Spontaneously orbital-selective superconductivity in a three-orbital Hubbard model

Kosuke Ishigaki,1 Joji Nasu,1 Akihisa Koga,1 Shintaro Hoshino,2 and Philipp Werner3

1Department of Physics, Tokyo Institute of Technology, Meguro, Tokyo 152-8551, Japan
2Department of Physics, Saitama University, Saitama 338-8570, Japan
3Department of Physics, University of Fribourg, 1700 Fribourg, Switzerland

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We study a three-orbital Hubbard model with negative Hund coupling in infinite dimensions, combining dynamical mean-field theory with continuous time quantum Monte Carlo simulations. This model, which is relevant for the description of alkali-doped fullerides, has previously been shown to exhibit a spontaneous orbital selective Mott phase in the vicinity of the superconducting phase. Calculating the pair potential and double occupancy in each orbital, we study the competition between different homogeneous ordered states and determine the corresponding finite temperature phase diagram of the model. We identify two distinct types of spontaneous orbital-selective Mott states and show that an orbital-selective s-wave superconducting state with one superconducting and two metallic orbitals is spontaneously realized between the conventional s-wave superconducting phase and these two kinds of spontaneously orbital-selective Mott states.

Orbital degrees of freedom and their dynamics are known to play an essential role in strongly correlated electron systems as they couple to other degrees of freedom of the lattice system. This can lead to exotic phenomena such as colossal magnetoresistance in the manganites [1], and unconventional superconductivity in ruthenates [2] or iron pnictides [3]. An interesting phenomenon in this general context is the orbital-selective Mott (OSM) transition [4], which has been discussed in transition metal oxides such as Ca$_2$-SrRuO$_4$ [5, 6] and La$_{n+1}$Ni$_n$O$_{3n+1}$ [7–9]. The OSM transition results in a distinct electronic character of different orbitals, i.e., some orbitals are itinerant while the others are localized. This physics has been explored in simple two-orbital Hubbard models with different bandwidths [10–14] or crystal field splitting [15], where the difference of the effective Coulomb interaction or local energy induces the OSM state.

As electron systems have degenerate bands, it is essential to consider the degeneracy of orbitals. In many systems, such as the fullerene-based solids, the degeneracy of orbitals is lifted due to the presence of antiferromagnetic Hund coupling. This antiferromagnetic Hund coupling induces the OSM state and the corresponding finite temperature phase diagram of the model. We identify two distinct types of spontaneous orbital-selective Mott states and show that an orbital-selective s-wave superconducting state with one superconducting and two metallic orbitals is spontaneously realized between the conventional s-wave superconducting phase and these two kinds of spontaneously orbital-selective Mott states.

In this Letter, we study the three-orbital Hubbard model with antiferromagnetic Hund coupling at half filling, combining dynamical mean-field theory (DMFT) [18–20] with continuous-time quantum Monte Carlo (CTQMC) simulations [21, 22]. Calculating pair potentials and double occupancies, we clarify that at low temperatures, an s-wave SC state without orbital symmetry breaking is stabilized rather than the SOSM state. At higher temperatures, we find a new SOSM state where two orbitals are metallic while the third is in a paired Mott state. Most remarkably, we demonstrate that this SOSM phase transforms into a spontaneous orbital-selective superconducting (SOSSC) phase in the vicinity of the SC dome.

We consider the half-filled three-orbital Hubbard model described by the Hamiltonian

\[ \mathcal{H} = -t \sum_{\langle i,j \rangle_{\alpha \sigma}} c_{i \alpha \sigma}^\dagger c_{j \alpha \sigma} + U \sum_{i \alpha} n_{i \alpha \uparrow} n_{i \alpha \downarrow} + U' \sum_{i \alpha \alpha'} n_{i \alpha \sigma} n_{i \alpha' \bar{\sigma}} + (U' - J) \sum_{i \alpha \alpha'} n_{i \alpha \sigma} n_{i \alpha' \bar{\sigma}}, \]

