A new approach for perovskites in large dimensions

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Abstract

Using the Hubbard Hamiltonian for transition metal-3d and oxygen-2p states with perovskite geometry, we propose a new scaling procedure for a nontrivial extension of these systems to large spatial dimensions $D$. The scaling procedure is based on a selective treatment of different hopping processes for large $D$ and can not be generated by a unique scaling of the hopping element. The model is solved in the limit $D \to \infty$ by the iterated perturbation theory and using an extended non-crossing approximation. We discuss the evolution of quasi particles at the Fermi-level upon doping, leading to interesting insight into the dynamical character of the charge carriers near the metal insulator instability of transition metal oxide systems, three dimensional perovskites and other strongly correlated transition metal oxides.

The electronic structure of the strongly correlated copper oxide superconductors [1], other transition metal and rare earth based perovskites [2], and of transition metal oxides [3] NiO, CoO or MnO has been an enduring problem in the last years, still containing numerous unresolved questions. Among them are the applicability of a Fermi liquid description, the evolution and doping dependence of coherent quasi particles near the Fermi energy in a doped Mott Hubbard or charge transfer insulator and the transfer of spectral weight from high to low energy scales. In these materials, it is of importance to take the local spin and charge fluctuations of the oxygen states explicitly into account. A theoretical approach that maintains the dominating local correlations and which allows the calculation of the excitation spectrum of strongly correlated systems was recently proposed by Metzner and Vollhard [4]. They introduced a dynamical mean field theory, where the system is mapped on a local problem coupled to an effective bath. [4–7] This dynamical mean field approach is exact in the limit of large coordination numbers or equivalently for large spatial dimensions ($D \to \infty$). In most applications of this approach, the one
band Hubbard model is considered. A scaling of the hopping element like \( t = t^*/\sqrt{D} \) with fixed \( t^* \) leads for \( D \to \infty \) to a remarkable simplification of the many body problem while retaining the nontrivial local dynamics of the elementary excitations and other main features of the model. [6,7] Only few attempts have been made to extend the dynamical mean field theory to systems with more than one orbital degree [8,9].

Considering the \( D \)-dimensional extension of the perovskite lattice, where the TM sites sit on a hypercubic lattice and the oxygen sites are located between every two nearest neighbour TM sites, the following two band Hubbard Hamiltonian for the TM 3\( d \) and O 2\( p \) orbitals results:

\[
H = \sum_{k\sigma} \left( \varepsilon_d d_{k\sigma}^\dagger d_{k\sigma} + \varepsilon_p p_{k\sigma}^\dagger p_{k\sigma} \right) + \sqrt{2Dt} \sum_{k\sigma} \left( \gamma_k d_{k\sigma}^\dagger p_{k\sigma} + H.c. \right) + U \sum_i d_{i\uparrow}^\dagger d_{i\uparrow} d_{i\downarrow}^\dagger d_{i\downarrow}. \tag{1}
\]

Here, \( d_{k\sigma}^\dagger \) (\( p_{k\sigma}^\dagger \)) creates a hole in a TM (O-2p) orbital with momentum \( k \) and spin \( \sigma \). \( \varepsilon_d \) and \( \varepsilon_p \) are the corresponding on-site energy, respectively. \( t \) is the amplitude of the nearest neighbour TM-O hopping integral and \( U \) is the Coulomb repulsion of TM holes. The coherence factor is given by \( \gamma_k^2 = 1 - \frac{1}{D\alpha_k} \) with \( \alpha_k = \sum_{\nu=1}^D \cos k\nu \). There exists no scaling like \( t = t^*/D^\beta \) with exponent \( \beta \) leading to a nontrivial limit of Eq. 1 for \( D \to \infty \). This results from the TM coordination number being 2\( D \) whereas the oxygen coordination number is 2. A selective consideration of different hopping paths on the \( D \)-dimensional lattice can be obtained by the following separate scaling of the hopping element in the coherence factor: \( t^2D\gamma_k^2 = 2(t^*)^2 - \sqrt{\frac{2}{D}}(t^*)^2\alpha_k \). Here, the TM-O-TM hopping precesses with identical and with different TM-sites are scaled differently such that for two different TM-sites, the transfer function \( t^2/(\omega - \varepsilon_p) \) and not \( t \) itself is scaled similar to the one band Hubbard model. Consequently, the decoupling of different TM-O ”dimers” of Ref. [8] does not occur. Based on this new scaling procedure, it follows similar to the one band case, that the self energy \( \Sigma_d(\omega) \) of TM states is momentum independent and that the system can be mapped onto an Anderson model with effective hybridization, which has to be calculated self consistently. In the following, we solve the model using the iterated perturbation theory [6] (IPT) and a modified version of the non-crossing approximation [10,11] (NCA).

