Cellular Dynamical Mean Field Approach to Strongly Correlated Systems

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We propose a cellular version of dynamical mean field theory which gives a natural generalization of its original single-site construction and is formulated in different sets of variables. We incorporate a possible nonorthogonality of the tight-binding basis set and prove that the resulting equations lead to *manifestly causal* self-energies.

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Dynamical mean field theory (DMFT) has been very successful in describing many aspects of strongly correlated electron systems [1], and presently much effort is being put into implementing it for realistic calculations of material properties of solids [2]. This method is exact in the limit of infinite lattice coordination [3] and describes correctly local correlations including magnetically *ordered* phases [4,5]. However, many applications require generalizations of the DMFT to capture short-range correlations, in the absence of broken symmetries. This is an active area of research and several methods have already been put forward [1,6–10].

The idea behind cluster methods is to treat local (cluster) degrees of freedom exactly while replacing the remaining degrees of freedom with a bath of noninteracting electrons which hybridize with the cluster so that the translation invariance is restored. The basic difficulty here is determining a suitable self-consistency condition without generating nonphysical solutions violating causality [11].

In this paper we propose an alternative generalization of DMFT, which can easily be formulated in real space, and in an arbitrary basis set, and which has excellent convergence properties as a function of cluster size. DMFT can be viewed as an extension of the coherent potential approximation (CPA), and several approaches to incorporate $\mathbf{k}$ dependence in the self-energy have been considered [11,12] in this framework. Our proposal can be viewed as an extension of those generalizations of CPA for *interacting systems*. We prove two central points: (I) The DMFT construction [1] can be formulated in a large class of basis sets. This observation frees us from the need to introduce sharp boundaries in real space. This approach is inspired by ideas from electronic structure, in which one achieves a cellular description by means of orbitals which can have a variable spatial extension. (II) This cellular DMFT (CDMFT) construction is manifestly causal, i.e., the self-energies that result from the solution of the cluster equations obey $\mathrm{Im}\,\Sigma(k,\omega) \leq 0$, eliminating *a priori* one of the main difficulties encountered earlier in devising practical cluster schemes.

It is useful to separate the three essential elements of a cluster scheme: (a) The definition of the cluster degrees of freedom, which are represented by impurity degrees of freedom in a bath described by a Weiss field matrix function $\hat{G}_0$. The solution of the cluster embedded in a medium results in a cluster Green's function matrix and a cluster self-energy matrix. (b) The expression of the Weiss field in terms of the Green's function or the self-energy of the cluster, i.e., the self-consistency condition of the cluster scheme. (c) The connection between the cluster self-energy and the self-energy of the lattice problem. The impurity solver estimates the local correlations of the cluster, while the lattice self-energy is projected out from the cluster self-energy by exploiting the use of additional information, i.e., the periodicity of the original lattice.

Our construction applies to very general models. It should be thought of as an extension of the band structure formalism in the Korringa-Kohn-Rostocker formulation that takes into account the electron-electron interactions. The lattice Hamiltonian, $H[f_{i\sigma}, f^{\dagger}_{i\sigma}]$, (one example could be the well-known Hubbard Hamiltonian), is expressed in terms of creation and annihilation operators $f_{i\sigma}$ and $f^{\dagger}_{i\sigma}$, where i runs over the sites of a d-dimensional infinite lattice $i = (i_1, \ldots, i_d)$ and the index σ denotes an internal degree of freedom such as a spin index or a spin orbital or band index if we consider an orbitally degenerate solid.

(a) Selection of cluster variables.—The first step in a mean field approach to a physical problem is a selection of a finite set of relevant variables. We do that by choosing a tight-binding basis for supercells with translation vectors R_n, i.e., $|R_n\alpha\rangle$ partially localized around R_n with $\alpha = 1, \ldots, N$ denoting an internal *cluster* index. The relation between the new wave functions, $|R_n\alpha\rangle$, and the old wave functions, $|i\sigma\rangle$, is encoded in a transformation matrix, $S_{R_n\alpha,i\sigma}$, such that $|R_n\alpha\rangle = \sum_{i\sigma} |i\sigma\rangle S^{-1}_{i\sigma,R_n\alpha}$. Because of the translation symmetry of the lattices, we obtain $S_{R_n\alpha,i\sigma} = S_{\alpha\sigma}[r(i) - R_n]$, where $r(i)$ is the position of site i. The creation and annihilation operators of the new basis are related to the operators of the old basis by $c_{R_n\alpha} = \sum_{i\sigma} S_{R_n\alpha,i\sigma} f_{i\sigma}$, and the operators that contain the "local" information that we want to focus our attention on are $c_\alpha \equiv c_{(R_n=0)\alpha}$, i.e., the operators of the cluster at the origin. We will refer to these operators as the cluster operators. Note that we do not require that the wave function basis be orthogonal, and the nonorthogonality is

summarized in an overlap matrix $O^{mn}_{\mu\nu} = O_{\mu\nu}(R_m - R_n) \equiv \langle R_m\mu \,|\, R_n\nu\rangle$.

