A new numerical algorithm for the double-orbital Hubbard model — Hund-coupled pairing symmetry in the doped case

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In order to numerically study electron correlation effects in multi-orbital systems, we propose a new type of discrete transformation for the exchange (Hund’s coupling) and pair-hopping interactions to be used in the dynamical mean field theory + quantum Monte Carlo method. The transformation, which is real and exact, turns out to suppress the sign problem in a wide parameter region including non-half-filled bands. This enables us to obtain the dominant pairing symmetry in the double-orbital Hubbard model, which shows that the spin-triplet, orbital-antisymmetric pairing that exploits Hund’s coupling is stable in a wide region of the band filling.

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In the physics of correlated electron systems, as highlighted by the high-$T_c$ superconductivity, the prototypical transition-metal oxides have arrested attention because of a rich variety of physical properties, among which are anisotropic pairing symmetries in the cuprates and in Sr$_2$RuO$_4$, the colossal magnetoresistance in manganites, and a complex phase diagram for La$_{1-x}$Sr$_x$MnO$_3$. Since the $d-$orbitals are relevant, we are actually talking about multi-orbital systems, and, while some of the phenomena should be generically captured within single-band models, understanding of the effect of electron correlation in multi-orbital system remains a fundamental problem.

Namely, the orbital degrees of freedom should couple to other degrees of freedom such as charge, spin, and lattice distortion, and we can expect even richer physical properties. Indeed, the colossal magnetoresistance results from Hund’s coupling and the complex phase diagrams of Mn and Co compounds are considered to arise from a competition between Hund’s coupling and the Jahn-Teller distortion. In perovskite-type oxides, the crystal field splits the five $3d-$orbitals into three-fold degenerate ($t_{2g}$) and two-fold degenerate ($e_g$) levels. When the degenerate levels are not fully filled, the degeneracy may be lifted by the Jahn-Teller effect, where the system is effectively mapped to a single-orbital model when the splitting is large enough. We take here the small splitting limit to concentrate on the physics specifically caused by the orbital degrees of freedom.

Both numerical and analytic methods have been developed to study correlated electron systems. The methods should preferably be non-perturbative if one wants to examine the effect specific to the electron correlation such as Mott’s metal-insulator transition. The dynamical mean field theory (DMFT)\cite{DMFT}, which can fully include temporal fluctuations while spatial fluctuations are neglected, first succeeded in describing this transition both from metallic and insulating sides. In this method a lattice system (such as the Hubbard model) is mapped to an impurity model, which becomes an exact mapping in the limit of infinite spatial dimension. A standard procedure is to solve the impurity problem with the (auxiliary-field) quantum Monte Carlo (QMC) method\cite{QMC}, which involves no approximations except for the Trotter decomposition. So the DMFT+QMC method should be a desirable candidate for the multi-orbital cases. Unfortunately, it is difficult to extend the QMC method to multi-orbitals: First, it is impossible to express those (exchange and pair-hopping) interactions that are specific to multi-orbital cases in terms of the usual auxiliary fields. Second, even when we can accomplish this, the negative sign problem, a notorious problem in QMC calculations, is usually difficult to avoid for multi-orbitals.

This has motivated us to propose here a new auxiliary-field transformation that is applicable to Hund’s and pair-transfer terms. The transformation, which is real and discrete, turns out to suppress the sign problem in a wide parameter region including non-half-filled bands. This enables us to examine the role of Hund’s coupling and pair-transfer in a double-orbital model. One of the most intriguing questions for correlated electrons on multi-orbits is what should be the symmetry of the superconducting pairing that arises from the electron-electron interaction. We have examined the dominant pairing symmetry in the double-orbital Hubbard model with the DMFT+QMC method. The result reveals that the spin-triplet $\otimes$ orbital-antisymmetric $\otimes$ even-frequency pairing, which exploits Hund’s coupling, is stable in a wide region of the band filling.