In Fig. 1 we show our results for the TM- and O-densities of states obtained within the IPT for half filling \( x = 0 \) \( (x = n_d + n_p - 1) \). Two Hubbards bands dominated by TM-states are separated by \( U \) and two oxygen dominated bands in the neighborhood of \( \varepsilon_p \) are visible. Similar to the one band case, the IPT leads to an interesting structure of the density of states, where now four bands are occuring. However, the expected insulating behaviour at half filling (for large \( U \)) does not occur. This results from the overestimation of the spectral
weight of the lower Hubbard band within the IPT by $\propto 1 - n_d$. The occupation of oxygen sites ($n_p > 0$) due to TM-O hybridization leads to an overcounting of copper sites which can be occupied without paying any Coulomb energy. Therefore, the success of the IPT in the one band case does not occur within the two band model. This is due to the absence of the particle-hole symmetry and the change of the TM-occupation number $n_d$ as function of $t$ at half filling. This shortcoming of the IPT results from the occupation of O sites and is expected to vanish for small $n_p$ which occurs in the limit of a large value for $\Delta = \varepsilon_p - \varepsilon_d$.

However, for the physically interesting situation $\Delta \approx U/2$, the IPT leads to qualitatively wrong results and one has to develop theoretical approaches which take the local TM-O many body states explicitly into account. Therefore, we extended the NCA, which was shown to be in excellent agreement with quantum Monte Carlo (QMC) simulations in the one band case, such that a local TM-O hybrid with 16 local eigenstates is coupled to an effective medium. Here, the TM-O singlet and triplet states which are the important excitations near half filling are explicitly taken into account. In Fig. 2, we show our results for the densities of states obtained from this hybrid-NCA in the whole energy range and in the neighborhood of the Fermi level ($\omega = 0$). One clearly recognizes the two TM-dominated Hubbard bands and the O-dominated states near $\varepsilon_p$. Even the double peak structure of the Hubbard bands is in qualitative agreement with QMC calculations for the two dimensional three band Hubbard model [12]. The O-dominated state at the Fermi level results from the formation of a local TM-O (Zhang-Rice) singlet state [13]. The strong temperature dependence of this low energy singlet state is shown in the inset of Fig. 2, where the formation of coherent quasi particles on a temperature scale of 300 K occurs. This results from the spin fluctuations between a singly occupied TM site and the TM-O singlet state. Interestingly, these coherent quasi particles occur on this temperature scale only for doping values larger than $x \approx 0.04$. Finally, we indeed find an insulating state for half filling.

In conclusion, we proposed a new scaling procedure for perovskite systems in large spatial dimensions. It is based on a selective treatment of different hopping processes and can not be generated by a unique scaling of the hopping element. Solving this model within the iterated perturbation theory and an extended non-crossing approximation, it is shown that this procedure and the NCA reproduce important physics of the low dimensional situation including the formation of coherent quasi particles for low temperatures, while shortcomings of the IPT are clearly visible.

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Fig. 1. TM (solid line) and O (dashed line) density of states obtained within the iterated perturbation theory for half filling.

Fig. 2. TM (solid line) and O (dashed line) density of states obtained within the hybrid-NCA for $x = 0.1$. The inset shows the temperature dependence of the O-DOS for $x = 0.08$: $T = 10000$ K (dashed line), $T = 1000$ K (dotted line), and $T = 300$ K (solid line).
\( U = 8 \text{ eV} \)
\( \Delta = 3 \text{ eV} \)
\( t = 1.5 \text{ eV} \)
$\rho_{d(p)}(\omega)$ [eV$^{-1}$]

$U = 10 \text{ eV}$

$\Delta = 5 \text{ eV}$

$t = 1 \text{ eV}$