The next step is to express the Hamiltonian in terms of the complete set of operators $c_{R_m\mu}$. In terms of the new set of variables it has the form

$$H = -\sum_{R_m\mu R_n\nu} t_{\mu\nu}(R_m - R_n) c^+_{R_m\mu} c_{R_n\nu} + \sum_{R_1\mu R_2\nu R_3\rho R_4\varsigma} U_{\mu\nu\rho\varsigma}(\{R_i\}) c^+_{R_1\mu} c^+_{R_2\nu} c_{R_4\varsigma} c_{R_3\rho}\,. \quad (1)$$

Equation (1) is a general Hamiltonian of electrons in a solid in some tight-binding nonorthogonal basis. The Hamiltonian is now split into three parts, $H = H_c + H_{cb} + H_b$ where H_c involves only the cluster operators, H_b contains $c_{R_n\mu}$ with $R_n \neq 0$ only and plays the role of a "bath," and finally H_{cb} contains both $c_{R_n\mu}$ with $R \neq 0$ and the cluster operators c_μ. Physically H_{cb} couples the cluster with its environment. A similar separation can be carried out at the level of the action, in the coherent state functional integral formulation of this problem, where the partition function and the correlation functions are represented as averages over Grassmann variables,

$$Z = \int \prod_{R_n\alpha} D c^+_{R_n\alpha} D c_{R_n\alpha} e^{-S}, \quad (2)$$

where the action is given by

$$S = \int_o^\beta d\tau \left(\sum_{R_m\mu R_n\nu} c^+_{R_m\mu} O^{mn}_{\mu\nu} \partial_\tau c_{R_n\nu} - H[c^+_{R_m\mu}, c_{R_n\nu}] \right) \equiv S_c + S_{cb} + S_b\,. \quad (3)$$

The effective action for the cluster degrees of freedom is obtained conceptually by integrating out all the variables $c_{R_n\mu}$ with $R_n \neq 0$ in a path integral to obtain an effective action for the cluster variables c_μ, i.e.,

$$\frac{1}{Z_{\rm eff}} e^{-S_{\rm eff}[c^+_\mu c_\mu]} \equiv \frac{1}{Z} \int \prod_{R_m\neq 0,\mu} D c^+_{R_m\mu} D c_{R_m\mu} e^{-S}. \quad (4)$$

Note that the exact knowledge of $S_{\rm eff}$ allows us to calculate *all the local* correlation functions involving cluster operators. As described in [1], this cavity construction, if carried out exactly, would generate terms of arbitrarily high order in the cluster variables. Two approximations are done at this stage. All interactions whose range goes beyond the extent of the cluster are neglected since only $U_{\mu\nu\rho\varsigma}(\{0\})$. Furthermore, the renormalization of the quartic and higher order terms in the effective action are not taken into account. The extended dynamical mean field approach introduced in Ref. [10] was designed to improve precisely this aspect of the cavity construction, and these improvements can be combined with the suggestions in this paper. The numerical results discussed below, however, suggest that these corrections are numerically small, at least when the range of the interactions is smaller than the cluster size, making this method relevant for the treatment of interactions beyond the on-site Hubbard interaction. Since the action S_{cb} contains only boundary terms, the effects of these operators will decrease as the size of the cluster increases. Our numerical results below assume that they are not important in models with short-range interactions, even in small clusters. Within these assumptions, the effective action is parametrized by $G_{0,\mu\nu}(\tau - \tau')$, the Weiss function of the cluster, and has the form

$$S_{\rm eff} = -\int_0^\beta d\tau d\tau' \sum_{\mu\nu} c^+_\mu(\tau) G^{-1}_{0,\mu\nu}(\tau - \tau') c_\nu(\tau') + \int_0^\beta d\tau_1\, d\tau_2\, d\tau_3\, d\tau_4 \times \Gamma_{\mu\nu\rho\varsigma} c^+_\mu(\tau_1) c^+_\nu(\tau_2) c_\varsigma(\tau_4) c_\rho(\tau_3)\,, \quad (5)$$