So we take the two-fold degenerate Hubbard model\cite{Hubbard} with a Hamiltonian,

\begin{equation}
H = H_0 + H_1 + H_2,
\end{equation}

\begin{align*}
H_0 &= -t \sum_{i\sigma} \sum_{m} c_{im\sigma}^\dagger c_{im\sigma} - \mu \sum_{im\sigma} n_{im\sigma}, \\
H_1 &= U \sum_{i,m} n_{im\uparrow} n_{im\downarrow} + U' \sum_{i\sigma} n_{i1\sigma} n_{i2\sigma} - J \sum_{i\sigma} n_{i1\sigma} n_{i2\sigma},
\end{align*}
$H_2 = J \sum_{i,m \neq m'} \left( c_{im\sigma}^\dagger c_{im'\sigma} c_{im\sigma} c_{im'\sigma} + c_{im\sigma}^\dagger c_{im'\sigma} c_{im'\sigma}^\dagger c_{im\sigma}^\dagger \right)$.

Here $c_{im\sigma}$ creates an electron of spin $\sigma$ in the orbital $m (=1,2)$ at site $i$, and $n_{im\sigma} \equiv c_{im\sigma}^\dagger c_{im\sigma}$. We only consider the nearest-neighbor hopping between the similar orbitals, electron-electron interactions are assumed to be intra-atomic with the intra-(inter-)orbital Coulomb interaction denoted as $U(U')$, while the exchange and pair-hopping interactions as $J$. The Hamiltonian is rotationally invariant not only in spin, but also in real space if we fulfill the condition $U = U' + 2J$ (as is the case with $d$-orbitals). We have divided the interaction into the density-density interactions $H_1$ and the exchange and pair-hopping interactions $H_2$.

The DMFT+QMC method has been used by many authors for the single-orbital Hubbard model. For the usual on-site Hubbard interaction, a decoupling is done with the discrete Hubbard-Stratonovich(HS) transformation \[ e^{-a[n_i, n_i] - \frac{1}{2}(n_i+n_i)} = \left\{ \begin{array}{ll}
\frac{1}{2} \sum_{n=1}^{+\infty} e^{\lambda s(n_i-n_i)} (a \geq 0) \\
\frac{1}{2} \sum_{n=-\infty}^{-1} e^{\lambda s(n_i+n_i)-1} + 1/2 (a < 0) \end{array} \right. \tag{2}
\]

with $\lambda \equiv \log(e^{[\alpha]} + \sqrt{e^{[\alpha]} - 1})$, which transforms the two-body interaction into one-body interactions summed over an auxiliary field $s$. Applying this to each interaction term on the discretized imaginary time, we can decompose the partition function $Z$ of the many-body system into a sum of the partition functions $Z_{\{s_i\}}$ of one-body systems as $Z = \sum_{\{s_i\}} Z_{\{s_i\}}$. The QMC samples the single-particle systems according to the weight $Z_{\{s_i\}}$. The negative sign refers to the fact that the weight is not positive-definite. The sign problem does not occur, as far as the DMFT is concerned, in the single-orbital Hubbard model because the impurity problem lacks the electron hopping terms.

While the auxiliary-field QMC method has been applied to some multi-orbital Hubbard models by neglecting the terms other than the density-density interactions ($H_1$ in eq. (1)), the QMC algorithm becomes a challenging problem for the exchange and pair-hopping interactions ($H_2$): the HS transformation (2) is obviously inapplicable to these terms. While a decoupling, $\exp(J \Delta \tau c_1^\dagger c_2 c_3^\dagger c_4) = (1/2) \sum_{s=1}^{+\infty} \exp(s \sqrt{J \Delta \tau} (c_1^\dagger c_2 - c_1 c_4^\dagger))$, is possible after breaking $e^{-\Delta \tau H_2}$ into a product of exponentials, it leads to a serious sign problem. Another attempt by Motome and Imada \[ e^{-\Delta \tau H_2} = \frac{1}{2} \sum_{r=\pm 1} e^{\lambda r \left(f_1-f_i\right)} e^{a(n_i+N_i)+bN_i N_i}, \tag{3} \]

where $\lambda \equiv \frac{1}{2} \log(e^{2J \Delta \tau} + \sqrt{e^{4J \Delta \tau} - 1})$, $a \equiv \log(cosh(\lambda))$, $b \equiv \log(cosh(J \Delta \tau))$, $f_\sigma \equiv c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}$, $N_\sigma \equiv n_{1\sigma} + n_{2\sigma} - 2n_{1\sigma} n_{2\sigma}$.

