Generalization of the iterative perturbation theory and metal-insulator transition in multi-orbital Hubbard bands

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The iterative perturbation theory of the dynamical mean field theory is generalized to arbitrary electron occupation in case of multi-orbital Hubbard bands. We present numerical results of doubly degenerate $E_g$ bands in a simple cubic lattice. The spectrum shows the electron ionization and affinity levels of different electron occupations. For sufficiently large Coulomb integral, a gap opens in the spectrum at integer filling of electrons and the system becomes insulator. The present scheme is easy to combine with the LSDA electronic structure theory.

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The first principle electronic structure theory has been developed much recently in the field of strongly correlated electron systems, e.g., the LSDA+U method, the GW-approximation\[1\] etc. The LSDA+U method can include the spin, charge and orbital fluctuation in real space, and the GW approximation is formulated in the framework of the LSDA+U method or the GW-approximation. The metal-insulator transition and the anomalous metallic phase is now at the stage of our understanding in a unified picture of both low and high energy excitations due to application of the dynamical mean field theory (DMFT) developed by Georges and Kotliar\[3, 4, 5].

On the other hand, the DMFT might not be completely established. Several methods have been developed in order to calculate the Green’s function in the DMFT. The DMFT projects a system onto the single impurity Anderson model and this treatment is exact in the limit of the infinite dimension where the off-site Coulomb interaction can be neglected. The combination with the GW approximation is being developed to include the off-site Coulomb interaction\[6]. It is also tried to extend the single impurity approximation to the cluster approximation\[7, 8]. The effects of multi-orbitals were discussed\[9, 10, 11\] and the case of electron filling up to 1 was shown to be essentially the same as the case of the non-degenerate orbital\[12, 13\]. The combination with the LDA or the LSDA is also desirable\[14\].

To calculate the Green’s functions of the DMFT, one can use several computational schemes such as the quantum Monte Carlo simulation (QMC), the iterative perturbation theory (IPT), the non-crossing approximation (NCA), the exact diagonalization (ED), the numerical renormalization group (NRG) etc.\[15\]. QMC, ED and NRG are exact and, because of the exact calculation, would be limited for wide variety of applications. Other methods, IPT and NCA, are approximate methods and might be easier to combine with other calculation in realistic complex materials. The aim of this paper is to generalize the IPT to multi-orbital bands and to apply it to the metal-insulator transition in the doubly degenerate $E_g$ orbitals.

We start with the Hubbard-type Hamiltonian:

\[
H = \epsilon_d \sum_{j, m, \sigma} c_{j, m, \sigma}^\dagger c_{j, m, \sigma} + \sum_{j, m, \sigma, \sigma'} J_{j, m, \sigma} c_{j, m, \sigma}^\dagger c_{j, m, \sigma'} + \frac{1}{2} \sum_{j, m, \sigma} U_{j, m, \sigma} n_{j, m, \sigma} n_{j, m, \sigma'} - \frac{1}{2} \sum_{j, m, \sigma, \sigma'} (U_{j, m, \sigma} - J_{j, m, \sigma}) n_{j, m, \sigma} n_{j, m, \sigma'} .
\]

In the following calculations, we would simplify as $U_{j, m, \sigma} = U$ and $J_{j, m, \sigma} = J = 0$, but its generalization is just straightforward. The local Green’s function is then defined, since we neglect the $\textbf{k}$-dependence of the self-energy $\Sigma$ as usual in the DMFT, as

\[
G_{\sigma, \sigma'}(i\omega) = \frac{1}{V} \int d\textbf{k} \left[ (i\omega + \mu) \mathbbm{1} - \epsilon_d^0 \mathbbm{1} - h(\textbf{k}) - \Sigma(i\omega) \right]^{-1}_{\sigma, \sigma'} .
\]

where $h(\textbf{k})$ is the transfer matrix in $\textbf{k}$-space, $\mu$ is the chemical potential, $\Sigma(i\omega)$ is the self-energy, and $\mathbbm{1}$ is the unit matrix. In case of a paramagnetic and orbitally degenerate system, which is the case in the present paper, the self-energy $\Sigma_{\sigma, \sigma'}(i\omega)$ are diagonal with respect to orbitals and spins and independent on them. The local Green’s function $G(i\omega)$ can be then in a form of

\[
G(i\omega) = \left[ i\omega + \mu - \Delta(i\omega) - \Sigma(i\omega) \right]^{-1} ,
\]

where $\Delta(i\omega)$ is the effective hybridization function. From the definition of the “effective” medium, one defines an Green’s function of the effective medium as

\[
G^0(i\omega) = \left[ i\omega + \tilde{\mu} - \Delta(i\omega) \right]^{-1} .
\]

