SPECTRAL DENSITY FUNCTIONAL APPROACH TO ELECTRONIC CORRELATIONS AND MAGNETISM IN CRYSTALS

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Abstract  A novel approach to electronic correlations and magnetism of crystals based on realistic electronic structure calculations is reviewed. In its simplest form it is a combination of the “local density approximation” (LDA) and the dynamical mean field theory (DMFT) approaches. Using numerically exact QMC solution to the effective DMFT multi-orbital quantum-impurity problem, a successful description of electronic structure and finite temperature magnetism of transition metals has been achieved. We discuss a simplified perturbation LDA+DMFT scheme which combines the T-matrix and fluctuation-exchange approximation (TM-FLEX). We end with a discussion of cluster generalization of the non-local DMFT scheme and its applications to the magnetism and superconductivity of high-T_c superconductors.
1. Introduction

The theory of electronic structure and magnetism of solids historically was split into two distinct parts, namely, the model investigations of many-body effects and the calculations of the energy spectra and properties of specific compounds in the framework of density functional (DF) scheme [1, 2]. Recently, within the dynamical mean-field theory (DMFT, for a review see Ref. [3]) the correlation effects have been incorporated into realistic electronic structure calculations [4-10]. This method has been successfully applied to a number of classical problems of solid state physics such as the finite-temperature magnetism of iron-group metals [11], $\alpha - \delta$ transition in plutonium [12], electronic structure of doped Mott insulators [9]. In contrast with standard DF theory, in this new approach known as “LDA+DMFT” [4] or “LDA++” [5] the total energy of the system (or, more accurately, the thermodynamic potential $\Omega$) is considered as a functional of the Green function instead of the density matrix [7, 8, 13, 14]. To stress this new feature more explicitly we will use the term “spectral density functional (SDF)”. The analytical properties of the Green function guarantee that the knowledge of the spectral density is equivalent to the knowledge of the time-dependent Green function whereas the density matrix is just static value of the latter [15]. Here we will describe the basic ideas of the SDF method, both in the framework of a standard DMFT and from a more general point of view, discuss possible cluster generalizations of the DMFT, and consider the applications of SDF to correlation effects and magnetism in transition metals.

2. Dynamical Mean Field Theory: an effective action perspective

A most economical approach to unifying the various dynamical mean-field approximations in use, is provided by the effective action construction [16]. The idea is to select a set of variables which is relevant to the physics of the problem and to write down a functional of the relevant variables. The extremum of this functional yields the values of those variables in equilibrium, and the value of this functional at stationarity gives the free energy of the system in equilibrium.

Density functional theory is the simplest example of this construction [17], here the total energy of the solid is expressed in terms of the density of the electrons. Another well tested example is the spin density functional theory in which the total energy is expressed in terms of the spin and charge densities. The construction of an explicit expression of the
exact effective-action functional is usually not available except for the case of very simple examples and the success of the method relies on the availability of good approximations to this functional. The LDA and the LSDA approaches have been extraordinarily successful for weakly correlated systems.

The dynamical mean-field approach to model Hamiltonians of strongly correlated systems on a lattice can also be brought to such an effective action perspective. More important this construction can be easily generalized to incorporate the so-called extended dynamical mean-field approach (E-DMFT) [18], by constructing an effective action for both the local Green function and the local density-density and local spin-spin autocorrelation function. The effective-action approach also allows the formulation of the E-DMFT approach in the case where spatial and spin or charge symmetries are broken. [8, 19].

The effective-action approach, allows us a simple combination of the density functional theory and the DMFT. By Legendre transformation techniques one can construct a functional of the density and of the local spectral function of the heavy orbitals whose extremization would yield the exact density local spectral function of the heavy orbitals, and the total energy of the system. Again the exact form of this functional are not known in explicit form, but useful approximations to it are available, and can be used to study interesting problems.

This realistic DMFT, LDA+DMFT approach or LDA++ approach, was first implemented ignoring the coupling between the density and the local spectral function. This amounts to performing first a LDA calculation to derive a tight-binding model Hamiltonian, and then performing a DMFT calculation for the spectra of the DMFT Hamiltonian. This is very close in spirit to the philosophy of model Hamiltonian calculations. Recently a full implementation of the self-consistent determination of the density and the local spectra was carried out by S. Savrasov [12], and this is now closer in spirit to traditional first-principles electronic structure calculations.

The effective action approach, can be generalized to clusters, if short-range correlations need to be taken into account. In the context of model Hamiltonians, a functional of the restriction of the Green function to a given cluster is defined. The extremization of this functional give rise to cluster dynamical mean field equations. There is no difficulty in constructing hybrid functionals for the self-consistent determination of both the density and the cluster Green functions, namely a CDMFT+LDA method.

In the next section we motivate the approximate DMFT form of those functionals from a perspective of a reduced fermionic description.
3. Fermionic reduced description and dynamical mean-field theory

In the SDF approach two-electron subsystems are introduced, one of them is described by the standard DF theory (usually this is the subsystem of \(sp\)-electrons) and dynamical interelectron correlations are taken into account for the another one (usually this is the subsystem of \(d\)- or \(f\)-electrons). Further simplifications can be connected with the local form of interelectron interactions (only Hubbard-type on-site electron correlations are considered), using some approximations like DMFT or its cluster generalizations [22, 23, 24], etc. From a general point of view all these approaches can be considered as specific cases of a “coarse graining” (reduced description) ideology [25] when all the variables describing the system can be separated into the “gross” and “slave” variables; the only assumption that a closed set of the equations of motion for the gross variables exist is sufficient to find an explicit form of these equations [26]. Here we demonstrate the coarse graining procedure for fermionic degrees of freedom for the system of interacting electrons in a crystal.

Let us start with the functional integral over the Grassman creation and annihilation electron fields \(c^+, c\), where the “measure” is \(\exp (-S)\),

\[
S = -\int dx \int_0^\beta d\tau \left[ c^+ (x, \tau) \left( \frac{\partial}{\partial \tau} + \mu - H_0 \right) c (x, \tau) \right] + S_{\text{int}} [c^+, c] \tag{1}
\]

Here \(\mu\) is the chemical potential, \(\beta = 1/T\) is the inverse temperature, and \(H_0\) is the “one-particle” part of the Hamiltonian. In the SDF approach

\[
H_0 = -\nabla^2 / 2 + V_{KS} \tag{2}
\]

where \(V_{KS}\) is the Kohn-Sham self-consistent potential [1].

In the spirit of a reduced description approach [25] we introduce “gross” variables \(d = f \cdot c\) (more explicitly, \(d_\mu = \sum_j f_{\mu j} c_j\) where \(j\) are site indices; e.g., in a standard DMFT \(f_{\mu j} = \delta_{\mu 0} \delta_{j0}\)). The effective action for the gross variables \(S_{\text{eff}}\) is defined by introducing a delta-functional \(\delta (d - f \cdot c) \delta (d^+ - f^* \cdot c^+)\) [27] into the functional integral.
\[ \exp(-S_{\text{eff}}) = \frac{1}{Z} \int DcDc^+ \exp(-S) * \int D\lambda D\lambda^+ \exp \left[ i\lambda (d^+ - f^* \cdot c^+) + i\lambda^+ (d - f \cdot c) \right] \] (3)

Unfortunately, this functional integral cannot be estimated exactly and approximations are needed. Here we consider only the simplest approximation to the derivation of the effective action, we replace \( S_{\text{int}}[c^+,c] \rightarrow \tilde{S}_{\text{CG}}[d^+,d] \) but compensate for the omission of the interaction terms away from those in the gross variables, namely in the medium by adding a medium self-energy in the standard DMFT

\[ S \rightarrow \tilde{S} = S + \sum_{i \geq 0} \int \int d\tau d\tau' c^+ (\tau,i) \Sigma (\tau - \tau',i,i) c (\tau',i) \] (4)

In other words, the DMFT approach treats exactly the interaction terms \textit{only in} those part of the interaction Lagrangian which can be written in terms of gross variables, while \textit{the rest of the interaction terms are handled in a gaussian approximation} by replacing the interaction terms by a medium self-energy.

Now the effective measure for the gross variables is given by

\[ \exp(-S_{\text{eff}}) = \frac{1}{Z} \int DcDc^+ \exp(-\tilde{S}_{\text{CG}} + d^+ \Sigma d) * \int D\lambda D\lambda^+ \exp \left[ i\lambda (d^+ - f^* \cdot c^+) + i\lambda^+ (d - f \cdot c) \right] \] (5)

where \( Z = \int DcDc^+ \exp(-\tilde{S}_{\text{CG}}) \), \( \tilde{S}_{\text{CG}} = S_{\text{med}} + S_{\text{int}}[d^+,d] \).

Passing to the Matsubara frequencies [15] one has

\[ S_{\text{med}} = -T \sum_{\omega_n,j} c^+ (i\omega_n,j) [i\omega_n + \mu - H_0 - \Sigma (i\omega_n)] c (i\omega_n,j) \] (6)

In the standard DMFT scheme the correlations between \( d(f) \)-electrons on a central site \( j = 0 \) are treated exactly while the correlations in the medium are treated in the Gaussian approximation. It is sometimes stated in the literature that this assumption violates the translational invariance of the system. The effective-action approach, is meant to construct a functional of the selected variables, so by construction it is not possible to discuss within this framework the issue of translation
invariance. The “self-consistency condition” does the best possible job within a Gaussian approximation to restore the equivalence of the central site and the medium.