This transformation is exact with the auxiliary field $(r)$ being real and all the operators being hermitian. Although a term $N_\sigma N_\bar{\sigma}$ which is forth order in $n$ appears on the right hand side, we can apply the usual HS transformation to this term due to a property $N^2_\sigma = N_\sigma$. The resulting terms in the form $mn$ can be combined with the Coulomb terms, which are transformed with eq. (2), so that we need in total only two auxiliary-fields for $H_2$. Furthermore the interaction parameters $U, U', J$ can be varied independently, which means that we can treat the rotational symmetric cases of $U = U' + 2J$. We note that recently Han \[ e^{-\Delta \tau H_2} = \frac{1}{2} \sum_{r=\pm 1} e^{\lambda r \left(f_1-f_i\right)} e^{a(n_i+N_i)+bN_i N_i}, \tag{3} \]

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The average sign plotted as a function of inverse temperature for $n = 1.8$, $U = U' = 1$, $J = 0.2$ calculated with the present algorithm (circles) and with the one due to Ref.[9] (squares).

\[
\sum_{\{s_i\}} Z_{\{s_i\}} / \sum_{\{s_i\}} |Z_{\{s_i\}}|, \text{ is plotted as a function of the band filling for two sets of values of } \beta \equiv 1/T \text{ and } J \text{ for the two-orbital Hubbard model on an infinite dimensional hypercubic lattice, whose density of states is Gaussian with the band width } W = 2. \text{ We can see that the sign problem is avoided almost independently of the band filling. Figure 2 depicts the temperature dependence, which shows that } (sign) \text{ decreases mildly as the temperature lowers, so we can go to lower temperatures with the present transformation. One reason why the transformation reduces the amount of negative weights is that it has a single auxiliary field } (r \text{ in eq. (3)}) \text{ as the source of the negative sign; the other auxiliary fields, related to the density-density interactions, are irrelevant to the negative weights.}

To test how the present method works, we have applied it to the calculation of the superconducting susceptibility for the Hamiltonian. Since off-site, anisotropic pairings (such as $p$ and $d$-waves) cannot be treated within the DMFT, we confined ourselves to $s$-wave pairing. Even within that channel, various pairing symmetries are possible in multi-orbital systems, since the total symmetry now consists of spin $\otimes$ orbital $\otimes$ frequency, so

\[
\begin{array}{ccc}
\text{spin} & \text{orbital} & \text{frequency} \\
\hline
1SE & \text{singlet} & \text{symmetric} & \text{even} \\
3AE & \text{triplet} & \text{antisymmetric} & \text{even} \\
1AO & \text{singlet} & \text{antisymmetric} & \text{odd} \\
3SO & \text{triplet} & \text{symmetric} & \text{odd} \\
\end{array}
\]

are the possibilities. The pairs that are formed across different orbitals are especially interesting. The orbital-symmetric pairs are

\[
\begin{align*}
S^a : & \quad c_1^\dagger c_1 \text{ and } c_2^\dagger c_2, \\
S^b : & \quad c_1^\dagger c_1 - c_2^\dagger c_2, \\
S^c : & \quad c_1^\dagger c_2 + c_2^\dagger c_1,
\end{align*}
\]

where $c_1^\dagger c_1$ and $c_2^\dagger c_2$ are combined into bonding and antibonding states due to the pair-hopping term, while the orbital-antisymmetric pair is

\[
A : \quad c_1^\dagger c_2 - c_2^\dagger c_1.
\]

Here we take, without the loss of generality, the $S_z = 0$ channel for spin triplets, where $S_z$ is the $z$-component of the Cooperon spin.

So the interest here is which pairing symmetry is favored in the double-orbital system, especially in the presence of Hund’s coupling. For the single-orbital case the sites are mostly singly-occupied by electrons around the half-filling, where the pairing (usually in $d$-wave channel) occurs. The question is what would be the corresponding picture for the double-orbital case around half-filling. So we have calculated the pairing susceptibility $P$, which is related to the two-particle Green function $\chi$,

\[
\chi_{ll',mm'}(i\omega_n, i\omega_{n'}) \equiv \int_0^\beta \int_0^\beta \int_0^\beta \int_0^\beta \int_0^\beta \int_0^\beta \int_0^\beta \int_0^\beta d\tau_1 d\tau_2 d\tau_3 \tau_4 e^{i\omega_n(\tau_1 - \tau_2) + i\omega_{n'}(\tau_3 - \tau_4)}
\]