The second order self-energy can be defined as

\[
\Sigma^{(2)}(\tau) = -U^2 (N_{\text{deg}} - 1) G^0(\tau) G^0(-\tau) ,
\]
where \( N_{\text{deg}} \) is the degeneracy with respect to spins and orbitals (here, in case of \( E_g \) orbitals, \( N_{\text{deg}} = 2 \times 2 = 4 \)).

The IPT was developed by Kajueter and Kotliar \[14\] for non-degenerate orbital. Here, we assume a simple form of the self-energy as

\[
\Sigma(i\omega) = U n(N_{\text{deg}} - 1) + \frac{A \Sigma^{(2)}(i\omega)}{1 - B(i\omega)\Sigma^{(2)}(i\omega)},
\]

where \( n_d = \sum_{m\sigma} n_j m\sigma = n N_{\text{deg}} \) and \( n \) is actually the occupation par each orbital. The coefficients \( A \) and \( B \) should be determined under the following guidelines: (1) \( A \) is determined so that the self-energy \( \Sigma(i\omega) \) is correct in the limit of \( i\omega \to \infty \). (2) \( B(i\omega) \) is determined so that the self-energy \( \Sigma(i\omega) \) is correct in the atomic limit, \( i.e. U \to \infty \).

The exact form of the self-energy in the limit of \( i\omega \to \infty \) is generally given as

\[
\lim_{i\omega \to \infty} i\omega \{ \Sigma_{m\sigma,m'\sigma}(i\omega) - \langle [H_1, c_{m\sigma}], c_{m'\sigma}^\dagger \rangle \} = \left( \langle [H_1, c_{m\sigma}], [c_{m'\sigma}^\dagger, H_1] \rangle - \langle [H_1, c_{m\sigma}], c_{m'\sigma}^\dagger \rangle \right)^2,
\]

where \( H_1 \) is the electron-electron interactions. The brackets \([\cdots]\) and \( \{\cdots\} \) are the commutator and anti-commutator, respectively. In the present case, the limiting form of the self-energy is

\[
\lim_{i\omega \to \infty} i\omega \{ \Sigma_{m\sigma,m'\sigma}(i\omega) - U n(N_{\text{deg}} - 1) \}
\]

\[
\simeq U^2(N_{\text{deg}} - 1)n(1 - n),
\]

where \( n^0 \) is the occupation number for the effective medium Green’s function \( G^0 \) and, together with \( \tilde{\mu} \), is determined so as to satisfy the Luttinger theorem. Then the final form of \( A \) should be

\[
A = \lim_{i\omega \to \infty} \frac{\Sigma(i\omega) - U n(N_{\text{deg}} - 1)}{\Sigma^{(2)}(i\omega)} = \frac{n(1 - n)}{n^0(1 - n^0)}. \tag{10}
\]

The required form of \( B(i\omega) \) may be

\[
B(i\omega) = \Sigma^{(2)\text{at}}(i\omega)^{-1} - \Sigma^{\text{at}}(i\omega) - U n(N_{\text{deg}} - 1)]^{-1} A,
\]

where \( \Sigma^{(2)\text{at}} \) is the second order self-energy in the atomic limit.