We show below that this reasoning in a cluster setting leads to the “cellular DMFT” approach \[24\]. In principle, the correlations between the gross and slave variables (e.g., the long-range part of the Coulomb interaction) also can be taken into account in a close analogy with the classical spin models \[25\]; the corresponding modification of the DMFT approach will be considered elsewhere. In accordance with the general scheme of the non-equilibrium statistical operator method \[26\], we add an “auxiliary field” conjugated to the Green function of the gross variables, namely, the term \(d^+ \bar{\Sigma} d\). These fields will restore the translational invariance for the fermionic Green function (see below, Eq. (11)).

Calculating the Gaussian integral over \(c, c^+\) one has

\[
\exp (-S_{\text{eff}}) = \exp \left( -S_{\text{int}}^{CG} [d^+, d] + d^+ \Sigma d \right) * \\
\int D\lambda D\lambda^+ \exp \left[ i\lambda d^+ + i\lambda^+ d - \lambda^+ f \cdot G_{\text{med}} \cdot f^* \lambda \right]
\]

where the medium Green function reads

\[
G_{\text{med}} = [i\omega_n + \mu - H_0 - \Sigma (i\omega_n)]^{-1}.
\]  

(8)

At last, calculating the Gaussian integral over \(\lambda, \lambda^+\) we obtain

\[
S_{\text{eff}} [d^+, d] = S_{\text{int}}^{CG} [d^+, d] - T \sum_{\omega_n} d^+ (i\omega_n) \, G_0^{-1} (i\omega_n) \, d (i\omega_n)
\]

(9)

where

\[
G_0^{-1} (i\omega_n) = (f \cdot G_{\text{med}} \cdot f^*)^{-1} + \bar{\Sigma}
\]

(10)

The self-consistency condition that determines \(\Sigma\) requires the equality of the local Green function computed from the reduced description and from the medium agree, i.e.,

\[
\langle Td(\tau)d^+(\tau') \rangle_S = \langle Td(\tau)d^+(\tau') \rangle_{S_{\text{eff}}}
\]

(11)

Further, \(\langle Tc(\tau)c^+(\tau') \rangle_S = G_{\text{med}}\) (because of the replacement \(S_{\text{int}} [c^+, c] \rightarrow S_{\text{int}}^{CG} [d^+, d]\)) and

\[
\langle Td(\tau)d^+(\tau') \rangle_S = f \cdot G_{\text{med}} \cdot f^*
\]

(12)

On the other hand, \(\langle Td(\tau)d^+(\tau') \rangle_{S_{\text{eff}}} = \mathcal{G}\) satisfies the Dyson equation

\[
\mathcal{G}^{-1} = \mathcal{G}_0^{-1} - \Sigma_{CG} = (f \cdot G_{\text{med}} \cdot f^*)^{-1} + \bar{\Sigma} - \Sigma_{CG}
\]

(13)
and we have $\bar{\Sigma} = \Sigma_{CG}$ where $\Sigma_{CG}$ is a “coarse-grained” self-energy. The most natural choice is $\Sigma_{CG} = f^* \cdot \Sigma \cdot f$.

To obtain the main equation of the DMFT [3] we have to choose $f$ as a projection operator on the central site. In this case Eq.(13) will take the desired form

$$G^{-1}_0(i\omega_n) = \left[ \sum_k \frac{1}{i\omega_n + \mu - t(k) - \Sigma(i\omega_n)} \right]^{-1} + \Sigma (i\omega_n)$$

(14)

where the quasimomentum $k$ runs the Brillouine zone, $t(k)$ is the Fourier transform of the Hamiltonian $H_0$ projected into the subspace of $d(f)$-electrons.

To consider possible cluster generalizations one can choose $f = PU^+ P$ where $P$ is the projection operator on the cluster and $U$ is an unitary transformation of the variables in this cluster. Thus our result reads:

$$G^{-1}_0(i\omega_n) = \left[ \frac{(L_c)}{L} \sum_{k_c} U^+ \frac{1}{i\omega_n + \mu - t(k_c) - \Sigma} U \right]^{-1} + U \Sigma U^+$$

(15)

where $k_c$ runs the “new” supercell Brillouin zone, $L$ and $L_c$ are the sizes of the crystal and cluster, correspondingly. In terms of $\Sigma_{CG} = U \Sigma U^+$,

$$G^{-1}_0(i\omega_n) = \left[ \left( \frac{L_c}{L} \right) \sum_{k_c} \frac{1}{i\omega_n + \mu - Ut(k_c) U^+ - \Sigma_{CG}} \right]^{-1} + \Sigma_{CG}$$

(16)

As it was shown in Ref. [24] this equation for general basis set can be very useful in the optimisation of interaction problem within the cluster or cellular DMFT scheme. Another version of the cluster DMFT [23] will be described below.

4. Spectral density versus density functionals

In a standard DF theory the thermodynamic potential for non-correlated conduction-“c” electrons $\Omega^c$ is represented as a functional of the electron density $\rho(r)$ which is, generally speaking, a matrix in spin indices. Formally it can be represented as a thermodynamic potential of the Kohn-Sham quasiparticles [1], $\Omega_{sp}$, minus the contribution of the so called “double counted” terms, $\Omega_{dc}$:

$$\Omega^c = \Omega_{sp} - \Omega_{dc}$$

$$\Omega_{sp} = -Tr \log[i\omega + \nabla^2/2 - V_{KS}]$$

$$\Omega_{dc} = \int V_{KS}(r)\rho(r)dr - \int V_{ext}(r)\rho(r)dr - \frac{1}{2} \int \frac{\rho(r)\rho(r')}{|r-r'|}drdr' - E_{xc}[\rho]$$

(17)
where \( T_r = T_{r, \omega i L \sigma}, T_{r, \omega} \) is the sum over Matsubara frequencies \( T_{r, \omega} = T \sum_{\omega} e^{i \omega (0^+)} \), \( \omega = \pi T (2n + 1), n = 0, \pm 1, \ldots \), \( T \) is the temperature, and \( i L \sigma \) are site numbers \( (i) \), orbital quantum numbers \( (L = l, m) \) and spin projections \( \sigma \), respectively, \( V_{ext}(r) \) is the external potential, \( E_{xc}[\rho] \) is the exchange-correlation energy, and the Kohn-Sham effective potential is defined as

\[
V_{KS}(r) = V_{ext}(r) + \int \frac{\rho(r')}{|r - r'|} dr' + \frac{\delta E_{xc}[\rho]}{\delta \rho(r)}.
\]

(18)

In contrast with the standard density functional theory, the SDF approach deals with the real dynamical quasiparticles for correlated “d-electrons” defined via local Green functions rather than with Kohn-Sham “quasiparticles” which are, strictly speaking, only auxiliary states to calculate the total energy. Therefore, instead of working with the thermodynamic potential \( \Omega \) as a density functional we have to start from its general expression in terms of an exact Green function [28]

\[
\Omega^d = \Omega_{sp}^d - \Omega_{dc}^d
\]

\[
\Omega_{sp}^d = -Tr \left\{ \ln \left[ \Sigma - G_0^{-1} \right] \right\}
\]

\[
\Omega_{dc}^d = Tr \Sigma G - \Phi
\]

(19)

where \( G, G_0 \) and \( \Sigma \) are an exact Green function, its bare value and self-energy, \( \Phi \) is the Luttinger generating functional (sum of the all connected skeleton diagrams without free legs), respectively. A complete SDF thermodynamic potential is equal to \( \Omega = \Omega^c + \Omega^d \). We have to keep in mind also the Dyson equation

\[
G^{-1} = G_0^{-1} - \Sigma
\]

(20)

and the variational identity

\[
\Sigma = \frac{\delta \Phi}{\delta G}
\]

(21)

When neglecting the quasiparticle damping, \( \Omega_{sp} \) will be nothing but the thermodynamic potential of “free” fermions but with exact quasiparticle energies. Formal analogies between Eqs.(17) and (19), (18) and (21) are obvious: the self-energy plays the role of the Kohn-Sham potential (without the external potential) and the Green function plays the role of the density matrix. As an example of this correspondence one can prove [7] in the framework of the SDF an useful identity known as the “local
force theorem” basically in the same way as it has been done within DF theory [29, 30].

For both parts of the thermodynamic potential, $\Omega^c$ and $\Omega^d$, the local force theorem is based on extremum properties with respect to the variation of the density matrix and Green function, respectively. Further we will consider the contribution $\Omega^d$ (omitting the index $d$ for brevity). In principle, fermionic reduced description scheme allows us to combined conduction “c” and correlated “d” part of the total SDF: $\Omega = \Omega^c + \Omega^d$ [14].