where $\Gamma_{\mu\nu\rho\varsigma} = U_{\mu\nu\rho\varsigma}(\{0\})$. By using the effective action (5), one can calculate the Green's functions of the cluster $G_{c,\mu\nu}(\tau - \tau')[\hat{G}_0] \equiv -\langle T_\tau c_\mu(\tau) c^+_\nu(\tau')\rangle[\hat{G}_0]$ and the cluster self-energies

$$\hat{\Sigma}_c \equiv \hat{G}_0^{-1} - \hat{G}_c^{-1}. \quad (6)$$

(b) Self-consistency condition.—The cluster algorithm is fully defined once a self-consistency condition which indicates how $\hat{G}_0$ should be obtained from $\hat{\Sigma}_c$ and $\hat{G}_c$ is defined. In the approach that we propose here the self-consistent equations become matrix equations expressing the Weiss field in terms of the cluster self-energy matrix $\hat{\Sigma}_c$.

$$\hat{G}_0^{-1} = \left(\sum_k \frac{1}{(i\omega + \mu)\hat{O}(k) - \hat{t}(k) - \hat{\Sigma}_c} \right)^{-1} + \hat{\Sigma}_c\,, \quad (7)$$

where $\hat{O}(k)$ is the Fourier transform of the overlap matrix, $\hat{t}(\mathbf{k})$ is the Fourier transform of the kinetic energy term of the Hamiltonian in Eq. (1), and $\mathbf{k}$ is now a vector in the reduced Brillouin zone (reduced by the size of the cluster). Equations (5) and (7) can be derived by scaling the hopping between the supercells as the square root of the coordination raised to the power of the Manhattan distance between the supercells and generalizing the cavity construction of the DMFT [1] from scalar to matrix self-energies. An important difference between Eq. (7) and the DCA equations [7] is that they yield cluster self-energies which are not diagonal in the cluster momenta. A detailed comparison between CDMFT and DCA will be presented elsewhere [13].

(c) Connection to the self-energy of the lattice.—In CDMFT the lattice self-energy does not participate in the self-consistent equations, and has to be estimated from the cluster self-energy, $\hat{G}_c$ and $\hat{\Sigma}_c$, using additional information, such as conservation laws and translation invariance. The simplest estimator uses the transformation matrix $S_{R_m\alpha,i\sigma}$ and is given by the equation

$$\Sigma_{lat,\sigma\sigma'}(k,\omega) = \sum_{\mu\nu} \tilde{S}^\dagger_{\sigma,\mu}(k) \Sigma_{c,\mu\nu}(\omega) \tilde{S}_{\nu,\sigma'}(k)\,, \quad (8)$$

where $\tilde{S}$ is the Fourier transform of the matrix S with respect to the original lattice indices i. Once a basis set

and a cluster shape are specified, improved estimates can be obtained to minimize finite size effects [13].

(d) Connection to impurity models.—As in single-site DMFT it is very convenient to view the cluster action as arising from a Hamiltonian,

$$H_{\mathrm{imp}} = \sum_{\rho\varsigma} \hat{E}_{\rho\varsigma} c_\rho^+ c_\varsigma + \sum_{\mu\nu\rho\varsigma} \Gamma_{\mu\nu\rho\varsigma} c_\mu^+ c_\nu^+ c_\rho c_\varsigma + \sum_{k\lambda} \epsilon_{k\lambda} a_{k\lambda}^+ a_{k\lambda} + \sum_{k\lambda,\mu} (V_{k\lambda,\mu} a_{k\lambda}^+ c_\mu + \mathrm{H.c.}). \tag{9}$$

Here $\epsilon_{k\lambda}$ is the dispersion of the auxiliary band and $V_{k\lambda,\mu}$ are the hybridization matrix elements describing the effect of the medium on the impurity. When the band degrees of freedom are integrated out, the effect of the medium is parametrized by a hybridization function,

$$\Delta_{\mu\nu}(i\omega_n)[\epsilon_{k\lambda}, V_{k\lambda}] = \sum_{k\lambda} \frac{V_{k\lambda,\mu}^* V_{k\lambda,\nu}}{i\omega_n - \epsilon_{k\lambda}}. \tag{10}$$

The hybridization function is related to the Weiss field function by expanding Eq. (7) in high frequencies:

$$\hat{G}_0^{-1}(i\omega_n) = i\omega_n \bar{O} - \hat{E} - \hat{\Delta}(i\omega_n) \tag{11}$$

with $\bar{O} = [\sum_k \hat{O}_k^{-1}]^{-1}$ indicating that the impurity model has been written in a nonorthogonal local basis with an overlap matrix $\bar{O}$.