The self-energy in the atomic limit can be calculated exactly, or even can be written down in an analytic form, through the atomic Green’s function for the \( N_{\text{deg}} \)-fold atomic level:

\[
\begin{align*}
G^{\text{at}}_{mm'\sigma}(i\omega) &= \delta_{mm'} \sum_{n_\sigma^{\prime} = 0}^{N_{\text{deg}} - 1} \frac{(N_{\text{deg}} - n_{\sigma}^{\prime}) p_{n_{\sigma}^{\prime}} + (n_{\sigma}^{\prime} + 1) p_{n_{\sigma}^{\prime} + 1}}{i\omega - \epsilon_0^{\prime} - U n_{\sigma}^{\prime} + \mu} \tag{12}
\end{align*}
\]

\( G^{\text{at}}_{mm'\sigma}(i\omega) \) is the probability of finding an atom in a state of \( n_{\sigma}^{\prime} \) electrons present. Here the atomic levels may be given as \( E_0^{\prime} + U n_{\sigma}^{\prime} \) according to the electron occupation \( n_{\sigma}^{\prime} = 0, 1, 2, \cdots \) and all configurations are degenerated when the electron occupation is identical. If we would adopt more realistic Coulomb and exchange integral parameters \( U_{mn}\) and \( J_{mn} \) rather than the present simplified ones, the atomic levels should be split and they depend on the total angular momentum.

In order to get a rigorous expression of the self-energy, one can assume the limit of the inverse temperature \( \beta \to \infty \) with respect to \( U \). Then the occupation probabilities of each configuration, for \( r < n_d < r + 1 \) (\( r \)): integer number), are

\[
p_r = 1 - (n_d - r), \quad p_{r+1} = (n_d - r), \quad p_{r+1} = 0 : r' \neq r, r + 1. \tag{13}
\]

The resultant expression of \( B(i\omega) \) may be as follows:

\[
B(i\omega) = \frac{i\omega - \epsilon_0^{\prime} + \tilde{\mu}}{U^2(N_{\text{deg}} - 1)n(1 - n^0)} \frac{(X - a)(X - b)}{(X - a)^2 - ab - U^2} - (a + b) ab, \tag{14}
\]

where, for \( r < n_d < r + 1 \) (\( r \): integer number),

\[
X = i\omega - \epsilon_0^{\prime} - U r + \mu, \quad q = n_d - r,
\]

\[
a + b \equiv U \frac{N_{\text{deg}}}{N_{\text{deg}} - 1} \{- q(N_{\text{deg}} - 1) + r\},
\]

\[
ab \equiv - \frac{U^2}{N_{\text{deg}}} \{N_{\text{deg}}(1 - q) - r + 2rq + q\}. \]

Then finally we get an analytic expression of the self-energy of the present generalized IPT. The resultant expression of the self-energy Eq. \( [13] \) with Eqs. \( [10] \) and \( [14] \) are actually simple extension of the IPT \( [14] \) and it is reduced to the known expression when \( N_{\text{deg}} \to 2 \). It is also similar to the results given in \( [14] [11] \) but not identical.

We now present our results of the doubly degenerate \( E_g \) orbitals \( (N_{\text{deg}} = 4) \). We consider the Slater-Koster type tight-binding Hamiltonian of the doubly degenerate \( E_g \) orbitals in a simple cubic lattice with \( \epsilon_0^{\prime} = 0 \). The effective transfer integrals are assumed only between nearest neighbor pairs and \( V_{\sigma\sigma}^D = 1/3, V_{\sigma\sigma}^D = -2/3V_{\sigma\sigma}^D, V_{\sigma\sigma}^D = 1/6V_{\sigma\sigma}^D \). The relation among \( V_{\sigma\sigma}^D, V_{\sigma\sigma}^D \) and \( V_{\sigma\sigma}^D \) is the scaling properties of bare two-center integrals in the LMTO method. \[13\] Here the band width is \( 6(V_{\sigma\sigma}^D + V_{\sigma\sigma}^D) = 7/3 \) (the half band width \( D = 7/6 \). In a simple cubic lattice, there is no \( \delta \sigma \) interaction within the \( E_g \) orbitals and \( V_{\sigma\sigma}^D \) does not appear. Figure \[13\] shows the \( \omega(k) \)-curve and the real and imaginary parts of the local Green’s function with \( \Sigma = 0 \) and \( \beta \to \infty \).