5. Effective exchange interactions

Let us discuss the problem of calculation of effective exchange interactions ($J_{ij}$) in correlated systems. In principle the $J_{ij}$ parameters are not well defined for arbitrary magnetic systems, and the traditional way to study spin excitations related to the calculation of non-local frequency-dependent spin-susceptibility [3, 32]. In this case the auxiliary space-time dependent magnetic field is added to the Hamiltonian: $\sigma h(r, \tau)$ and the second derivative of the free-energy with respect to the magnetic field gives the interacting spin-susceptibility: $\chi^{-1} = \chi_0^{-1} - \Gamma$, where $\chi_0$ is an empty-loop susceptibility and $\Gamma$ is the vertex correction [3, 32]. Here we consider a simple approximation of “rigid spin rotation” of spectral density for a small angle:

$$\delta e_i = \delta \varphi_i \times e_i$$  \hspace{1cm} (22)

where $e_i$ is a general direction of constrained effective spin-dependent potential on site $i$ and $\delta \varphi_i$ is a rotation vector. In this case it is useful to write explicitly the spinor structure of the self-energy and Green functions:

$$\Sigma_i = \Sigma_i^c + \Sigma_i^s \sigma$$  \hspace{1cm} \hspace{1cm} (23)

$$G_{ij} = G_{ij}^c + G_{ij}^s \sigma$$

where $\Sigma_i^{(c,s)} = \frac{1}{2} \left( \Sigma_i^\uparrow + \Sigma_i^\downarrow \right)$, $\Sigma_i^s = \Sigma_i^s e_i$, $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ are Pauli matrices, $G_{ij}^c = \frac{1}{2} Tr_\sigma (G_{ij})$ and $G_{ij}^s = \frac{1}{2} Tr_\sigma (G_{ij} \sigma)$. We suppose that the bare Green function $G^0$ does not depend on spin direction and all the spin-dependent terms including the Hartree-Fock terms are incorporated in the self-energy. In the rigid spin approximation we assume that the unit vector $e_i$ does not depend on the energy and orbital indices and represents the direction of the average local magnetic moment on the site $i$. Note, that the thermodynamic potential $\Omega$ should be considered as a constrained SDF which depends on $e_i$ as on external parameters.
(cf. Ref. [33]). Then the variation of the thermodynamic potential with respect to small spin-rotation can be written as

$$\delta \Omega = \delta^* \Omega_{sp} + \delta_1 \Omega_{sp} - \delta \Omega_{dc}$$  \hspace{1cm} (24)

where $\delta^*$ is the variation without taking into account the change of the "self-consistent potential" (i.e. self-energy) and $\delta_1$ is the variation due to this change of $\Sigma$. Taking into account Eq. (21) it can be easily shown (cf. Ref. [28]) that

$$\delta_1 \Omega_{sp} = \delta \Omega_{dc} = TrG\delta \Sigma$$  \hspace{1cm} (25)

and hence

$$\delta \Omega = \delta^* \Omega_{sp} = -\delta^* Tr \ln \left[ \Sigma - G_0^{-1} \right]$$  \hspace{1cm} (26)

which is an analog of the "local force theorem" in the density functional theory [30].

In the case of rigid spin rotation the corresponding variation of the thermodynamic potential can be written as

$$\delta \Omega = V_i \delta \varphi_i$$  \hspace{1cm} (27)

where the torque $V_i$ is equal to

$$V_i = 2Tr_{\omega L} \left[ \Sigma_i^s \times G_{i\ell}^s \right]$$  \hspace{1cm} (28)

Based on the expansion of this expression (28) in a sum of pairwise contributions one can obtain [7] useful formula for the effective magnetic interactions:

$$J_{ij} = -Tr_{\omega L} \left( \Sigma_i^s \Sigma_j^s \Sigma_j^s \Sigma_i^s \Sigma_j^s \right)$$  \hspace{1cm} (29)

and, correspondingly, for the stiffness tensor of a ferromagnet:

$$D_{\alpha\beta} = -\frac{2}{M}Tr_{\omega L} \sum_k \left( \Sigma_i^s \frac{\partial G^\dagger_i(k)}{\partial k_\alpha} \Sigma_i^s \frac{\partial G^\dagger_j(k)}{\partial k_\beta} \right)$$  \hspace{1cm} (30)

where $M$ is the magnetic moment per unit cell. These results generalize the LSDA expressions of Ref. [30] to the case of correlated systems.

Note that passing from Eq.(28) to Eq.(29) is not accurate, since the exchange parameters are connected with the second variations of the $\Omega$-potential and use of the local force theorem can not be justified. Eq. (29) corresponds to the "empty loop" approximation neglecting the vertex corrections. At the same time, for the stiffness the latter are absent and Eq.(30) appears to be exact provided that the self-energy and three-leg vertex are local (as in the DMFT) [13].

The fact that vertex corrections to the spin stiffness are absent within DMFT, is also suggested by the analogy between electric and spin transport developed in Ref. [20]. The spin-wave stiffness can be obtained from
the zero-frequency limit of the spin conductivity. Within DMFT, the charge current-current correlation function does not require vertex corrections [21] and can therefore be obtained directly from the convolution of the one electron spectra. These arguments are independent of the spin structure, and can therefore be used for the spin-wave stiffness.

In order to elucidate the approximation behind the expression for the exchange parameters (Eq. (29)), we consider the energy of a spiral magnetic configuration with the rigid rotation of the spinor-electron operators by the polar angles $\theta$ and $\varphi$:

$$c_{im} \rightarrow U \left( \theta_i, \varphi_i \right) c_{im}$$

where

$$U \left( \theta, \varphi \right) = \begin{pmatrix} \cos \theta/2 & \sin \theta/2 \exp \left( -i\varphi \right) \\ -\sin \theta/2 \exp \left( i\varphi \right) & \cos \theta/2 \end{pmatrix}$$

(31)

assuming that $\theta_i = \text{const}$ and $\varphi_i = qR_i$ where $R_i$ is the site-lattice vector. Since we take into account only on-site correlation effects the interaction term in the Hamiltonian is invariant under that transformation, and the change of the Hamiltonian is

$$\delta H = \sum_{ij} Tr_{m\sigma} \left[ t_{ij} c_i^+ \left( U_i^+ U_j - 1 \right) c_j \right] = \delta_1 H + \delta_2 H$$

$$\delta_1 H = \sin^2 \frac{\theta}{2} \sum_k Tr_{m\sigma} \left[ \left( t(k+q) - t(k) \right) c_k^+ c_k \right]$$

$$\delta_2 H = \frac{1}{2} \sin \theta \sum_{ij} Tr_m \left[ t_{ij} c_i^+ c_j^\dagger \right] \left( \exp\left( i q R_i \right) - \exp\left( i q R_j \right) \right)$$

(32)

Consider further the case of small $\theta$, we can calculate change of the total energy to lowest order in $\theta$ corresponds to the first order in $\delta_1 H$ and the second order in $\delta_2 H$:

$$\delta E = \frac{\theta^2}{4} \left\{ \sum_k \left[ t(k+q) - t(k) \right] n_k - i Tr_m \int \frac{d^4k}{(2\pi)^4} \left[ t(k+q) - t(k) \right] * \gamma(k, q) G(k+q) [t(k+q) - t(k)] G^\dagger(k) \right\}$$

where $n_k = Tr_{m\sigma} \langle c_k^+ c_k \rangle$, $q$ is a four-vector with component $(q,0)$, and $\gamma$ is the three-leg vertex. Our main approximation is to neglect of the vertex corrections ($\gamma = 1$). In this case the previous equation takes the following form:

$$\delta E = - \frac{\theta^2}{4} Tr_m \{ i \int \frac{d^4k}{(2\pi)^4} \left[ t(k+q) - t(k) \right] *$$

(33)
\[ G_\uparrow (k+q) \left[ G_\uparrow^{-1} (k+q) - G_\uparrow^{-1} (k) + t (k+q) - t (k) \right] G_\uparrow (k) \]

Using the following consequence of the Dyson equation:

\[ t (k+q) - t (k) = G_\uparrow^{-1} (k) - G_\uparrow^{-1} (k+q) + \Sigma_\uparrow (E) - \Sigma_\downarrow (E) \]  

(34)

one can rewrite Eq. (33) in the form: \( \delta E = \frac{q^2}{T} [J (0) - J (q)] \) with the exchange integrals corresponding to Eq. (29). We conclude that the expression for \( J_{ij} \) is accurate if the vertex corrections can be neglected. Note that in the limit of small \( q \) this can be justified rigorously, provided that the self-energy and three-leg scalar vertex are local. Therefore, the expression for the stiffness constant of the ferromagnet (Eq. (30)) appears to be exact in the framework of DMFT [13].

It should be stressed that the exchange integrals discussed here are just static characteristics connected with energy of inhomogeneous spin configurations. They determine the frequencies of spin excitation in itinerant-electron systems only under adiabatic (rigid-spin) approximation. For more general consideration one should calculate \( q \)- and \( \omega \)-dependent spin susceptibility.

6. Electron correlations and finite-temperature magnetism in transition metals

Now we describe the applications of the SDF approach to a classical problem of finite-temperature magnetism of the iron-group transition metals. Despite a lot of attempts starting from seminal works by Heisenberg and Frenkel (for review of early theories see e.g. [31, 34, 35]) we have yet no complete quantitative theory describing their magnetic and spectral properties. The reason is that to describe the properties of Fe, Co, and Ni one has to solve the problem of taking into account moderately strong electronic correlations where approaches developed both for weakly correlated systems such as normal-group metals and to highly correlated systems such as Mott insulators are not, generally speaking, reliable. There were many attempts to introduce by some way correlation effects in band structure calculations of these metals [36, 37, 38, 39, 40], not referring to numerous purely model works. But the question of applicability of specific approximations such as lowest order perturbation theory [37, 40], moment method [39] or three-body Faddeev equations [38] is not clear and one needs a reliable approach which would be checked carefully for model systems and demonstrate its applicability for moderately correlated systems. It has been demonstrated in Ref. [11] that the \textit{ab initio} dynamical mean-field theory does give a very success-
ful description of both correlation effects in the electron energy spectra and the finite-temperature magnetic properties of Fe and, especially, Ni. Here we present the corresponding results.

We start with the LDA Hamiltonian in the tight-binding orthogonal LMTO representation \( H_{LDA}^{mm'}(k) \) [41], where \( m \) describes the orbital basis set containing 3d-, 4s- and 4p-states, and \( k \) runs over the Brillouin zone (BZ). The interactions are parameterized by a matrix of screened local Coulomb interactions, \( U_{mm'} \), and a matrix of exchange constants, \( J_{mm'} \), which are expressed in terms of two screened Hubbard parameters, \( U \) and \( J \), describing the average Coulomb repulsion and the interatomic ferromagnetic exchange, respectively. We use the values \( U = 2.3 \) (3.0) eV for Fe (Ni) and the same value of the interatomic exchange, \( J = 0.9 \) eV for both Fe and Ni, a result of constrained LDA calculations [42, 4, 5]. These parameters, which are consistent with those of many studies result in a very good description of the physical properties of Fe and Ni.

