Dynamical vertex approximation for nanoscopic systems

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With an increasing complexity of nanoscopic systems and the modeling thereof, new theoretical tools are needed for a reliable calculation of complex systems with strong electronic correlations. To this end, we propose a new approach based on the recently introduced dynamical vertex approximation. We demonstrate its reliability already on the one-particle vertex (i.e., dynamical mean field theory) level by comparison with the exact solution. Modeling a quantum point contact with 110 atoms, we show that the contact becomes insulating already before entering the tunneling regime due to a local Mott-Hubbard transition occurring on the atoms which form the point contact.

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Introduction. In recent years, we have seen a tremendous experimental progress in the direction of man-made nanostructures. For example, with the wizardry of modern semiconductor technology quantum effects in quantum dots could be revealed \(^1\)\(^-\)\(^7\) in the area of molecular electronics transport through single molecules can now be studied \(^8\)\(^-\)\(^10\); and for magnetic storage technology nanoclusters of transition metals on surfaces become relevant \(^11\). In all three examples electronic correlations play a decisive role since the restriction to nanostructures brings the electrons close to each other so that their mutual Coulomb interaction becomes large (compared to their kinetic energy or tunneling rates). As a matter of fact, electronic correlations are not only genuine to nanostructures, but they also make them fascinating, both from the basic research point of view, with new physics occurring, and from the point of applications since strong correlations result in spectacular physical properties. An example is the Kondo-type physics which overcomes the Coulomb blockade and which has been observed in the conductance of quantum dots \(^2\)\(^-\)\(^5\)\(^,\)\(^7\) as well as for small clusters and individual adatoms \(^12\)\(^-\)\(^15\).

The theoretical modeling of strong correlations in nanostructures attached to some environment (bath) such as the source and drain electrode in case of the quantum dot or the surface for the transition metal cluster is hitherto based on generalizations of the Anderson impurity model \(^5\)\(^-\)\(^7\)\(^,\)\(^13\)\(^-\)\(^18\). However, if one is not only dealing with a single or two "sites" (say the number of quantum dots), the numerical effort to solve the corresponding Anderson impurity model becomes prohibitively expensive. More precisely, the effort grows exponentially with the number of "sites" for an exact \(^19\)\(^-\)\(^20\) or numerical renormalization group \(^18\) treatment. This restricts these methods in effect to \(O(2)\) sites coupled to a bath. Related dynamical matrix renormalization group (DMRG) approaches \(^21\) might allow for slightly larger systems but ultimately suffer from the same non-polynomial problem, except for truly one dimensional geometries. Potentially more efficient quantum Monte-Carlo methods \(^22\) on the other hand exhibit a growing sign problem with increasing system size. Hence a good theory for correlated nano-systems with even a few coupled nano-objects is presently missing. Such a theory is however mandatory since future technological applications will require the engineering of complex networks of such nano-objects – be it for a quantum computer or for a von Neuman computer based on such small structures so that quantum effects are no longer negligible.

On the other side, dynamical mean field theory (DMFT) \(^23\)\(^-\)\(^26\) along with its cluster \(^22\) and diagrammatic extensions such as the dynamical vertex approximation (DFA) \(^27\) and the dual fermion approach \(^28\) has been applied to strongly correlated electron systems with great success: On the model level, among others, the Mott-Hubbard transition \(^24\)\(^,\)\(^25\), magnetism \(^29\), and kinks in strongly correlated systems \(^30\)\(^-\)\(^32\) could be better understood or have even been discovered. Merging DMFT with density functional theory in the local density approximation (LDA) \(^33\)\(^-\)\(^35\) turned out to be a breakthrough for the calculation of actual materials with strong correlations. By construction, these DMFT calculations are done in the thermodynamic limit, i.e., for a macroscopically large system.

There has been one attempt by S. Florens \(^37\) to establish a nanoscopic version of DMFT. The idea behind Florens’ nanoDMFT approach is the DMFTesque limit of a large number of neighbors with a central site in the middle, surrounded by many neighbors which in turn are coupled to many neighbors etc. In such a geometry (e.g., of a Cayley-type tree), one gets a recursive method where the inner sites depends on their outer neighbors but not vice versa. Experimentally however, such a geometry with more and more neighbors is hardly realizable, and the approach has been scarcely used in practice \(^38\).