where \( c_{i \alpha \sigma} \) is an annihilation operator for an electron with spin \( \sigma (\uparrow, \downarrow) \) and orbital index \( \alpha (=1, 2, 3) \) at the \( i \)th site and \( n_{i \alpha \sigma} = c_{i \alpha \sigma}^\dagger c_{i \alpha \sigma} \). \( t \) is the transfer integral between nearest neighbor sites, \( U \) (\( U' \)) is the intraband (interband) Coulomb interaction and \( J \) is the Hund coupling. We assume the relation \( U = U' + 2J \) and neglect the exchange part of the Hund coupling and the pair hopping for simplicity. The effects of these interactions are discussed later. In the present calculations, we fix the Hund coupling as \( J/U = -1/4 \), which is large compared to ab-initio estimates [23] but allows us to reveal the relevant physics at moderate computational expense. An important point is the negative sign of the (anti-ferromagnetic) coupling, which is characteristic of fullerene-based solids [24–26]. This coupling disfavors singly occupied orbitals since the interorbital Coulomb interaction dominates \( (U' > U) \). At half-filling, the intraorbital Coulomb interaction can be regarded as effectively attractive in the weak coupling region. On the other hand, in the strongly interacting
half-filled case, empty, singly and doubly occupied orbitals are realized on a given site and hence large orbital fluctuations are expected in the system. This is in stark contrast to the ferromagnetic Hund coupling case with \( U > U' \), where each orbital wants to be singly occupied and orbital fluctuations are suppressed. Therefore, in our model with antiferromagnetic Hund coupling \( (J < 0) \), interesting orbital-selective states may emerge due to orbital fluctuations.

In the present study, we mainly make use of DMFT. In this approach, the lattice model is mapped to an effective impurity problem, where local electron correlations can be taken into account precisely. The Hubbard model with degenerate orbitals has been extensively discussed in the framework of DMFT, and interesting phenomena have successfully been clarified such as the Mott transition [27–34], orbital-selective Mott transitions [10–15, 35], magnetism [36–38], and superconductivity [39, 40]. In the present study, we focus on the half-filled model and neglect translational symmetry breaking phases such as charge density waves, antiferromagnetically or antiferroorbitally ordered states. This assumption is justified in a system with next-nearest-neighbor hopping \( t' \) comparable to the nearest-neighbor hopping \( t \) [41], which should be relevant for fcc-type fullerene-based compounds.

To examine the competition of the SC and SOSM states at low temperatures, we calculate the pair potential in the \( \alpha \)th orbital \( \psi_\alpha = |\langle c^\dagger_{i\alpha}c_{i\alpha}\rangle| \) as an order parameter of the SC state. In contrast, the order parameter for the SOSM states is not obvious since no difference in the average orbital occupations appears [16]. Here, we calculate the double occupancy for orbital \( \alpha \), \( d_\alpha = \langle n_{i\alpha}n_{i\alpha}\rangle \), and characterize the SOSM state by the appearance of orbital-dependent double occupancies. In the following, we set the unit of energy to the half-bandwidth \( D \).

Figure 1 plots the pair potential and double occupancy for each orbital at \( T/D = 0.01 \). In the noninteracting case \( (U = 0) \), a metallic state is realized with \( d_\alpha = 1/4 \) and \( \psi_1 = 0 \) for all orbitals. Turning on the interaction \( U \) slightly increases the double occupancy since the onsite interaction in this half-filled system is effectively attractive due to the antiferromagnetic Hund coupling. This interaction is expected to enhance pair correlations, and indeed a second-order phase transition occurs to the \( \psi \)-wave SC state with finite \( \psi_1 = \psi_2 = \psi_3 \) around \( U/D \sim 0.87 \), as shown in Fig. 1(b).

A further increase of the Coulomb interaction leads to a maximum in the pair potentials near \( U/D \approx 1.4 \). Around \( U/D \sim 2.0 \), the physical quantities jump and a first-order phase transition occurs, as shown in Figs. 1(a) and 1(b). In the state with \( 2.0 \leq U/D \leq 2.8 \), the pair potentials vanish, while the double occupancies take two distinct values \( d_1 < d_2 = d_3 \). This means that a metallic state is realized in orbital 1. In contrast, a paired Mott state with \( d \sim 0.5 \) is realized in orbitals 2 and 3, which are dominated by empty and doubly occupied configurations. From these observations, we can conclude that an SOSM state is indeed realized in this region [16]. A similar orbital symmetry breaking has also been identified in the two-dimensional system [42]. Beyond \( U/D \sim 2.8 \), the orbital-selective features disappear, and the double occupancies exhibit an orbital-independent value. In the strong coupling region, three electrons are localized at each site in a configuration with empty, singly, and doubly occupied orbitals, and a Mott state is realized with \( d_\alpha \sim 1/3 \).