As was discussed above, the DMFT maps the many-body system onto a multi-orbital quantum impurity, i.e. a set of local degrees of freedom in a bath described by the Weiss field function \( G \). The impurity action (here \( n_{m\sigma} = c_{m\sigma}^+ c_{m\sigma} \) and \( c(\tau) = [c_{m\sigma}(\tau)] \) is a vector of Grassman variables) is given by:

\[
S_{\text{eff}} = -\int_0^\beta d\tau \int_0^\beta d\tau' \text{Tr}[c^+(\tau)G^{-1}(\tau,\tau')c(\tau')] + \frac{1}{2} \sum_{m,m',\sigma} \int_0^\beta d\tau [U_{mm'}n_{\sigma}^m n_{-\sigma}^{m'} + (U_{mm'} - J_{mm'})n_{\sigma}^m n_{\sigma}^{m'}] \quad (35)
\]

It describes the spin, orbital, energy and temperature dependent interactions of a particular magnetic 3d-atom with the rest of the crystal and is used to compute the local Greens function matrix:

\[
G_{\sigma}(\tau - \tau') = -\frac{1}{Z} \int D[c,c^+] e^{-S_{\text{eff}}(c) c^+(\tau')} 
\]

\((Z \) is the partition function) and the impurity self-energy \( G^{-1}_\sigma(\omega_n) - G^{-1}_\sigma(\omega_n) = \Sigma_\sigma(\omega_n) \).

The Weiss field function is required to obey the self-consistency condition (14), which can be specified for a given case as

\[
G_\sigma(\omega_n) = \sum_k [(i\omega_n + \mu)1 - H_{LDA}^{LDA}(k) - \Sigma_{dc}^{LDA}(\omega_n)]^{-1} \quad (37)
\]

The local matrix \( \Sigma_{dc}^{LDA} \) is the sum of two terms, the impurity self energy and a so-called “double counting ” correction, \( E_{dc} \) which is meant to subtract the average electron-electron interactions already included in
the LDA Hamiltonian. For metallic systems we proposed the general form of d-c correction: $\Sigma_{dc}^{\sigma}(\omega) = \Sigma_{\sigma}(\omega) - \frac{1}{2} Tr_{\sigma} \Sigma_{\sigma}(0)$. This is motivated by the fact that the static part of the correlation effects are already well described in the density functional theory. Only the $d$-part of the self-energy is presented in our calculations, therefore $\Sigma_{dc}^{d} = 0$ for $s$- and $p$-states as well as for non-diagonal $d-s$, $p$ contributions. In order to describe the finite temperature ferromagnetism of transition metals we use the non spin-polarized LDA Hamiltonian $H^{LDA}(k)$ and accumulate all temperature-dependent spin-splittings in the self-energy matrix $\Sigma_{dc}^{\sigma}(\omega_n)$.

We used the impurity QMC scheme (see Appendix) for the solution of the multiband DMFT equations [45]. The Hirsch discrete Hubbard-Stratonovich transformations [43, 44] introduces $(2M-1) M$ auxiliary Ising fields $S_{m' m}^{\sigma \sigma'}$ where $M$ is the orbital degeneracy of the $d$-states and calculate $G_{\sigma}(\tau)$ by an exact integration of the fermion degrees of freedom in the functional integral (Eq.(36)) [3]. In order to sample efficiently all the spin configurations in the multi-band QMC scheme, it is important to use “global” spin-flips: $[S_{m' m}^{\sigma \sigma'}] \rightarrow [-S_{m' m}^{\sigma \sigma'}]$ in addition to the local moves of the auxiliary fields. The number of QMC sweeps was of the order of $10^5$. A parallel version of the DMFT program was used to sample the 45 Ising fields for $3d$-orbitals. We used 256 $k$-points in the irreducible part of the BZ for the $k$ integration. 10 to 20 DMFT iterations were sufficient to achieve convergence far from the Curie point. Due to the cubic symmetry of the bcc-Fe and fcc-Ni lattices the local Green function is diagonal in the basis of real spherical harmonics. The spectral functions for real frequencies were obtained from the QMC data by applying the maximum entropy method [46].

Our results for the local spectral function for iron and nickel are shown in Figs.1 and 2, respectively. The SDF approach describes well all the qualitative features of the density of states (DOS), which is especially non-trivial for nickel. Our QMC results reproduce well the three main correlation effects on the one particle spectra below $T_C$ [47, 48, 49]: the presence of a famous 6 eV satellite, the 30% narrowing of the occupied part of $d$-band and the 50% decrease of exchange splittings compared to the LDA results. Note that the satellite in Ni has substantially more spin-up contributions in agreement with photoemission spectra [49]. The exchange splitting of the $d$-band depends very weakly on temperature from $T=0.6 \ T_C$ to $T=0.9 \ T_C$. Correlation effects in Fe are less pronounced than in Ni, due to its large spin-splitting and the characteristic bcc-structural dip in the density of states for spin-down states near Fermi level, which reduces the density of states for particle hole excitations.
Now we discuss the applications of the SDF approach to the description of the finite-temperature magnetic properties of iron and nickel. While density functional theory can in principle provide a rigorous description of the thermodynamic properties, at present there is no accurate practical implementation available. As a result the finite-temperature properties of magnetic materials are estimated following a simple suggestion [30], whereby constrained DFT at $T = 0$ is used to extract exchange constants for a classical Heisenberg model, which in turn is solved using approximation methods (e.g. RPA, mean field) from classical statistical mechanics of spin systems [30, 50, 51, 52]. The most recent implementation of this approach gives good values for the transition temperature of iron but not of nickel [53]. While these localized spin models give, by construction, at high temperatures a Curie-Weiss like magnetic susceptibility, as observed experimentally in Fe and Ni, they encounter difficulties in predicting the correct values of the Curie constants [54].

The uniform spin susceptibility in the paramagnetic state, $\chi_{q=0} = dM/dH$, was extracted from the QMC simulations by measuring the induced magnetic moment in a small external magnetic field. It includes the polarization of the impurity Weiss field by the external field [3]. The dynamical mean field results account for the Curie-Weiss law which is observed experimentally in Fe and Ni. As the temperature increases above $T_C$, the atomic character of the system is partially restored resulting in an atomic like susceptibility with an effective moment:

$$\chi_{q=0} = \frac{\mu_{eff}^2}{3(T - T_C)}$$

The temperature dependence of the ordered magnetic moment below the Curie temperature and the inverse of the uniform susceptibility above the Curie point are plotted in Fig. 3 together with the corresponding experimental data for iron and nickel [55]. The LDA+DMFT calculations describes the magnetization curve and the slope of the high-temperature Curie-Weiss susceptibility remarkably well. The calculated values of high-temperature magnetic moments extracted from the uniform spin susceptibility are $\mu_{eff} = 3.09\ (1.50)\mu_B$ for Fe (Ni), in good agreement with the experimental data $\mu_{eff} = 3.13\ (1.62)\mu_B$ for Fe (Ni) [55].

We have estimated the values of the Curie temperatures of Fe and Ni from the disappearance of spin polarization in the self-consistent solution of DMFT problem and from the Curie-Weiss law in Eq.(38). Our
Figure 1. LDA+DMFT results for ferromagnetic iron ($T = 0.8 \, T_C$). The partial densities of d-states (full lines) is compared with the corresponding LSDA results at zero temperature (dashed lines) for the spin-up (arrow-up) and spin-down (arrow-down) states. The insert shows the spin-spin autocorrelation function for $T=1.2 \, T_C$.

Figure 2. Same quantities as in Fig.1 for ferromagnetic nickel ($T = 0.9 \, T_C$). The insert shows the spin-spin autocorrelation function for $T=1.8 \, T_C$. 
estimates $T_C = 1900 \ (700) K$ are in reasonable agreement with experimental values of 1043 \ (631) K for Fe (Ni) respectively\[55\], considering the single-site nature of the DMFT approach, which is not able to capture the reduction of $T_C$ due to long wavelength spin-waves. These effects are governed by the spin-wave stiffness. Since the ratio of the spin-wave stiffness ($D$) to $T_C$, $T_C/a^2D$ is nearly a factor of 3 larger for Fe than for Ni\[55\] (a is the lattice spacing), we expect the DMFT $T_C$ to be much higher than the observed Curie temperature in Fe than in Ni. Note that this is a consequence of the long-range oscillating character of exchange interactions in iron compared to short-range ferromagnetic exchange interactions in nickel \[53\]. Quantitative calculations demonstrating the sizeable reduction of $T_C$ due to spin waves in Fe in the framework of a Heisenberg model were performed in Ref \[53\].

