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Double occupancy in DMFT and the Dual Boson approach

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We discuss the calculation of the double occupancy using Dynamical Mean-Field Theory (DMFT) in finite dimensions. The double occupancy can be determined from the susceptibility of the auxiliary impurity model or from the lattice susceptibility. The former method typically overestimates, whereas the latter underestimates the double occupancy. We illustrate this for the square-lattice Hubbard model. We propose an approach for which both methods lead to identical results by construction and which resolves this ambiguity. This self-consistent dual boson scheme results in a double occupancy that is numerically close to benchmarks available in the literature.

The double occupancy plays an important role in the study of correlated systems. It is indicative of the Mott metal-insulator transition and of local moment formation. In optical lattice experiments, the double occupancy1–3 gives information about the phase. On the theory side, the double occupancy has been used to benchmark approximations4. It also enters the calculation of total energies and forces in LDA+DMFT studies of strongly correlated materials5. Given this important role, it is worthwhile to look at how the double occupancy is determined. Here, we consider the double occupancy in DMFT and its extension Dual Boson (DB). We compare our results with benchmark results4 available in the literature.

Over the past two decades, DMFT6,7 has become the dominant approximation for strongly correlated electron systems8. DMFT solves a self-consistently determined auxiliary single-site problem (the impurity problem) to determine properties of the original lattice problem. Initially, DMFT studies focused on infinite-dimensional systems, where the approximation becomes exact. In this case, the double occupancy is given by the double occupancy of the auxiliary impurity problem9. Nowadays, DMFT has become an accepted approximation also for finite-dimensional systems. In these calculations, it is often assumed that the double occupancy is equal to that of the impurity problem10. Here we show that this assumption is potentially problematic.

To be concrete, we study the Hubbard model as a prototypical example of a strongly correlated system. It is described by the Hamiltonian

\[ H = -t \sum_{(ab)\sigma} c_{a\sigma}^{\dagger} c_{b\sigma} + \sum_a U n_{a\uparrow} n_{a\downarrow}, \]  

where \( t \) is the hopping amplitude of an electron with spin \( \sigma \) between nearest-neighbor sites \( a \) and \( b \), and \( U \) is the on-site repulsion between electrons. The local repulsion \( U \) disfavors doubly-occupied sites and hence decreases the double occupancy \( d = \langle n_{\uparrow} n_{\downarrow} \rangle \).

The double occupancy is a local two-particle correlation function. It can be written as the difference between the charge and spin susceptibility. In obvious short-hand notation,

\[ d = \frac{1}{4} \left[ \langle nn \rangle - \langle S_z S_z \rangle \right] 
   = \frac{1}{4} \left[ X_{\text{loc}}^\text{ch} - X_{\text{loc}}^\text{sr} + \langle n \rangle \langle n \rangle - \langle S_z \rangle \langle S_z \rangle \right]. \]  

Here \( X_{\text{loc}}^\text{ch} = \langle nn \rangle - \langle n \rangle \langle n \rangle \) and \( X_{\text{loc}}^\text{sr} = \langle S_z S_z \rangle - \langle S_z \rangle \langle S_z \rangle \) are the equal-time, local correlation functions of the charge density \( n = n_{\uparrow} + n_{\downarrow} \) and the magnetization \( S_z = n_{\uparrow} - n_{\downarrow} \), respectively11. In DMFT, the charge and spin susceptibilities in turn are either approximated by the respective impurity susceptibility, or by the DMFT lattice susceptibilities. The latter are computed by summing ladder diagrams containing the lattice Green’s function and a local irreducible vertex7,12. The computation of the DMFT double occupancy is ambiguous, because the two-particle impurity correlation functions are not identical to the local lattice correlation functions. Recently, we showed this for the compressibility13.

To emphasize their difference, we can decompose the susceptibility into the impurity susceptibility \( \chi_{\text{imp}} \) and a
For comparison, DCA results extrapolated to infinite lattice size from Ref. 4 are shown (green circles). They can be considered the numerically exact solution of this model. The nonlocal vertex corrections in (3) include additional correlation effects that tend to reduce the double occupancy, making the system more insulating. On the other hand, due to its mean-field character, DMFT overestimates the Néel temperature. In particular, this means that the DMFT susceptibility (blue triangles) overestimates \( X^\text{up} \) and thus, according to (2), underestimates \( d \). This happens particularly at large \( U \), where the tendency towards antiferromagnetism is more pronounced and hence the strongest deviations occur. In fact, we find that the DMFT susceptibility results in a negative double occupancy at sufficiently large values of \( U \), which is clearly unphysical. The double occupancy of the auxiliary impurity, on the other hand, gets close to the DCA results at large \( U \). As \( U \) increases, the electrons localize and nonlocal corrections are less important.