Let us now consider some examples of this approach.

(i) Single-site DMFT.—The simplest example is the single-site dynamical mean field theory which is exact in the limit of infinite dimensions. In this case the cluster is just a single site denoted by 0, and the cluster operators are the creation and annihilation operators of that site, $c_{0\sigma}^+, c_{0\sigma}$. The cluster Hamiltonian is diagonal in the spin variables and reduces to the effective action of the Anderson impurity model. The next step is a scalar equation $\hat{G}_0^{-1} = \hat{G}_c^{-1}[G_0] + \hat{\Sigma}_c[G_0]$. Finally the last step identifies the self-energy of the cluster with the lattice self-energy.

(ii) Free cluster.—The next example is a free cluster scheme for the one-band Hubbard model. The method divides the lattice into supercells, and views each supercell as a complex "site" to which one can apply ordinary DMFT. Here R_n is the supercell position and α labels the different sites within the unit cell and the spin. Introducing a spin label σ and a supercell notation where an atom is denoted by the supercell, R_n, and the position inside the supercell, l, $\alpha = (\sigma', l)$ and $S_{R_n\alpha,i\sigma} = \delta_{\sigma,\sigma'}\delta_{R_n+l,r_i}$ is diagonal in spin and position. In this case the overlap matrix is the identity. This real space cluster method was investigated using quantum Monte Carlo methods by Katsnelson and Lichtenstein [14].

(iii) Multiorbital DMFT in a nonorthogonal basis.—Another important special case for our general construction is the implementation of single-site DMFT in a nonorthogonal basis. In this case the supercell is a single site, but the wave functions defining the cluster operators are chosen so that they are very localized in real space.

An implementation of this method, in conjunction with a generalization of the interpolative perturbation theory, as an impurity solver, has resulted in new advances in the theory of plutonium [2]. Here the flexibility in the choice of basis is crucial for the success of the DMFT program. DMFT neglects from the start the interactions which are not on site. A high degree of localization requires a nonorthogonal basis and the formalism introduced in this Letter.

(d) Other bases.—Finally we point out that the most attractive feature of this method is that it would allow its formulation in terms of wave functions which are partially localized in real and momentum space like wavelet functions. This flexibility is most appealing for treating problems such as the Mott transition, where both the particlelike and the wavelike aspects of the electron need to be taken into account, requiring a simultaneous consideration of real and momentum space.

We now prove that the CDMFT approach gives manifestly causal Green's functions. For this we assume that we start the DMFT iteration with a guess for the bath function $\hat{\Delta}$ which is causal. The self-energy which is generated in the process of solving the "impurity model" is also causal. Furthermore, any sensible approximation technique used to compute the self-energy of the cluster respects causality, so our proof is valid not just for exact solutions of the CDMFT scheme but also for approximate solutions as long as the impurity solvers used in the solution of the cluster impurity problem preserve causality. The next step is to show that, if a causal self-energy is introduced in the self-consistency condition [Eq. (7)], the resulting bath function $\hat{\Delta}$ is causal. Since both $\hat{\Sigma}_c$ and $\hat{\Delta}$ are matrices, the causality condition needs to be formulated precisely. A fermionic matrix function, $A(\omega)$, is causal if it is analytic in the upper half of the complex frequency plane and has a spectral representation with the positive-definite spectral density matrix $\frac{-1}{2\pi i}\{A(\omega) - A^\dagger(\omega)\}$. It is easy to see that the DMFT equation leads to the correct analytic properties, and the following proof establishes the positivity of the bath spectral density. By writing $\Sigma_R = \epsilon - i\gamma$ with ϵ, γ Hermitian and γ positive definite, we get

$$(\hat{\Delta}_R^\dagger - \hat{\Delta}_R) = -2i\gamma + \sqrt{\gamma}\left(\left\{\sum_k \frac{1}{i + d_k}\right\}^{-1} - \left\{\sum_k \frac{1}{-i + d_k}\right\}^{-1}\right)\sqrt{\gamma}. \tag{12}$$