Within dynamical mean field theory one can also compute the local spin susceptibility defined by

$$\chi_{\text{loc}} = \frac{g^2}{3} \int_0^\beta d\tau \langle S(\tau) S(0) \rangle$$ (39)
where \( g_s = 2 \) is the gyromagnetic ratio and \( S = \frac{1}{2} \sum_{m,\sigma,\sigma'} c_{m\sigma}^\dagger \sigma \sigma' c_{m\sigma'} \) is single-site spin operator and \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) are Pauli matrices. It differs from the \( q = 0 \) susceptibility by the absence of spin polarization in the Weiss field of the impurity model. Eq.(39) cannot be probed directly in experiments but it is easily computed in DMFT-QMC. Its behavior as function of temperature gives a very intuitive picture of the degree of correlations in the system. In a weakly correlated system we expect Eq.(39) to be nearly temperature independent, while in a strongly correlated system we expect a leading Curie-Weiss behavior at high temperatures \( \chi_{\text{local}} = \mu_{\text{loc}}^2/(3T + \text{const}) \) where \( \mu_{\text{loc}} \) is an effective local magnetic moment. In the Heisenberg model with spin \( S \), \( \mu_{\text{loc}}^2 = S(S+1)g_s^2 \) and for well-defined local magnetic moments (e.g., for rare earth magnets) this quantity should be temperature independent. For the itinerant electron magnets, \( \mu_{\text{loc}} \) is temperature-dependent, due to a variety of competing many-body effects such as Kondo screening, the induction of local magnetic moment by temperature [35] and thermal fluctuations which disorders the moments [56]. All these effects are included in the DMFT calculations. The \( \tau \)-dependence of the correlation function \( \langle S(\tau)S(0) \rangle \) results in the temperature dependence of \( \mu_{\text{loc}} \) and is displayed in the inserts on the Figs.1,2. Iron can be considered as a magnet with very well-defined local moments above \( T_C \) (the \( \tau \)-dependence of the correlation function is relatively weak), whereas nickel is more of an itinerant electron magnet (stronger \( \tau \)-dependence of the local spin-spin autocorrelation function).

The comparison of the values of the local and the \( q = 0 \) susceptibility gives a crude measure of the degree of short-range order which is present above \( T_C \). As expected, the moments extracted from the local susceptibility Eq.(39) are a bit smaller (2.8 \( \mu_B \) for iron and 1.3 \( \mu_B \) for nickel) than those extracted from the uniform magnetic susceptibility. This reflects the small degree of the short-range correlations which remain well above \( T_C \) [58]. The high-temperature LDA+DMFT clearly show the presence of a local-moment above \( T_C \). This moment, is correlated with the presence of high energy features (of the order of the Coulomb energies) in the photomission. This is also true below \( T_C \), where the spin dependence of the spectra is more pronounced for the satellite region in nickel than for that of the quasiparticle bands near the Fermi level (Fig. 2). This can explain the apparent discrepancies between different experimental determinations of the high-temperature magnetic splittings [57, 59, 60] as being the results of probing different energy regions. The resonant photoemission experiments [59] reflect the presence of local-moment polarization in the high-energy spectrum above the Curie temperature in nickel, while the low-energy ARPES
investigations [60] results in non-magnetic bands near the Fermi level. This is exactly the DMFT view on the electronic structure of transition metals above \( T_C \). Fluctuating moments and atomic-like configurations are large at short times, which results in correlation effects in the high-energy spectra such as spin-multiplet splittings. The moment is reduced at longer time scales, corresponding to a more band-like, less correlated electronic structure near the Fermi level.

7. Approximate solution for the self-energy: TM-FLEX method

The QMC method described above is probably the most accurate way of solving the effective impurity problem in the DMFT. However, it is rather cumbersome and expensive computationally; besides that, it deals with the “truncated” two-indices interaction matrix (see Eq.(35)) instead of the complete four-indices one. Therefore a scheme has been proposed in Ref. [6] based on a multiband spin-polarized generalization of the “fluctuating exchange” (FLEX) approximation by Bickers and Scalapino [61]. The original formulation of the FLEX approximation treats both particle-hole (PH) and particle-particle (PP) channels on an equal footing. But their roles in magnetism are completely different. The interaction of electrons with spin fluctuations in PH channel leads to the most relevant correlation effects [35] whereas PP processes are important for the renormalizations of the effective interactions in spirit of the \( T \)-matrix approach (“ladder approximation”) by Galitskii [62] and Kanamori [63]. Therefore we used in Ref. [6] a “two-step” procedure when, at first, the bare matrix vertex is replaced by a \( T \)-matrix, and, secondly, PH channel processes with this effective interaction are taken into account explicitly. This approximation has high enough accuracy both for the Hubbard model and for real systems with moderate correlations \( U < W/2 \) where \( U \) is the Hubbard on-site repulsion energy and \( W \) is the bandwidth (see [6] and Refs therein). However, specific form of the approximation used in [6] can be improved further by taking into account the spin-dependence of the \( T \)-matrix. Here we present the formulation of this \( T \)-matrix-FLEX approximation.

Let us start, as in Refs. [5, 6], with the general many-body Hamiltonian for a crystal in the LDA+\( U \) scheme [64]:

\[
H = H_t + H_U \\
H_t = \sum_{\lambda \lambda' \sigma} t_{\lambda \lambda'} c_{\lambda \sigma}^\dagger c_{\lambda' \sigma}
\]
\[ H_U = \frac{1}{2} \sum_{\{\lambda_i\}} \langle \lambda_1 \lambda_2 | v | \lambda_1' \lambda_2' \rangle c_{\lambda_1 \sigma}^+ c_{\lambda_2 \sigma'}^+ c_{\lambda_2' \sigma'} c_{\lambda_1' \sigma} \]  

(40)

where \( \lambda = im \) are the site number \((i)\) and orbital \((m)\) quantum numbers, \(\sigma = \uparrow, \downarrow\) is the spin projection, \(c^+, c\) are the Fermion creation and annihilation operators, \(H_t\) is the effective single-particle Hamiltonian from the LDA, corrected for the double-counting of average interactions among correlated electrons as it was described above, and the Coulomb matrix elements are defined in the standard way

\[
\langle 12 | v | 34 \rangle = \int dr dr' \psi_1^*(r) \psi_2^*(r') v(r - r') \psi_3(r) \psi_4(r'),
\]

(41)

where we define for briefness \(\lambda_1 \equiv 1\) etc. Following Ref. [62] we take into account the ladder \((T\text{-matrix})\) renormalization of the effective approximation:

\[
\langle 13 | T^{\sigma\sigma'} (i\Omega) | 24 \rangle = \langle 13 | v | 24 \rangle - \frac{1}{\beta} \sum_{\omega} \sum_{5678} \langle 13 | v | 57 \rangle \times G_{56}^\sigma (i\omega) G_{78}^{\sigma'} (i\Omega - i\omega) \langle 68 | T^{\sigma\sigma'} (i\Omega) | 24 \rangle
\]

(42)

Further we rewrite the perturbation theory in terms of this effective interaction matrix.

At first, we take into account the “Hartree” and “Fock” diagrams with the replacement of the bare interaction by the \(T\text{-matrix}\)

\[
\Sigma_{12,\sigma}^{(TH)} (i\omega) = \frac{1}{\beta} \sum_{\Omega} \sum_{34\sigma'} \langle 13 | T^{\sigma\sigma'} (i\Omega) | 24 \rangle G_{43}^{\sigma'} (i\Omega - i\omega)
\]

\[
\Sigma_{12,\sigma}^{(TF)} (i\omega) = -\frac{1}{\beta} \sum_{\Omega} \sum_{34} \langle 14 | T^{\sigma\sigma} (i\Omega) | 32 \rangle G_{34}^\sigma (i\Omega - i\omega)
\]

(43)

Note that \(\Sigma^{(TH)} + \Sigma^{(TF)}\) contains exactly all the second-order contributions. Now we have to consider the contribution of particle-hole excitations to sigma. Similar to [6] we will replace in the corresponding diagrams the bare interaction by the static limit of the \(T\text{-matrix}\). However, we improve the approximation [6] by taking into account its spin dependence. When considering the particle-hole channel we replace in the Hamiltonian (40) \(v \rightarrow T^{\sigma\sigma'}\) which is the solution of Eq.(42) at \(\Omega = 0\).

Eq. (43) is exact in the limit of low electron (or hole) density which is important for the criterion of magnetism e.g. in the case of nickel (with almost completely filled \(d\)-band).

Now we rewrite the effective Hamiltonian (40) with the replacement \(\langle 12 | v | 34 \rangle\) by \(\langle 12 | T^{\sigma\sigma'} (\Omega = 0) | 34 \rangle\) in \(H_U\). To consider the correlation
effects due to PH channel we have to separate density ($d$) and magnetic ($m$) channels as in [61] 

$$d_{12} = \frac{1}{\sqrt{2}} \left( c_{1\uparrow}^+ c_{2\uparrow} + c_{1\downarrow}^+ c_{2\downarrow} \right)$$

$$m_{12}^0 = \frac{1}{\sqrt{2}} \left( c_{1\uparrow}^+ c_{2\uparrow} - c_{1\downarrow}^+ c_{2\downarrow} \right)$$

$$m_{12}^+ = c_{1\uparrow}^+ c_{2\uparrow},$$

$$m_{12}^- = c_{1\downarrow}^+ c_{2\downarrow},$$

(44)

Then the interaction Hamiltonian can be rewritten in the following matrix form 

$$H_U = \frac{1}{2} Tr \left( D^+ * V^\parallel * D + m^+ * V^\perp_m * m^--m^- * V^\perp_m * m^+ \right)$$

(45)

where $*$ means the matrix multiplication with respect to the pairs of orbital indices, e.g.

$$(V^\perp_m * m^+)_{11'} = \sum_{34} (V^\perp_m)_{11',22'} m^+_{22'},$$

the supervector $D$ defined as

$$D = (d, m^0), D^+ = \left( d^+, m^+_0 \right),$$

and the effective interactions have the following form:

$$(V^\perp_m)_{11',22'} = -\langle 12 | T^\uparrow \downarrow | 2'1' \rangle,$$

$$V^\parallel = \left( \begin{array}{cc} V^{dd} & V^{dm} \\ V^{md} & V^{dd} \end{array} \right),$$

$$V^{dd}_{11',22'} = \frac{1}{2} \sum_{\sigma\sigma'} \langle 12 | T^{\sigma\sigma'} | 1'2' \rangle - \frac{1}{2} \sum_{\sigma} \langle 12 | T^{\sigma\sigma} | 2'1' \rangle,$$

$$V^{mm}_{11',22'} = \frac{1}{2} \sum_{\sigma\sigma'} \langle 12 | T^{\sigma\sigma'} | 1'2' \rangle - \frac{1}{2} \sum_{\sigma} \langle 12 | T^{\sigma\sigma} | 2'1' \rangle,$$