\[
\times \sum_{kk'} \sum_{k'k''m'm} \langle \tau_1 \rangle c_{kk'}^{\downarrow}(\tau_1) c_{\downarrow k''}(\tau_2) c_{\downarrow k'}^{\uparrow}(\tau_3) c_{k''m'}^{\uparrow}(\tau_4) \rangle \chi_{ll',mm'}(i\omega_n, i\omega_{n'})
\]

through the equation,

\[
P = g^\dagger \chi g = \sum_{ll',mm'} \sum_{i\omega_n, i\omega_{n'}} g_{ll',mm'}(i\omega_n, i\omega_{n'}) \chi_{ll',mm'}(i\omega_n, i\omega_{n'})
\]

Here $g$ is the form factor describing the pairing symmetry in orbital-frequency space, which is either even or odd in $\omega$ for each orbital component. We adopt $g_{odd}(\omega) = \text{sign}(\omega)$ and $g_{even}(\omega) \equiv 1$ here.

The two-particle Green function $\chi$ is obtained from the Bethe-Salpeter equation,

\[
\chi = \chi_0 + \chi_0 \Gamma \chi,
\]

where $\chi$ is defined in eq. (4), $\chi_0$ the bare two-particle Green function calculated as a product of the one-particle Green function, $G_\uparrow(\mathbf{k}, \omega)G_\downarrow(\mathbf{k} - \mathbf{q}, -i\omega)$, summed over the momenta, and $\Gamma$ the vertex function, all of them being matrices with respect to orbital and frequency indices. Within the DMFT we can replace $\Gamma$ with the local one,

\[
\Gamma_{loc} = (\chi_0^{loc})^{-1} - (\chi^{loc})^{-1},
\]

in the limit of infinite dimensions, where $\chi_0^{loc}$ and $\chi^{loc}$ are respectively the bare and the dressed two-particle Green functions for the effective impurity model, which are computed in the QMC.

The temperature dependence of the pairing susceptibilities is shown in Fig. 3 for $n = 1.8$, $U' = 0.7$, $J = 0.4$, and $U = U' + 2J = 1.5$. We can see that the spin-triplet, orbital-antisymmetric, even frequency pairing (denoted as 3AE) becomes dominant at low temperatures. The
enhancement of such a pairing should be due to Hund’s coupling, which tends to align electron spins across different orbitals. The result is consistent with Han’s[10] for the semi-elliptical density of states. While in the work of Han the susceptibility diverges at $T/W \sim 0.06$ for $J/W = 0.15, U'/W = 0.45$, we do not detect the divergence up to $T/W = 1/180$. This should be because electrons are less correlated in the hypercubic lattice than in the Bethe lattice (which the semi-elliptic density of states would represent), since the Gaussian density of states for the hypercubic lattice, with high-energy tails, has an effectively larger band width.

Figure 4 shows the band-filling dependence of the pairing susceptibilities at $T/W = 1/120$ for $U' = 0.7, J = 0.4$, and $U = U' + 2J = 1.5$. A new finding here is that the spin-triplet, orbital-antisymmetric (3AE) pairing induced by Hund’s coupling remains dominant in a rather wide range ($n = 1.6 \sim 2$) of hole-doping at this temperature. It declines, however, at low fillings ($n = 1 \sim 1.4$). This result may be related to a mechanism of superconductivity proposed by Capone[14] for multi-orbital systems close to the Mott transition, where the electron repulsion $U$ is envisaged to assist a pairing even for $s$-waves.

In summary, we have constructed a new discrete transformation (which reduces the sign problem and is applicable to doped bands) for the exchange and pair-hopping terms in double-orbit models, and implemented this in a DMFT+QMC calculation. Superconducting susceptibilities of the $s$-wave pairings calculated with this method show that the spin-triplet, orbital-antisymmetric even frequency pairing, enhanced by Hund’s coupling and being dominant at half-filling, is robust against hole doping. We note that it is difficult to investigate ferromagnetism in this model even with the present transformation, since the ferromagnetism appears only in very strongly correlated regime[15], where the sign problem becomes serious again, so this is a future problem.

Numerical calculations were partly performed on SR8000 in ISSP, University of Tokyo.

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