These results confirm that the SOSM state with one orbital itinerant and two orbitals localized competes with the \( \psi \)-wave SC and Mott states at low temperatures. On the other hand, it is naively expected that another SOSM state with two orbitals itinerant and one orbital paired may also exist in the present system although such a state has not been previously discussed [16]. To clarify this, we examine the low temperature properties systematically, calculating the orbital-dependent quantities \( Z_\alpha = [1 - \text{Im} \Sigma_\alpha(\omega_0)/\omega_0]^{-1} \) and \( A_\alpha = -G_\alpha(1/2T)/\pi T \) as estimates of the renormalization factor and the density of states at Fermi level at finite temperatures [43], where \( \omega_0 = \) the self-energy for the orbital and \( \omega_0 = \pi T \). The results at the temperature \( T/D = 0.02 \) are shown in Figs. 2(a)-(c). In this parameter region, no pair potentials appear. It is found that two kinds of SOSM states are realized between the metallic and Mott states. These can be classified by the number of itinerant unpaired orbitals; the SOSM-1 state is associated with \( n = 1, 2 \) metallic orbital(s), which are schematically shown in Fig. 2(d). When \( 2.2 \leq U/D \leq 2.5 \), \( d_1 < d_2 = d_3 \) and the SOSM-1 state is realized with a metallic orbital 1. On the other hand, in the region \( 2.1 \leq U/D \leq 2.3 \), \( d_1 = d_2 < d_3 \) and the SOSM-2 state is realized with metallic orbitals 1 and 2. The phase transitions between metallic, SOSM-2, SOSM-1, and Mott states should be of first order...
although no hysteresis is visible around $U/D \approx 2.1$.

To clarify the nature of the phase transition, we employ a Landau theory, where the symmetry of the system is taken into account correctly. As discussed above, the orbital-dependent double occupancies $d_o$ are appropriate to characterize the SOSM states [16]. Since the Hamiltonian (1) is invariant under permutations of the three orbitals, the free energy $F$ should be expanded as

$$F = F_0 + a(X^2 + Y^2) + bX(X^2 - 3Y^2) + c(X^2 + Y^2)^2,$$  \hspace{1cm} (2)

where $X = (d_1 + d_2 - 2d_3)/\sqrt{3}, Y = d_1 - d_2$, with constants $F_0, a, b, c (> 0)$. $X$ and $Y$ correspond to the order parameters characteristic of the SOSM states and their forms derive from the $3 \times 3$ Gell-Mann matrices $\lambda_8$ and $\lambda_5$, respectively. The orbital permutation is then represented by the $C_{3\nu}$ symmetry in the $X$-$Y$ plane. This yields the third-order term in the free energy and the phase transition to the SOSM states is of first order (related to the Lifshitz condition).

The nontrivial solutions can be classified into two classes. The solution with $(X, Y) = (-\frac{1}{2}R, -\sqrt{3}/2R)$ ($R > 0$ is a radius), which is equivalent to $(R, 0)$ and $(-\frac{1}{2}R, \sqrt{3}/2R)$ under the $C_{3\nu}$ symmetry, corresponds to the SOSM-1 state with $d_1 < d_2 = d_3$. The other class is the SOSM-2 solution with $(X, Y) = (-R, 0)$, where $d_1 = d_2 < d_3$. These solutions in the $X$-$Y$ plane are schematically shown in Fig. 2(e). Namely, the SOSM-1 (SOSM-2) state is stabilized in the negative (positive) $b$ case. Therefore, in the system at $T/D = 0.02$, the sign of $b$ changes around $U/D \sim 2.25$.

At lower temperatures, an interesting orbital-selective state appears. Figure 3 shows the double occupancy and pair potential for each orbital when $T/D = 0.013$. When $U/D \leq 1.90,$ the $s$-wave SC state is realized with $\psi_1 = \psi_2 = \psi_3 \neq 0$. In the region with $U/D \geq 2.03$, the SOSM-2 state is realized with $d_1 = d_2 < d_3$ and $\psi_1 = \psi_2 = \psi_3 = 0$. Between these two states (1.90 $U/D \leq 2.03$), we find in Fig. 3 that the pair potentials as well as the double occupancies take two distinct values. In particular, one of the three orbitals has a finite pair potential while it vanishes for the other two. This implies the realization of a spontaneously orbital-selective superconducting (SOSSC) state.