(46)

$$V^{dm}_{22',11'} = \frac{1}{2} \left[ \langle 12 | T^{\uparrow\downarrow} | 1'2' \rangle - \langle 12 | T^{\downarrow\uparrow} | 1'2' \rangle - \langle 12 | T^{\uparrow\downarrow} | 1'2' \rangle + \langle 12 | T^{\downarrow\uparrow} | 2'1' \rangle + \langle 12 | T^{\downarrow\uparrow} | 2'1' \rangle \right]$$
To calculate the PH contribution to the electron self-energy we first have to write the expressions for the generalized susceptibilities, both transverse $\chi^\perp$ and longitudinal $\chi^\parallel$. The corresponding expressions are the same as in [6] but with another definition of the interaction vertices. One has

$$\chi^\pm(i\omega) = \left[1 + V_m^\pm * \Gamma^\pm(i\omega)\right]^{-1} * \Gamma^\pm(i\omega),$$

(47)

where

$$\Gamma_{12,34}^{\sigma\sigma'}(\tau) = -G_{23}^{\sigma}(\tau) G_{41}^{\sigma'}(-\tau)$$

(48)

is an “empty loop” susceptibility and $\Gamma(i\omega)$ is its Fourier transform. The corresponding longitudinal susceptibility matrix has a more complicated form:

$$\chi^\parallel(i\omega) = \left[1 + V^\parallel * \chi^\parallel_0(i\omega)\right]^{-1} * \chi^\parallel_0(i\omega),$$

(49)

and the matrix of bare longitudinal susceptibility:

$$\chi^\parallel_0 = \frac{1}{2} \begin{pmatrix} \Gamma^{\uparrow\uparrow} + \Gamma^{\downarrow\downarrow} & \Gamma^{\uparrow\downarrow} - \Gamma^{\downarrow\uparrow} \\ \Gamma^{\downarrow\uparrow} - \Gamma^{\uparrow\downarrow} & \Gamma^{\uparrow\uparrow} + \Gamma^{\downarrow\downarrow} \end{pmatrix},$$

(50)

in the $dd$-, $dm^0$, $m^0d^*$, and $m^0m^0$- channels ($d, m^0 = 1, 2$ in the super-matrix indices). An important feature of these equations is the coupling of longitudinal magnetic fluctuations and of density fluctuations. It is absent in one-band Hubbard model due to the absence of the interaction of electrons with parallel spins. For this case Eqs. (47,49) coincide with the well-known result [65].

Now we can write the particle-hole contribution to the self-energy. According to [6] one has

$$\Sigma_{12,\sigma}^{(ph)}(\tau) = \sum_{34,\sigma'} W_{13,42}^{\sigma\sigma'}(\tau) G_{34}^{\sigma'}(\tau),$$

(51)

with P-H fluctuation potential matrix:

$$W^{\sigma\sigma'}(i\omega) = \begin{bmatrix} W^{\uparrow\uparrow}(i\omega) & W^{\uparrow\downarrow}(i\omega) \\ W^{\downarrow\uparrow}(i\omega) & W^{\downarrow\downarrow}(i\omega) \end{bmatrix},$$

(52)

where the spin-dependent effective potentials are defined as

$$W^{\uparrow\uparrow} = \frac{1}{2} V^\parallel * \left[\chi^\parallel - \chi^\parallel_0\right] * V^\parallel$$
The approach to electronic correlations

\[ W^{\downarrow\downarrow} = \frac{1}{2} V^\parallel \ast [\tilde{\chi}^\parallel - \tilde{\chi}_0^\parallel] \ast V^\parallel \]

\[ W^{\uparrow\downarrow} = V_m^\perp \ast [\chi^{+-} - \chi_0^{+-}] \ast V_m^\perp \]

\[ W^{\downarrow\uparrow} = V_m^\perp \ast [\chi^{-+} - \chi_0^{-+}] \ast V_m^\perp \]

where \( \tilde{\chi}^\parallel, \tilde{\chi}_0^\parallel \) differ from \( \chi^\parallel, \chi_0^\parallel \) by the replacement of \( \Gamma^{\uparrow\uparrow} \Leftrightarrow \Gamma^{\downarrow\downarrow} \) in Eq.(50). We have subtracted the second-order contributions since they have already been taken into account in Eq.(43).

Our complete expression for the self energy is

\[ \Sigma = \Sigma^{(TH)} + \Sigma^{(TF)} + \Sigma^{(PH)} \quad (53) \]

This expression takes into account accurately spin-polaron effects because of the interaction with magnetic fluctuations [6], the energy dependence of \( T \)-matrix which is important for describing the satellite effects in Ni [36], contains the exact second-order terms in \( v \) and is rigorous (because of the first term) for almost filled or almost empty bands. In spirit of the DMFT approach we have to use \( G_0 \) instead of \( G \) in all the expressions when calculating the self-energy on a separated central site. It should be noted that this TM-FLEX scheme is not conserved (or “\( \Phi \)-derivable”) therefore one need to enforce the Luttinger theorem by introducing the \( \mu_0 \) for the bath Green function as in iterative perturbation theory[66].

We have started from the spin-polarized LSDA band structure of ferromagnetic nickel within the TB-LMTO method [41] in the minimal \( s,p,d \) basis set and used numerical orthogonalization to find the \( H_t \) part of our starting Hamiltonian. We take into accounts the Coulomb interactions only between \( d \)-states. Semiempirical analysis of the appropriate interaction value gives \( U \approx 2 - 4 \text{ eV} \). The difficulties with choosing the correct value of \( U \) are connected with complicated screening problems, definitions of orthogonal orbitals in the crystal, and contributions of the intersite interactions. In the quasitomic (spherical) approximation the full \( U \)-matrix for the \( d \)-shell is determined by the three parameters \( U, J \) and \( \delta J \) or equivalently by effective Slater integrals \( F^0, F^2 \) and \( F^4 \) [64]. For example, \( U = F^0, J = (F^2 + F^4)/14 \) and we use the simplest way of estimating \( \delta J \) or \( F^4 \) keeping the ratio \( F^2/F^4 \) equal to its atomic value 0.625 [64]. Note that the value of intra-atomic (Hund) exchange interaction \( J \) is not sensitive to the screening and approximately equals 0.9 eV in different estimations [64].
Figure 4. Spin-up (full lines) and spin-down (dashed lines) density of d-states for ferromagnetic nickel in the LSDA and the LDA+SPTF (LSDA+SPTF) calculations for different average Coulomb interaction $U$ with $J = 1$ eV and temperature $T=200$ K.
The resulting DOS for Ni (Fig. 4) shows that spin-polarized TM-FLEX calculations approximately reproduce the satellite structure and reduction of the band width in satisfactory agreement with exact QMC-result (Fig. 2).

8. Cluster DMFT approach: antiferromagnetism and superconductivity

As it was stressed above the SDF approach does not necessarily connect with the standard DMFT scheme; one can use more general choice of fermion coarse-grained variables, e.g., considering the case of an effective cluster instead of an effective impurity. It is especially important for the problems where intersite correlations are involved from the beginning such as $d$-wave superconducting pairing [22, 23] or charge ordering [67]. Here we consider, following Ref. [23], one of the cluster generalizations of the DMFT, and its application to the problem of magnetism and high-temperature superconductivity (HTSC) of copper-oxide compounds.

The microscopic theory of high-temperature superconducting cuprates is still far from a final understanding [68, 69, 70]. One of the most important recent experimental achievements was the discovery of the pseudogap (PG) phenomenon above the superconducting transition temperature [71] and existence of a sharp 41-meV resonance below $T_c$ related to some collective antiferromagnetic excitations [72]. Thus, an interplay of an antiferromagnetism (AFM) and $d$-wave superconductivity ($d$-SC) in cuprates could be a natural way of discussing different HTSC phenomena. This requires a quantitative electronic structure theory including two different types of the order parameters: AFM and $d$-SC. Within such approach one can in principle analyze the phase diagram of HTSC compounds and resolve the long-standing problem of competition between antiferromagnetism and $d$-wave superconductivity in cuprates [73, 74].