Positivity is reduced to proving that the following matrix is negative [15], i.e.,

$$2 - \left\{\sum_k \frac{1}{1 - id_k}\right\}^{-1} - \left\{\sum_k \frac{1}{1 + id_k}\right\}^{-1} \le 0. \tag{13}$$

Here $\omega\hat{O}(k) - \hat{t}(k) - \epsilon \equiv \sqrt{\gamma}\, d_k \sqrt{\gamma}$. By performing a change of variables $d_k = i\,\frac{e^{i\theta_k}+e^{-i\theta_k}}{e^{i\theta_k}-e^{-i\theta_k}}$ with θ_k, a Hermitian matrix, Eq. (13) reduces to proving that

$$1 \leq \min_x \frac{\langle x|\,(1-z)^{-1} + (1-z^\dagger)^{-1}\,|x\rangle}{\langle x\,|\,x\rangle}. \qquad (14)$$

where $z \equiv \langle\langle e^{-2i\theta_k}\rangle\rangle$, with $\langle\langle \ldots\rangle\rangle$ denoting an average over the Brillouin zone. By performing the substitution $|x\rangle = (1-z)\,|\,y\rangle$, Eq. (14) reduces to

$$1 \leq \min_{||y||=1} \frac{2 + \langle y|z + z^\dagger|\,y\rangle}{1 + \langle y|z^\dagger z|y\rangle + \langle y|z + z^\dagger|\,y\rangle} \qquad (15)$$

which clearly holds due to the fact that z is an average of unitary matrices, and has the property $||z^\dagger z|| \leq 1$. From Eqs. (15) and (8) it follows that the imaginary part of the retarded self-energy is always less or equal to zero, completing the proof of causality. This proof generalizes Ref. [7] from scalar to matrix CPA equations.

Convergence properties.—This cluster formulation does not introduce any discontinuities in momentum space, suggesting that it will have superior convergence properties as a function of cluster size. We tested this hypothesis in a very simple model, a one-dimensional version of the model introduced by Affleck and Marston [16] in its uniform phase. This model contains an SU(N) nearest-neighbor spin-spin Heisenberg interaction with strength J/N and chemical potential μ to control the density and a nearest-neighbor hopping term with strength t. This model is exactly soluble in the large N limit, allowing a simple test of the convergence properties of the solution of the CDMFT equations. In Fig. 1 the convergence of the fermion equal time correlation function $\langle f^\dagger_{i\sigma} f_{j,\sigma}\rangle$, for i, j nearest neighbors, is plotted for various CDMFT cluster sizes, and the fast approach to the infinite cluster answer (dotted line) is indicated. More extensive studies are consistent with this rapid convergence of the CDMFT scheme and will be published elsewhere [13], together with comparisons with other cluster methods.

In conclusion, DMFT has produced a wealth of information on problems where the physics is local, and cluster methods promise to be equally fruitful in more complex problems where correlations between more sites and orbitals need to be taken into account. All the techniques which have been used for the solution of the single-site DMFT are applicable to this cluster extension. In this paper we limited ourselves to states without broken symmetries; further extensions of the cellular DMFT approach to states with broken symmetries as well as solutions of the CDMFT equations using exact diagonalization techniques are currently under investigation [17].

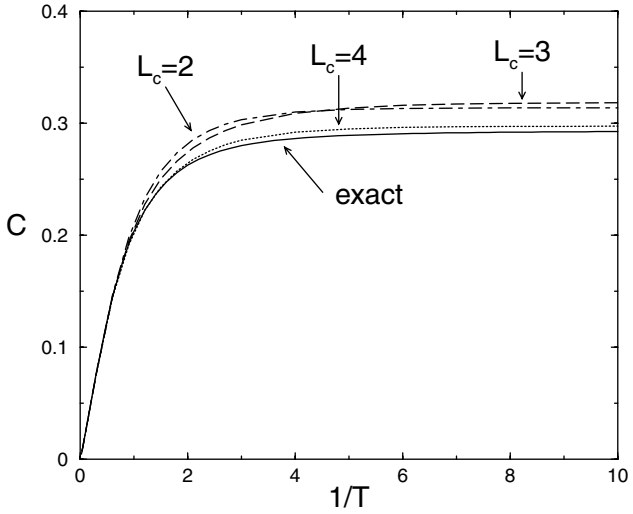

FIG. 1. The correlation function C is plotted as a function of the inverse temperature for $\mu = 1$ and $t = 1$. The solid line is the exact solution. The dot-dashed, dashed, and dotted lines are, respectively, the CDMFT predictions for two-, three-, and four-site clusters.

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