such plateau for $J = 0$. In the plateau region, $n_{00} \sim 0.5$, i.e., the (0, 0) patch is nearly half filled. Comparing Figs. 1(c) and (d), we find that the spin-flip and pair-hopping terms give a quantitatively large difference in the results: the density-type Hamiltonian [Fig. 1(d)] drastically overestimates the tendency toward the filling plateau. In the following, we concentrate on the spin-SU(2)-symmetric Hamiltonian and elucidate the physical
Although ImΣ at the first Matsubara frequency ω0 = πT, for (0,0) and (π,π) patches. The panels (a) and (c) [(b) and (d)] show the results for J = U/4 [J = 0].

FIG. 2: (Color online) (a),(b) Green’s functions at the imaginary time τ = β/2, G_K(β/2), and (c),(d) the imaginary parts of the self-energies, ImΣ_K(ω0), at the first Matsubara frequency ω0 = πT, for (0,0) and (π,π) patches. The panels (a) and (c) [(b) and (d)] show the results for J = U/4 [J = 0].

origin of the plateau behavior.

Since the present two-patch DCA does not have enough resolution in the momentum space, we avoid to discuss the shape of the Fermi surface. Instead, we plot in Figs. 2(a) and 2(b) Green’s function of each patch at the imaginary time τ = β/2, G_K(β/2), as a measure of the low-energy spectral weight. In the non-interacting limit (U = 0), G_00(β/2) is finite and G_ππ(β/2) ∼ 0, as the whole Fermi surface is encompassed in the (0,0) patch [Fig. 1(a)]. As U increases, these low-energy spectral weights evolve differently depending on J. In J = 0 case [Fig. 2(b)], G_ππ(β/2) is always nearly zero up to U = 16 while G_00(β/2) starts from the finite value at U = 0, gradually decreases as U increases, and eventually ends up with a very small value at around U = 12. According to the behavior of G_00(β/2), -ImΣ_00(ω0), which approximates the scattering rate, shows a rapid increase at around U = 12 [Fig. 2(d)]. These results show that the (π,π) patch does never acquire a low-energy spectral weight and that the whole system transits from a Fermi-liquid-like metal to the Mott insulator at a strong interaction (U ∼ 12).

A remarkable difference is seen in the result for J = U/4 [Fig. 2(a)]. In this case, G_ππ(β/2) acquires a finite weight in a region of U ∼ 6, where G_00(β/2) loses its weight. For U ≥ 12, G_ππ(β/2) dominates the low-energy weight and G_00(β/2) seems to vanish, indicating an anomalous electronic state distinct from both the Fermi liquid and the Mott insulator. As seen in Fig. 2(c), -ImΣ_00(ω0) remains a moderate value (∼ 1) in this region. Interestingly, this new state of matter emerges in the same parameter region as that of the filling plateau [Fig. 1(a)], as we have indicated by green color. Although a similar Hund-induced enhancement of the critical U can also be seen in the single-site DMFT results [3], the exotic behavior seen in our result crucially relies on the momentum differentiation, which is neglected in the single-site DMFT.

![FIG. 3: (Color online) (a) The onsite spin-spin correlation function \( \langle S^z_i (\tau) S^z_i (0) \rangle \) and orbital-orbital correlation function \( \langle \hat{n}_{li}(\tau) \hat{n}_{mi}(0) \rangle \) for J = U/4. The spin-spin and orbital-orbital correlation functions take positive and negative values, respectively. (b),(c) The intersite spin-spin correlation function \( \langle \hat{S}^z_i \hat{S}^z_j \rangle \), intraorbital density-density correlation function \( \langle \hat{n}_{li}(\tau) \hat{n}_{ij}(0) \rangle /4 \), and interorbital density-density correlation function \( \langle \hat{n}_{li}(\tau) \hat{n}_{mj}(0) \rangle /4 \) for J = U/4 and J = 0, respectively.](image-url)
tion functions; the spin-spin channel, $\langle \hat{S}_i^z \hat{S}_j^z \rangle$, intraorbital density-density channel, $\langle \hat{n}_{li} \hat{n}_{lj} \rangle / 4$, and interorbital density-density channel, $\langle \hat{n}_{li} \hat{n}_{mj} \rangle / 4$. In the absence of Hund’s coupling ($J = 0$), the intersite spin-spin correlation is always antiferromagnetic (negative) in the range from $U = 0$ to $U = 16$. However, for $J = U/4$, the intersite spin-spin correlation changes from antiferromagnetic to ferromagnetic around $U = 10$. The ferromagnetic region agrees well with the region of filling plateau in Fig. 1(c). Thus, in the filling-plateau region, $(0,0)$ patch loses [gains] the low-energy spectral weight, and the intersite spin-spin correlation becomes ferromagnetic.

In order to understand the relationship between these behaviors, we consider spin-orbital configurations of four electrons in the two-site cluster (with three orbitals at each site) in two different limiting cases. First, in the non-interacting limit ($U = 0$), the three orbitals at each site form bonding and antibonding orbitals in the two-site cluster. The bonding and antibonding orbitals correspond to $(0,0)$ and $(\pi, \pi)$ components, respectively [20]. The electron hopping between the two sites lifts the degeneracy of the bonding and antibonding orbitals. This energy offset can be regarded as an effective crystal-field splitting between the molecular orbitals. Then, in the ground state all the four electrons occupy the bonding orbitals (Fig. 4 small $U$).

As a result, the high-spin state is realized in $(0,0)$ patch and the patch becomes almost half-filled. Then, $(0,0)$ patch would develop strong correlations, as is indeed seen as the loss of the low-energy spectral weight and the enhancement of the scattering rate in Fig. 2(a). The formation of the high-spin state makes it difficult to change the filling, explaining the filling plateau of the $(0,0)$ patch seen in Fig. 1(c).

**Conclusion.** We have explored spatial correlation effects in multiorbital systems by means of the two-patch DCA. In the degenerate three-orbital Hubbard model at $n = 2$, we have found that Hund’s coupling, in combination with the electron hopping, brings about a drastic rearrangement in the momentum distribution of the electron density. In particular, the $(0,0)$ patch is almost fixed at half filling and becomes strongly correlated. There the low-energy spectral weight emerges in the $(\pi, \pi)$ sector, indicating that a new state of matter appears in between the Fermi-liquid metal and the Mott insulator. By calculating the two-particle correlation functions, we have identified the mechanism of this anomalous behavior with the intersite ferromagnetic correlations enhanced by Hund’s coupling. We have found similar behaviors also in the two-orbital Hubbard model at the filling $n = 1.5$, which suggests that these exotic behaviors are generic in multiorbital systems away from half filling.

Although larger clusters are not tractable at this moment, we expect that once some patches become high-spin state and almost half-filled, it leads to a filling rearrangement in the momentum space, and thereby cause a momentum differentiation. Especially, the high-spin state is realized in a patch which includes the fermi surface, it might lead to the loss of its low-energy spectral weight. The behaviors may be relevant to the pseudogap behaviors recently observed in several multiorbital materials: nickelates [26], irridates [27], and the iron-based superconductors [28 32]. It will be interesting to study these materials, by combining the present scheme with realistic band-structure calculations.

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Since we consider the paramagnetic solution, the configuration with three down-spin electrons in the bonding orbitals and one down-spin electron in anti-bonding orbitals is equally realized in the Monte Carlo sampling. Note that configurations in which two or more electrons occupy the anti-bonding orbitals also have a small weight.