A standard theoretical tool for cuprates electronic structure consists of the two-dimensional Hubbard model [68]. We start with the extended-hopping Hubbard model on the square lattice:

$$H = \sum_{ij} t_{ij} c_{i\sigma}^+ c_{j\sigma} + \sum_i U_i n_{i\uparrow} n_{i\downarrow}$$

where $t_{ij}$ is an effective hopping and $U_i$ local Coulomb interactions. We have chosen the nearest-neighbor hopping $t = 0.25$ eV and the next nearest hopping $t'/t = -0.15$ for the model of La$_{2-x}$Sr$_x$CuO$_4$ [75]. The total band width is $W=2$ eV and all Coulomb parameters set to be $U=1.2$ eV ($U/W = 0.6$). Let us introduce the “super-site” as an $2\times2$ square plaquet. The numeration of the atoms in the super-site is shown...
in the Fig.5. It is useful to introduce the superspinor $C^\dagger_i = \{c^\dagger_{i\alpha}\}$ where $\alpha = 0, 1, 2, 3$ (the spin indices are not shown). Taking into account the spin degrees of freedom, this is the 8-component superspinor creation operator. Then the crystal Green function for the Hubbard model can be rewritten as

$$G(k,i\omega) = [i\omega + \mu - h(k) - \Sigma(i\omega)]^{-1}$$  \hfill (54)

where $h(k,i\omega)$ is the effective hopping supermatrix with self-energy corrections. For simplicity we will write all the formulas in the nearest-neighbor approximations:

$$h(k) = \begin{pmatrix}
0 & tK_x^+ & 0 & tK_y^+
\hline
kK_x^- & 0 & tK_y^+ & 0
0 & tK_x^- & 0 & tK_x^-
tK_y^- & 0 & tK_x^+ & 0
\end{pmatrix}$$ \hfill (55)

where $K_{x(y)} = 1 + \exp(\pm i k_{x(y)} a)$, $a$ is the lattice constant, and each element is a $2 \times 2$ matrix in spin space. Within the cluster-DMFT approach we introduce the intra-atomic self-energy $\Sigma_0$ and the inter-atomic self-energies $\Sigma_x, \Sigma_y$, and both functions are of intra-site nature in the sense of our super-site:

$$\Sigma(i\omega) = \begin{pmatrix}
\Sigma_0 & \Sigma_x & 0 & \Sigma_y
\hline
\Sigma_x^* & \Sigma_0 & \Sigma_y & 0
0 & \Sigma_y^* & \Sigma_0 & \Sigma_x^*
\Sigma_y^* & 0 & \Sigma_x & \Sigma_0
\end{pmatrix}$$ \hfill (56)

For the small $2 \times 2$ cluster it is useful to introduce the translationally invariant ($k$-dependent) self-energy and rewrite $h(k) + \Sigma(i\omega) \rightarrow h(k,i\omega)$ where

$$h(k,i\omega) = \begin{pmatrix}
\Sigma_0 & t_xK_x^+ & 0 & t_yK_y^+
t_x^*K_x^- & \Sigma_0 & t_yK_y^+ & 0
0 & t_y^*K_y^- & \Sigma_0 & t_x^*K_x^-
t_y^*K_y^- & 0 & t_xK_x^+ & \Sigma_0
\end{pmatrix}.$$  \hfill (57)

The effective Hamiltonian is defined through the renormalized energy dependent hoppings: $t_x = t + \Sigma_x$, $t_y = t + \Sigma_y$. The functions $\Sigma_0(i\omega)$, $\Sigma_x(i\omega)$, $\Sigma_y(i\omega)$ are found self-consistently within the cluster DMFT scheme and for the $d$-wave superconducting state $\Sigma_x \neq \Sigma_y$. It is straightforward to generalize this scheme for a next-nearest neighbor hopping as well as the long-range Green function and the self-energy. In this case
we can renormalized also the second-nearest hopping: $t_{xy} = t' + \Sigma_{xy}$ for the $2 \times 2$ cluster, where $\Sigma_{xy}$ (or $\Sigma^{02}$) is the non-local self-energy in the $xy$ direction.

In the cluster version of the DMFT scheme one can write the matrix equation for the bath Green function matrix $\mathcal{G}$ which describe an effective interaction with the rest of crystal:

$$\mathcal{G}^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega),$$

where the local cluster Green function matrix is equal to $G_{\alpha\beta}(i\omega) = \sum_{\mathbf{k}} G_{\alpha\beta}(\mathbf{k}, i\omega)$, and the summation is run over the Brillouin zone of the square lattice. If instead of Eq.(57) we use Eq.(55), it would corresponds to a free cluster DMFT scheme or the simplest case of the so-called cellular-DMFT. Note that we use a translationally invariant self-energy obtained from the cluster DMFT scheme (Eq.(57)) or the simplest version of so-called cellular DMFT approach [24]. We believe that for a given choise of the small $(2 \times 2)$ cluster, the renormalization of the hopping parameter by $\Sigma_x$, $\Sigma_y$ terms is physically essential. The
present “matrix” form of a cluster DMFT with the self-energy which is not periodic inside the cluster allow us to study a multicomponent ordered state. Unfortunately, in contrast with the so-called Dynamical Cluster Approximation (DCA) [22] or cellular-DMFT approach [24] (see above, Section 1), we were unable to prove the causality of this approach for arbitrary band structure and interaction parameters. However, the causality of the Green function for the realistic choice of the parameters has been checked numerically.

In this case we have the standard DMFT problem with four “orbital” states per super-site. It has been solved by the multi-orbital QMC technique described above (Section 3). We used the generalized Nambu technique [76] to analyze the coexistence of the magnetic ordering and superconductivity. Let us introduce the superspinor

$$\Psi_i^+ (\tau) \equiv (\psi_{1i}^+, \psi_{2i}^+, \psi_{3i}^+, \psi_{4i}^+) = (c_{i\uparrow}^+, c_{i\downarrow}^+, c_{i\uparrow}, c_{i\downarrow})$$

and the anomalous averages describing the (collinear) antiferromagnetism $$\langle c_{i\uparrow} c_{j\downarrow} \rangle$$ and the superconductivity $$\Delta_{ij} = \langle c_{i\downarrow} c_{j\uparrow} \rangle$$.

The generalization of the Hirsch-Fye QMC-algorithm [44] for the superconducting problem [77] have been used. In the 4-spinor case a discrete Hubbard-Stratonovich transformation has the following form:

$$\exp[-\Delta \tau U_i n_{i\uparrow} n_{i\downarrow} + \frac{\Delta \tau U_i}{2} (n_{i\uparrow} + n_{i\downarrow})] = \frac{1}{2} \sum_{\sigma = \pm 1} \exp[\lambda_i \sigma (\psi_{1i}^+ \psi_{1i} - \psi_{2i}^+ \psi_{2i} - \psi_{3i}^+ \psi_{3i} + \psi_{4i}^+ \psi_{4i})]$$ (58)

where $$\lambda_i = \frac{1}{2} \arccosh[\exp(\frac{1}{2} \Delta \tau U_i)]$$.

Since we take into account only the singlet pairing, we obtain the following nonzero elements of the d-SC energy gap parameters: $$\Delta = \Delta_{01} = -\Delta_{12} = \Delta_{23} = -\Delta_{30}$$. One can chose $$\Delta_{ij}$$ to be real and therefore symmetric: $$\Delta_{ij} = \Delta_{ji}$$. Separating normal and anomalous parts of the Green function we have

$$G (\mathbf{k}, \tau, \tau') = \begin{pmatrix} G (\mathbf{k}, \tau, \tau') & F (\mathbf{k}, \tau, \tau') \\ F^+ (\mathbf{k}, \tau, \tau') & -G (-\mathbf{k}, \tau', \tau) \end{pmatrix}$$ (59)

where $$G (\mathbf{k}, \tau, \tau') = -\langle T_\tau C_{\mathbf{k}} (\tau) C_{\mathbf{k}}^+ (\tau') \rangle$$ and $$F (\mathbf{k}, \tau, \tau') = -\langle T_\tau C_{\mathbf{k}} (\tau) C_{-\mathbf{k}} (\tau') \rangle$$ are the matrices in spin and “orbital” space. It is convenient to expand the anomalous Green function in Pauli matrices $$F = (F^0 + F \sigma) i\sigma^y$$ and use the symmetry properties [78]:

$$F^0 (\mathbf{k}, \tau, \tau') = F^0 (-\mathbf{k}, \tau', \tau)$$ (60)

$$F (\mathbf{k}, \tau, \tau') = -F (-\mathbf{k}, \tau', \tau)$$.
then a $4 \times 4$ spinor formalism is reduced to $2 \times 2$ one in the collinear antiferromagnetic case with the d-wave superconductivity with the following spin-matrix form of the local Green function for the super-site:

$$G(\tau, \tau') = \begin{pmatrix} G_{\uparrow}(\tau, \tau') & F(\tau, \tau') \\ F(\tau, \tau') & -G_{\downarrow}(\tau', \tau) \end{pmatrix}$$

(61)

and the QMC formalism for the antiferromagnetic superconducting state is equivalent to the previous non-magnetic one [77]. Using the discretization of $[0, \beta]$ interval with $L$-time slices: $\Delta \tau = \beta/L$ ($\beta = 1/T$ is an inverse temperature) the $G_{\sigma}$- and $F$- Green functions become the matrices of $2NL$ dimension, where $N$ is the number of atoms in the cluster. After Fourier transform to the Matsubara frequencies the Green function matrix has the following form:

$$G(i\omega) = \begin{pmatrix} G_{\uparrow}(i\omega) & F(i\omega) \\ F(i\omega) & -G_{\downarrow}^*(i\omega) \end{pmatrix}$$

(62)

In superconducting states the self-energy defined as [3]:

$$G^{-1}(i\omega) - G^{-1}(i\omega) = \begin{pmatrix} \Sigma_{\uparrow}(i\omega) & S(i\omega) \\ S(i\omega) & -\Sigma_{\downarrow}^*(i\omega) \end{pmatrix},$$

(63)

and the inverse crystal Green function matrix is equal to:

$$G^{-1}(k,i\omega) = \begin{pmatrix} i\omega + \mu - h(k,i\omega) & s(k,i\omega) \\ s(k,i\omega) & i\omega - \mu + h^*(k,i\omega) \end{pmatrix}$$

(64)

where $s(k,i\omega)$ is the translationally invariant anomalous part of the self-energy $S(i\omega)$ similar to Eq.(55).