Here, we hence take another route based on the DΓA concept of the locality of the fully irreducible \(n\) particle vertex \(\Gamma\). While the calculations in this first paper will be for \(n = 1\), i.e. on a DMFTesque level, we call the approach nanoDΓA in following – also to distinguish it from...
the aforementioned nanoDMFT. Below, we introduce the approach, validate its range of applicability against the exact numerical solution for system sizes where this is still possible, and demonstrate its potential by hands of calculations for a quantum point contact with 110 sites.

**Method.** As pointed out in the introduction, we are interested in a nanoscopic system consisting of nano-objects (sites) which are hybridized via tunnelling channels $V_{ij}$, interacting by a Coulomb repulsion $U_i$ and coupled by $V_{i\nu k}$ to some non-interacting environment, see Fig. 1. The Hamiltonian hence reads

$$H = \sum_{i\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i} U_i c_{i\uparrow}^\dagger c_{i\downarrow} + \sum_{i\nu k} V_{\nu k} c_{i\nu}^\dagger l_{\nu k\uparrow} + h.c. + \sum_{\nu k\sigma} \epsilon_{\nu k} l_{\nu k\sigma}^\dagger l_{\nu k\sigma},$$

(1)

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) and $l_{\nu k\sigma}^\dagger$ ($l_{\nu k\sigma}$) denote the creation (annihilation) operators for an electron with spin $\sigma$ on site $i$ and in lead $\nu$ state $k$ with energy $\epsilon_{\nu k}$, respectively. While we consider a single band situation in the following, Hamiltonian (1) can easily be generalized to include orbital realism, leading to an additional orbital index in the second quantization operators and orbital matrices in the Green functions and self energies below.

As an exact solution of Hamiltonian (1) is possible at most for $O(10)$ interacting sites, we here propose an approximate DΓA solution. To this end, we first need to calculate the fully irreducible $n$-particle vertex on every site $i$ with the interacting Green function $G_{ii}(\omega)$ and Coulomb interaction $U_i$ as an input, see Fig. 2. In practice, this is done by numerically calculating the corresponding $n$-particle vertex of an associated Anderson impurity model. Note that the effort for this computationally most expensive step only grows *linearly* with the number of sites and is easily parallelizable. From the $n$-particle vertex in turn, we recalculate the Green function and proceed with the first step until convergence. In the case $n=1$, the one-particle fully irreducible vertex is simply the self energy $\Sigma(\omega)$ which is directly related to $G_{ii}(\omega)$ through the Dyson equation given in Fig. 2 in matrix notation for the site indices $ij$. For $n=2$, one needs instead to use the parquet equations to go from the irreducible vertex to the reducible one and the exact equation of motion to get the self energy before proceeding with the Dyson eq., similarly as discussed in Ref[27] for an infinite system.

Let us note that the approach becomes exact in several limits: (i) $U \to 0$, (ii) $V \to \infty$, (iii) number of connections to neighbor sites $Z \to \infty$, and, if each site couples to its own lead (iv) $t \to 0$. While the exact quantum Monte Carlo simulation is impossible for large clusters due to the so-called sign problem, our method is sign problem-free. For $n=2$ the approach also takes into account the Cooperon diagrams so that weak localization physics is explicitly included, as are spin fluctuations.

**Validation vs. exact result.** As a first test case and to validate the approach for $n=1$ against the numerically
exact solution, we consider the 6-site benzene geometry of Fig. 1 (upper panel) with a constant density of states \( \rho \) in the contacts from \(-D\) to \(D\) around the Fermi level \((D = 2t, \ t = 1\) sets our unit of energy in the following); and a site-diagonal hybridization \(V_{i\nu k} = V \delta_{i\nu}\). Two topologies are considered: (i) hopping \( t_{ij} \) restricted to the two nearest neighbors in the hexagon \(\text{“nn } t\text{”}\) and (ii) an equal hopping to all sites \(\text{“all } t\text{”}\). Both DFA and the exact solution are calculated by means of Monte-Carlo simulations [11] for \(U = 5t\), temperature \(T = 0.05t\).