We now turn our attention to the self-consistent DB (sc-DB) approach. The hallmark of this approach is the self-consistency condition

\[
X_{\text{loc}, \omega} = \chi_{\omega}^{\text{imp}},
\]

which resolves the above ambiguity by construction. This self-consistency condition is achieved by introducing a frequency-dependent interaction \( \Lambda \) to the auxiliary impurity model. The effect of this interaction is twofold. First of all, the auxiliary impurity problem and the associated \( \chi^{\text{imp}} \) are different from their DMFT values. Second, the lattice susceptibility has to be calculated as \( X^{-1} = X_{\text{DMFT}}^{-1} + \Lambda \), where \( X_{\text{DMFT}} \) is the usual DMFT susceptibility obtained from the two-particle ladder, and the equation should be understood in momentum and frequency space. It resembles the RPA expression for the susceptibility, where \( X_{\text{DMFT}} \) plays the role of the bare susceptibility and \( -\Lambda \) the role of the interaction.

We determine a self-consistent field both for the density and the spin (\( S_z \)) channel, as they differ in general.

The self-consistent dual boson approach described here shares certain characteristics with the two-particle self-consistent approach (TPSC). Both approaches determine an effective interaction for the spin and charge channel according to a self-consistency condition. Note however, that the original TPSC approach takes the RPA as its starting point, sc-DB starts from DMFT. Strong correlation effects are included in DB from the start. Furthermore, the effective interaction in TPSC is static. The Moriya-esque \( \lambda \) correction in DΓA is also somewhat similar in spirit. It, too, is a correction to the susceptibility that is used to impose self-consistency. Like in TPSC, the Moriya-esque correction is static. Therefore, in both cases, these can only fix certain sum-rules. The effective interaction in sc-DB, on the contrary, may be regarded as a frequency-dependent \( \Lambda \)-correction, which removes the ambiguity of calculating local susceptibilities.

We note that it was difficult to converge our scheme for \( U > 8 \). This coincides with the parameter region where...
The retarded interactions appear as both in the charge and in the spin channel, normalized by the difference between the screened and bare interaction is shown.

FIG. 3. Screening of the impurity interaction in sc-DB. The different values of \( U \) correspond to the point in Fig. 2. The difference between the screened and bare interaction is shown both in the charge and in the spin channel, normalized by the bare interaction. The retarded interactions appear as \( n\Lambda^h n + S_i\Lambda^s S_i \) in the impurity action, so both the positive sign in the charge channel and the negative sign in the spin channel suppress the double occupancy.

The dimensionality of the model plays an important role in DMFT-based studies. To illustrate the effect of the dimensionality, we have performed similar calculations in a 3d simple cubic lattice. The results are shown in Fig. 4. A direct comparison is difficult by the change in energy scales that occurs when changing the dimension. To overcome this, we use the Z-factor, \( Z = (1 - d\Re \Sigma_\omega / d\omega)^{-1} \), obtained in DMFT, to indicate the importance of interaction effects on the one-particle level. The renormalization of the double occupancy by nonlocal processes, \( d_{\text{sc-DB}} / d_{\text{DMFT}} \), is shown as a function of \( Z \). The figure clearly shows that the sc-DB susceptibility contains important nonlocal corrections. Although it is clear that other factors than dimensionality also play a role, Fig. 4 also suggests that the nonlocal renormalization of the double occupancy is less important in the 3d system.

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In conclusion, we have proposed a scheme which resolves the ambiguity in the calculation of the double occupancy inherent to DMFT in finite dimensions. DMFT is not self-consistent on the two-particle level, and as a result, the single- and two-particle quantities are not compatible, as exemplified here by the difference between the double occupancy from the Galitskii-Migdal formula and from the susceptibility. This ambiguity extends to the determination of total energies and forces from DMFT.

The self-consistent dual boson approach gives results which are in good agreement with benchmarks in the literature. We have further shown that the DMFT impurity double occupancy typically overestimates the double occupancy, while the occupancy determined from the DMFT lattice susceptibility underestimates it significantly. For large interaction, this can even lead to unphysical negative results. We have seen that in two-dimensional systems and at a moderate $Z$-factor of 0.5, the results may differ by 10%-20%. The double occupancies determined from the DMFT lattice and impurity susceptibilities differ strongly also away from half filling. The discrepancy may have a significant effect on total energy and force calculations and hence the determination of equilibrium positions of the atoms in realistic LDA+DMFT calculations. This should be kept in mind when interpreting the results. In order to avoid unphysical results we recommend to approximate the double occupancy by the impurity double occupancy in LDA+DMFT. The accuracy of this approach however remains to be determined.

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\[ d = \int_\nu \frac{2T}{U} \Sigma_\nu G_{\nu}, \]
based on the Galitskii-Migdal formula for the total energy. In infinite dimensions and in the DMFT approximation, the self-energy is local, and the formula simplifies to
\[ d = \int_\nu \frac{2T}{U} \Sigma_\nu G_{\nu}^{\text{loc}}. \]
This yields exactly the same double occupancy as the impurity problem, since the impurity problem is solved exactly.
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