The two-component order parameters state which includes Neel antiferromagnetism and d-wave superconductivity (Fig.5a) lowered the symmetry of the effective cluster-DMFT problem. A self-consistent DMFT cluster problem with AFM and d-SC general order parameters have been solved within the QMC scheme for 8x8 matrix Green function with $L=64$ time slices. The resulting two order parameters for $\beta = 60$ eV$^{-1}$ ($T=190$ K) and $t' = 0$ presented in Fig.5c together with the generic HTSC phase diagram (Fig.5b) as function of the hole doping. In this case the ordered magnetic moment is directly related with imaginary-time Green function $G_{\sigma}(\tau)$: $M = G_{\uparrow}^{00}(0) - G_{\downarrow}^{00}(0)$ and for the d-SC order parameter we chose a positive value of superconducting imaginary time Green function $F_{01}^{01}(0)$. It is important, that we find no serious sign-problem for all QMC calculations with various doping level, probably due to “stabilized” antiferromagnetic dynamical mean fields acting on the atoms in...
Figure 6. Imaginary time normal ($G_{\sigma}$) and superconducting ($F$) Green functions for the $2\times2$ cluster DMFT solution with second-nearest neighbor hopping and inverse temperature $\beta = 50 \text{ eV}^{-1}$ ($T=230 \text{ K}$).

our $2\times2$ cluster. Note that the AFM cluster-DMFT solution exists for a much higher doping concentration than experimental AFM ordered state and describes a dynamical mean-field version of AFM-spin fluctuations related to pseudogap phenomena (the PG-region on Fig.5b). The maximum of d-SC order parameter corresponds to a doping level of about 15% in agreement with the generic HTSC phase diagram. The d-SC order parameter is zero close to the undoped region ($x=0$), due to presence of a large AFM-gap. When the AFM-gap is closed ($x \sim 5\%$) the d-SC states develop but for $x > 20\%$ the d-gap decrease again since AFM spin-fluctuations around $(\pi, \pi)$ point disappear [69]. The precise characteristic of the phase diagram including the interactions between the AFM and d-SC order parameters demands an extensive cluster-DMFT calculations for different temperatures and doping.

We would like to note that the existing of d-SC cluster-DMFT solution for such high temperatures does not necessary means that the superconducting transition temperature is larger then 190K in our model.
crude estimation shows that the d-SC solution disappears at $T=300$ K for $x=0.15$ and the AFM solutions for $x=0$ become unstable at the temperature just above $1000$ K. This could be the sign of a “local” AFM solution and a local d-wave solution, like local moments in magnetic systems [6]. Due to the multiscale nature of the problem under considerations, essentially different energies connected with local moment formation, long-range magnetic order, local d-wave pairs within the $2\times2$ plaquet, and finally coherent superconductivity, it is difficult to distinguish a real long-range ordering from slow dynamical fluctuations in our QMC simulations. We plan to separate these energy scales analytically and estimate superconducting transition temperature in a future publication.

The role of next-nearest hopping is to lower of the van-Hove singularity [75] which increases the density of state at the Fermi level for the hole-doped case and favors the d-SC solution for a moderate correlation strength. There is also a change in the spin-fluctuation spectrum related with the broadening of AFM-peak near the $(\pi, \pi)$ point due to formation of so-called extended van-Hove singularities with increasing of the $t'$. We show one of the AFM-dSC solution on the Fig.6 with the next nearest-neighbor hopping for the 10% doping level and $\beta = 50$ eV$^{-1}$. The resulting local magnetic moment is $M=0.28 \mu_B$ and the d-SC order parameter $F(0)=0.036$. One can see that the superconducting order parameter is really of the $d_{x^2-y^2}$ symmetry since diagonal elements ($F^{00}$) as well as the next nearest-neighbors elements ($F^{02}$) are all equal to zero and only the nearest neighbor superconducting Green functions ($F^{01}$) are non-zero and change the sign for $F_x$ and $F_y$ components. The normal local Green function ($G^{00}$) (plotted for the spin-up atom in the Fig.6) as well as ($G^{02}$) are spin-split, while the nearest-neighbor Green function ($G^{01}$) has no spin-splitting due to AFM spin symmetry (see Fig.5). The absence of magnetic polarization in the non-diagonal G-function along the x(y)-directions suppress the magnetic pair-breaking and makes the AFM-dSC coexistence possible.

9. Summary

The spectral density functional (SDF) approach allows us to study correlation effects in solids based on realistic electronic structure calculations. Among all possible applications we have chosen the magnetism of transition metals and high-temperature superconductivity. From these two examples one can see already all the main advantages of the new approach in comparison with standard density functional theory. First, we can describe now the spectral density transfer phenomena (e.g., the
formation of 6 eV satellite in Ni), the quasiparticle damping and other effects connected with the frequency dependence of the self-energy; they are absent completely not only in the DF approach but also in the Hartree-Fock, LDA+U, or self-interaction corrections approximation (see, e.g., [64]). Second, we can describe adequately the contribution of the Bose degrees of freedom (e.g., spin fluctuations) to the electronic structure and thermodynamic properties. In the DF-based calculations the temperature is really taken into account only via the thermal expansion and the Fermi distribution function [79]. It was the main reason for the failure of the standard band theory for the description of finite-temperature effects in magnetic metals. We show that the SDF gives a satisfactory solution of this problem.

Most of real applications of the SDF approach are connected with the single-site dynamical mean field theory. At the same time, for a number of problems this can be insufficient and some generalizations to take into account the non-local effects are necessary, for example, cluster ones. The first attempts of such generalizations are already leading to some interesting results in the theory of high-$T_c$ superconductivity [22, 23] but a lot of additional work is required in the area of cluster dynamical mean field theories to reach the level of understanding that was reached in single site DMFT.

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Appendix: Multi-orbital QMC scheme

The multi-orbital DMFT problem and general cluster DMFT scheme can be reduced to the general impurity action:

\[ S = - \int_0^\beta d\tau \int_0^\beta d\tau' \sum_{i,j} c_i^\dagger(\tau) G_{ij}(\tau - \tau') c_j(\tau') + \frac{1}{2} \int_0^\beta d\tau \sum_{i,j} n_i(\tau) U_{ij} n_j(\tau) \]

where \( i = \{ m, \sigma \} \) - orbital (site) and spin. Without spin-orbital coupling we have:

\[ G_{ij} = G_{m,m'}^{\sigma \sigma'} \delta_{\sigma \sigma'} \]

The auxiliary fields Green-function QMC use the discrete Hubbard-Stratanovich transformation introduced by Hirsch [43]

\[
\exp\{\Delta \tau U_{ij}[n_i n_j - \frac{1}{2}(n_i + n_j)]\} = \frac{1}{2} \sum_{S_{ij} = \pm 1} \exp\{\lambda_{ij} S_{ij}(n_i - n_j)\}
\]

where \( S_{ij}(\tau) \) are the auxiliary Ising fields for each pair of orbitals and time slice with the strength:

\[ \lambda_{ij} = \text{arccosh}\exp\{\frac{\Delta \tau}{2} U_{ij}\} \]

Using Hirsch transformation one can integrated out fermionic fields in the path integral [3] and resulting partition function and Green function matrix have the following form:

\[
Z = \frac{1}{2^{N_f L}} \sum_{S_{ij}(\tau)} \det[\hat{G}^{-1}(S_{ij})] \\
\hat{G} = \frac{1}{2^{N_f L}} \sum_{S_{ij}(\tau)} \hat{G}(S_{ij}) \det[\hat{G}^{-1}(S_{ij})]
\]

where \( N_f \) is the number of Ising fields, \( L \) is the number of time slices, and \( \hat{G}(S_{ij}) \) is the Green function in the auxiliary Ising fields:

\[ G_{ij}^{-1}(S) = G_{ij}^{-1} + \Delta_i \delta_{ij} \delta_{\tau \tau'} \]

\[ \Delta_i = (e^{V_i} - 1) \]

\[ V_i(\tau) = \sum_{j(\neq i)} \lambda_{ij} S_{ij}(\tau) \sigma_{ij} \]

Here we introduce the generalized Pauli matrix:

\[ \sigma_{ij} = \begin{cases} +1, & i < j \\ -1, & i > j \end{cases} \]

For efficient calculation of the Green function in arbitrary configuration of Ising fields \( G_{ij}(S) \) we use the following Dyson equation [44]:

\[ G' = [1 + (1 - G)(e^{V'} - 1)]^{-1} G \]
The QMC important sampling scheme allowed us to integrate over the Ising fields with the \( \text{abs}(\det[\hat{G}^{-1}(S_{ij})]) \) as a stochastic weight\cite{44,3}. For a single spin-flip \( S_{ij} \), the determinant ratio is calculated as following:

\[
\frac{\det[\hat{G}]}{\det[\hat{G}']} = \frac{R_i R_j - R_{ij}}{1 + [1 - G_{ii}(\tau,\tau)]\Delta_i(\tau)}
\]

\[
R_i = 1 + [1 - G_{jj}(\tau,\tau)]\Delta_j(\tau)
\]

\[
R_{ij} = G_{ij}(\tau,\tau)\Delta_j(\tau)
\]

and the Green function matrix updated in the standard manner\cite{44,3}:

\[
G'_{ij,2}(\tau_1,\tau_2) = G_{ij,2}(\tau_1,\tau_2) + [G_{ii,1}(\tau_1,\tau) - \delta_{i1}\delta_{\tau_1,\tau}]\Delta_i(\tau)/R_i(\tau)G_{ij,2}(\tau,\tau_2)
\]

\[
G'_{ij,2}(\tau_1,\tau_2) = G'_{ii,2}(\tau_1,\tau_2) + [G'_{i1,1}(\tau_1,\tau) - \delta_{i1}\delta_{\tau_1,\tau}]\Delta_j(\tau)/R_j(\tau)G'_{jj,2}(\tau,\tau_2)
\]

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SDF approach to electronic correlations

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