Now, let us consider the nature of the SOSSC state. As shown in Fig. 3, the phase transition at $U/D \sim 1.90$ is of first order whereas that at $U/D \sim 2.03$ appears to be continuous. This suggests that the SOSSC state is closely related to the higher $U$ state, i.e., the SOSM-2 state. In this state, the orbital 3 is in a paired Mott state with large $d_3$ and the others are metallic. Decreasing $U$ from the SOSM-2 phase, $\psi_3$ becomes finite at $U/D \sim 2.03$ with an accompanying rapid decrease of the double occupancy $d_3$. This indicates that the orbital 3 plays an essential role in the phase transition to the SOSSC state. The behavior observed in orbital 3 is similar to the low temperature properties of the single-band attractive Hubbard model at half filling, where a second-order phase transition occurs between the SC and paired Mott states [44, 45]. This is consistent with the present result that the phase transition at $U/D \sim 2.03$ is of second order.

The orbital-selective superconducting instability originates from the existence of paired Mott orbitals in the SOSM state.
Therefore, a different type of SOSSC state may exist adjacent to the SOSM-1 state, whose essential feature should be described in terms of the two-band Hubbard model. The SC state in the latter model is realized in a narrow parameter space [39], which suggests that the potential SOSSC state related to the SOSM-1 phase is less stable and is difficult to realize in the present parameter regime.

![Phase diagram of the three-orbital Hubbard model](image)

**FIG. 4.** Phase diagram of the three-orbital Hubbard model. Solid circles represent second-order phase transition points. Open circles (crosses) represent the transition points, where the strong (weak) coupling state disappears. Shaded areas bounded by these points indicate the regions with two competing solutions.

By performing similar calculations for different temperatures, we obtain the phase diagram shown in Fig. 4, which clarifies the competition between the different ordered phases. For example, this phase diagram indicates that the first-order phase boundary between the \( s \)-wave SC and SOSM-1 states shifts to larger \( U \) with decreasing temperature. In the stronger coupling region and at low temperatures, the SOSM-1 state is realized instead of the Mott state adjacent to the SC state. This means that, due to the breaking of orbital symmetry, the SOSM-1 state is more stable than the Mott state.

The dominant electronic configurations for the SOSM-1 and SOSM-2 states are similar, but the SOSM-2 state is stabilized only at higher temperatures. This can be explained as follows. The SOSM states possess both itinerant and paired-Mott orbitals. The entropies for itinerant and paired Mott orbitals should be given by \( S \sim \gamma T \) and \( S \sim \ln 2 \), respectively, where \( \gamma \) is the specific heat coefficient proportional to the effective mass. Therefore, at high temperatures, the metallic orbitals tend to have a large entropy. For this reason, the SOSM-2 state with two metallic orbitals is realizable only at intermediate temperatures. For similar reasons, the SOSSC state with one orbital superconducting and the others metallic is less stable than the SC state with all orbitals superconducting at zero temperature. Therefore, the SOSSC state is stabilized only at finite temperatures.

In the present DMFT calculations, we did not consider the spin exchange part of the Hund coupling and pair hopping term. For the system with antiferromagnetic Hund coupling, the low-spin state is favored and the spin-flip is irrelevant. On the other hand, it has been clarified in Ref. 16 that the pair hopping is relevant to stabilize the SOSM-1 and SC states. To reveal the effect on the SOSM-2 state, we adopt a phenomenological theory for simplicity (details are given in the Supplemental Material). In the case without pair hopping, the SOSM-2 state appears between the high-temperature metallic and low-temperature SOSM-1 states. This is consistent with the result obtained by the DMFT calculations, which supports the validity of our phenomenological theory for the present system. Applying this theory to the system with pair hopping yields the prediction that the SOSM-2 state also exists in the intermediate temperature region. This result suggests that the SOSM-2 state survives even in the presence of pairing.