The transfer matrix \( \{h(k)\} \) is diagonalized at every k-point. The local Green’s functions satisfy the relation \( G_{\sigma\sigma}^{x^2-y^2} = G_{\sigma\sigma}^{x^2-y^2} \) and \( G_{\sigma\sigma}^{x^2-y^2} = 0 \) after the k-integration within the
As shown in Fig. 2, for a simple cubic lattice. The high symmetry (left) and the local Green’s function (right) of Eg-bands in is unoccupied. The chemical potential decreases from ω bard bands with the electron-hole symmetry. At the critical region of the metal-insulator transition, and one can see very sharp coherent peak at ω = 0 when n = 1/4 and n = 1/2. In this region, the self-energy shows the common feature of the Fermi liquid.

With small hole doping, the spectrum changes very drastically especially when U = 2.2 and U = 3.0. A sharp peak appears at U below the lower Hubbard band, whose width depends upon the value U or the separation from the main lower Hubbard band. In this region of 1/4 < n < 1/2 or 1 < n_d < 2, the lower energy state is the mixture of the ground states of n_d = 1 and n_d = 2. Then, the ionization and affinity levels for both n_d = 1 and 2 configurations should appear in the spectrum. The satellite structure of all configurations n_d = 0, 1, 2, · · · could appear, but the intensity of satellites would depend on the energy separation. These satellite structure originates from the self-energy correctly given in the atomic limit.

Near the occupation n ∼ 1/4, the satellite at U above the upper Hubbard band shrinks. When the concentration n crossed the value 1/4, i.e. 0 < n_d < 1, the upper satellite disappears and the lower satellite merges into the main (lower) Hubbard band, since the main contribution to the spectrum comes from the mixture of n_d = 0 and 1, rather than n_d = 1 and 2 of the case 1 < n_d < 2. In fact at this concentration, the chemical potential shifts rapidly or jumps and the satellite grows to a main lower Hubbard band. Then the behavior for 0 < n < 1/4 is rather similar to the case of 0 < n < 1 of the non-degenerate orbital and the difference is only the fact that the intensity of the lower and upper bands starts from the ratio of 1 : 3 near n ∼ 1/4 rather than 1 : 1 of n ∼ 1/2 in case of the non-degenerate orbit.

The k-dependent spectrum can be analyzed through the k-dependent Green’s function $G_{mm'\sigma}(\omega, k) \equiv \left[(\omega + \mu)\Pi - \delta_{mm'}^0 \Pi - h(k) - \Sigma(\omega)\right]^{-1}_{\sigma'\sigma}$. The band dispersion width maybe sensitively depend upon the electron occupation. In the region of ω ∼ 0, the Green’s function behaves as $G(\omega, k) \approx Z/[\omega\Pi - Z h(k)]$, where Z is the renormalization factor. This situation may be seen, in case of the small hopping from half filling. The top of the coherent peak shows a small dip which is a characteristic feature of the lattice Green’s function of the $E_q$ state. The height of the coherent peak is almost equal to each other at n ∼ 1/2 and n ∼ 1/4. Then the renormalization factor or the effective mass may be also equal.

In general, it may be very important to have a simple scheme for solving the self-consistent Green’s function in multi-orbital cases, since further generalization is
FIG. 2: The electron occupation dependence of the imaginary part of the local Green’s functions for the doubly degenerate $E_g$ band of the half band width $D = 7/6$. (a) $U = 1.0$, (b) $U = 2.2$ and (c) $U = 3.0$. The numbers in the figure denotes the electron occupation $n$ for each spectrum. The inverse temperature $\beta = 30$. The region $\omega < 0$ is occupied and that $\omega > 0$ is unoccupied. In case of the electron occupation $n = 1/4$ and $1/2$ at $U = 3.0$, a gap opens and systems are in insulating phase. 

needed for application to realistic materials showing very interesting interplay of spin, charge and orbital ordering. For that purpose, the present scheme is very practical and straightforward and, furthermore, the generalization for magnetically ordered systems and clusters containing several atoms may be quite simple. 

In conclusion, we developed a new generalization of the self-energy of the DMFT-IPT applicable to arbitrary electron occupation $0 < n < 1$ or $0 < n_d < N_{\text{deg}}$. The spectrum shows the electron ionization and affinity levels of different electron occupations. For sufficiently large Coulomb integral, the system becomes the insulating state at integer filling of $n_d$. This generalization of the IPT may be easily applicable to more general cases and the combination with the LSDA in more complex systems.

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