Fig. 2 shows the calculated local spectral function in an interval \(2T\) around the Fermi level (set to zero) \(\text{“}A(0)\text{”} = \int d\omega A(\omega)/\{e^{\omega/2T} + e^{-\omega/2T}\}\). The results clearly show that nanoDFA is a reliable hybridization for any hopping \(V \gtrless t\) and if enough neighbors (in our case 5 in the \text{“all } t\text{”} topology) are involved in the hopping processes. There are some deviations for the nearest-neighbor-hopping-only case if \(V \lesssim t\) since in this situation non-local correlations such as those involved in forming a two-site singlet are relevant. If the hopping is to all neighbors though, no deviations could be identified all the way down to the last calculable point \(V = 0.8t\). Below \(V = 0.8t\), the sign problem becomes too severe [average sign \(O(10^{-3})\)], and only the nanoDFA solution is possible - in a situation very favorable for nanoDFA because of the many neighbors. Let us note that another indication for the small correction is the off-diagonal self-energy, which is smaller than \(10^{-2}t\) for \(V > t\) and nearest neighbor hopping as well as for all \(V\) and hopping to all neighbors.

In the inset to Fig. 3 we present the conductance \(G\) through the benzene-like nanostructure, depicted in the upper panel of Fig. 1, from one side of the molecule to the opposite side, calculated along the lines of Ref. [22]. As the local spectral function, the conductance again shows the reliability of the calculation already on the one-particle vertex level, with discernible deviations from the exact solution only for a low number of neighbors (i.e., \(2\) “nn only”) and small hybridization to the leads. The results can be understood as follows: At small hybridization \(V\), we have a conductance through two tunneling lead-benzene contacts, leading to an increase of the conductance \(\sim V^4\) (i.e., squared tunneling rate \(\Gamma = \pi V^2 \rho\)). In the large \(V\) region on the other hand, a Kondo resonance between the individual sites of the benzene molecule and the respective lead forms, which suppresses the competing inter-benzene hopping and hence the transport through the molecule. If hopping between all benzene sites is allowed ("all \(t\)"), this effect is less pronounced since there is a direct hopping channel between the opposite sites to which the voltage has been applied.

**Quantum point contact (QPC).** To demonstrate the suitability of the approach for more complex nanosystems, let us now consider a QPC. Experimentally it can be realized e.g. through a mechanically controllable break junction of a conducting wire, see e.g. Ref. [23-25]. The assumed geometry is based on a body-centered cubic basic structure narrowed to a double-cone-like junctions as shown in Fig. 1 (lower panel). For the moment we assume a single band which might be realizable in more complex wires such as cuprate and cobaltate wires, but realistic calculations, e.g., in the spirit
of LDA+DMFT \cite{33-36}, are certainly possible. The parameters are: $U = 4t$, $t = -1$, $t' = -0.4$, $V = 0.4$, and $T = 0.05t$. Each calculation takes about 10 hours with a mildly parallelization on 25 Nehalem Intel processors (X5550, 2.66GHz), showing that much bigger calculations or calculations with orbital realism are possible. When slowly breaking up the junction the hybridization (tunneling) $t_{QPC}$ between the two atoms forming the point contact will change most strongly and is hence varied.

Surprisingly, we observe a dramatic reduction of the conductance for $|t_{QPC}| < 0.5t$ in Fig. 4. Breaking up the junction triggers a local Mott-Hubbard “transition” (more precisely a crossover) of the two atoms forming the quantum point, see Fig. 3 inset. Therefore the conductance drop with increasing distance between the two atoms is faster than the exponential decay of $t_{QCP}$ with distance. Our findings might explain similar experimental observations \cite{44} in transition metals point contacts with partially filled d-shells, where the electrons are actually similarly strongly correlated as in our calculation. This effect could not have been revealed with hitherto employed methods \cite{45} such as LDA, Landauer formalism or Coulomb blockade calculations.

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