In summary, we have studied the three-orbital Hubbard model in infinite dimensions, combining DMFT with the CTQMC method. Calculating the pair potential and double occupancy in each orbital, we have determined the finite temperature phase diagram of the model. We have clarified that an orbital-selective \( s \)-wave superconducting state with one orbital superconducting and the others metallic is spontaneously realized, in addition to the conventional \( s \)-wave superconducting state and two kinds of spontaneously orbital-selective Mott states. Finally, we briefly discuss the relevance to real materials. The Jahn-Teller metal in the fullerene-based solids \( \text{A}_2\text{C}_{60} \) is a promising candidate for the SOSM-1 state [16]. Our results suggest that the higher-temperature part of the Jahn-Teller metal is in fact an SOSM-2 state. If one can experimentally distinguish the SOSM-2 state from the low-temperature SOSM-1 state, for example by a difference in the electrical conductivity, the new type of spontaneous orbital-selective superconducting state should exist in the vicinity of this transition line and the SC phase in \( \text{A}_2\text{C}_{60} \).

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In this material we develop a phenomenological theory that accounts for the thermodynamics of SOSM states. We introduce the labels 0, 1 and 2 to indicate orbital-symmetric metal state (three metallic orbitals), SOSM-1 (one metallic orbital / two paired orbitals) and SOSM-2 (two metallic orbitals and one paired orbital), respectively. A metallic orbital results in a free energy gain from the kinetic energy \((K)\) and the \(T\)-linear entropy \((S \propto T)\). For a paired orbital, on the other hand, there is a free energy gain from the effective attraction \((V)\), and also from the entropy \(S = \ln 2\) associated with the degrees of freedom of locating the pairs in the orbitals. The effective free energies can thus be expressed as

\[
F_0 = -3K - T(3\gamma T) \\
F_1 = -K - 2V - T(\ln 2 + \gamma T) \\
F_2 = -2K - V - T(\ln 2 + 2\gamma T)
\]

where \(K > 0, V > 0, \gamma > 0\) are the (renormalized) kinetic energy, effective attraction, and specific heat coefficient \((\gamma \sim 1/K)\), respectively. Our purpose here is to describe the thermodynamic stability of these states, while the kinetic energy of the pairs, which is necessary for e.g. superconductivity, is neglected. We focus on the low-energy region and neglect the \(T\) dependence of \(K, V, \gamma\).

We are interested in the stability of the SOSM-2 state on which the SOSSC is founded. The inequalities \(F_0 > F_2\) and \(F_1 > F_2\) are the necessary and sufficient condition for the realization of SOSM-2 as the most stable state. This leads to the relation

\[
V > K - \frac{(\ln 2)^2}{4\gamma},
\]

which determines the lower bound of the attractive interaction \(V\). The typical temperature dependences of the free energy are shown in the upper panels of Fig. 1. When the effective attraction is sufficiently large, the SOSM-2 state becomes the most stable one in the intermediate temperature range, which is qualitatively consistent with the results shown in the main text. This demonstrates that the above simple theory can capture the thermodynamics of the SOSM states realized in the DMFT study.

Since the above phenomenology works well for the effective description of the DMFT results, we now apply it to the system with pair hopping. The free energies in this case are

\[
\begin{align*}
F_0 &= -3K - T(3\gamma T) \\
F_1 &= -K - 2V - J - T(\gamma T) \\
F_2 &= -2K - V - T(\ln 2 + 2\gamma T)
\end{align*}
\]

where \(J > 0\) is the (effective) pair hopping. The pair hopping modifies only \(F_1\), since the two paired orbitals are quantum-mechanically mixed, which results in an energy gain of \(J\). The condition for the situation where SOSM-2 is most stable in the presence of pair hopping becomes

\[
\begin{align*}
V &> K - \frac{(\ln 2)^2}{4\gamma} \\
J &< \frac{\ln 2}{\gamma} \left( \ln 2 + \sqrt{(\ln 2)^2 + 4\gamma(V - K)} \right).
\end{align*}
\]

Thus, there is an upper bound for the magnitude of \(J\), in addition to a lower bound for \(V\). The typical temperature dependences of the free energies are shown in the lower panels of Fig. 1, and the necessary conditions described by Eqs. (4) and (5) are indicated in the right-most panel. Although it is not trivial to determine whether or not the realistic values are located inside of this region, there is a chance that the SOSM-2 state is realized if the effective attraction \(V\) is strong enough, which pushes the upper bound of \(J\) to higher values.
FIG. 1. Temperature dependences of the free energies without (top panels) and with (bottom panels) pair hopping. We have used $\gamma = \alpha/K$ with $\alpha = 1$. The intervals highlighted in red indicate the temperature range where the SOSM-2 is the most stable. The right-most panel in the bottom row shows the stability region of the SOSM-2 state determined by Eqs. (4) and (5).