Downfolded Self-Energy of Many-Electron Systems

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Starting from the full many-body Hamiltonian of interacting electrons the effective self-energy acting on electrons residing in a subspace of the full Hilbert space is derived. This subspace may correspond to, for example, partially filled narrow bands, which often characterize strongly correlated materials. The formalism delivers naturally the frequency-dependent effective interaction (the Hubbard U) and provides a general framework for constructing theoretical models based on the Green's function language. It also furnishes a general scheme for first-principles calculations of complex systems in which the main correlation effects are concentrated on a small subspace of the full Hilbert space.

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One of the most important quantities in studying the electronic structure of materials is the spectral function, a quantity that contains information about the electronic excitations of the system obtained from photoemission and inverse photoemission experiment corresponding to the addition and removal of an electron. A suitable tool to describe the spectral function is the one-electron Green's function. To calculate the Green function of the full many-body Hamiltonian is a tremendous task and such calculations, within the *GW* approximation (GWA) [1,2], have become feasible thanks to the rapid progress in computer performance. However, two serious difficulties are hampering further progress: first, for many cases, the system size is too large for realistic *GW* calculations and second, from the theoretical point of view, the GWA may not be sufficient to treat correlation problems, especially those in materials with strong correlations containing partially filled narrow bands. These materials with many intriguing properties have been discovered in recent years [3].

The traditional approach for treating such systems is to introduce a model Hamiltonian, focusing on a small subspace of the full Hilbert space that is considered to be most relevant for the correlation problem at hand. Notable examples are the Hubbard model and the Anderson impurity model with the assumption of a local statically screened Coulomb interaction known as the Hubbard U. These models have given us a lot of physical insight into correlation problems ranging from the Kondo effect to high temperature superconductivity. However, these models are not strictly derived from the many-body Hamiltonian and they contain an adjustable parameter U.

The purpose of the present work is to derive from the full many-body Hamiltonian an effective self-energy corresponding to a subspace of the full Hilbert space, expressed in terms of the one-particle Green's function of the subspace. This subspace could be identified as, for example, partially filled narrow bands crossing the Fermi level, where most of the correlations take place, a typical situation in materials with strong correlations. The task is then to derive an effective self-energy acting on electrons residing in this subspace. As a result of the derivation, the effective screened interaction among electrons belonging to the subspace, i.e., the Hubbard U, emerges naturally.

There have been several works related to the present one. One of the early works is due to Bassani *et al.* [4], who divided the full Hilbert space onto the core and valence electrons subspaces and derived an effective Hamiltonian only for the valence electrons. However, they neglected the coupling between the two subspaces, which in our context is indispensable. Another related work on downfolded Hamiltonian may be found in [5,6].

Let us start by defining some basic variables and quantities. The many-electron Hamiltonian is given by

$$H = \int d^3r \hat{\psi}^+(\mathbf{r}) h^0(\mathbf{r}) \hat{\psi}(\mathbf{r}) + \frac{1}{2} \times \int d^3r d^3r' \hat{\psi}^+(\mathbf{r}) \hat{\psi}^+(\mathbf{r}') v(\mathbf{r}-\mathbf{r}') \hat{\psi}(\mathbf{r}') \hat{\psi}(\mathbf{r}), \quad (1)$$

where h^0 is the one-particle part of the Hamiltonian and $\hat{\psi}(\mathbf{r})$ is the field operator. We aim at downfolding the many-body problem onto a subspace of the full Hilbert space, which can consist of, for example, $3d$ or $4f$ orbitals. From now on we refer to the subspace as the d subspace. We first divide the complete field operator into the d field and the rest, denoted by d and r, respectively:

$$\hat{\psi}(\mathbf{r}) = \hat{\psi}_d(\mathbf{r}) + \hat{\psi}_r(\mathbf{r}) = \sum_d \chi_d(\mathbf{r}) \hat{c}_d + \sum_r \chi_r(\mathbf{r}) \hat{c}_r. \quad (2)$$

χ_d and χ_r are the one-particle orbitals and $\hat{c}_d$ and $\hat{c}_r$ are the associated annihilation operators. We use a convention that $(\mathbf{r}t)$ is represented by a number. Since we will utilize the Schwinger functional derivative technique [7] to develop a closed set of equations for G^d, where a probing field $\varphi(1)$ is applied to probe the linear response of the Green function, we define the following Green's functions in the Dirac or interaction representation.

$$iG^d(1,2) = \frac{\langle T[\hat{S}\hat{\psi}_d(1)\hat{\psi}_d^+(2)]\rangle}{\langle\hat{S}\rangle}, \tag{3}$$

$$iG^{rd}(1,2) = \frac{\langle T[\hat{S}\hat{\psi}_r(1)\hat{\psi}_d^+(2)]\rangle}{\langle\hat{S}\rangle}, \tag{4}$$

$$\hat{S} = T\exp\left[-i\int d3\varphi(3)\hat{\psi}^+(3)\hat{\psi}(3)\right]. \tag{5}$$

Before proceeding further with a detailed derivation, we first summarize schematically our final result of a closed set of equations for the effective self-energy and the Green function of the d subspace.

$$\Sigma^{\text{eff}} = \Sigma^d + \Sigma^{rd} + \Sigma^{drd}, \tag{6}$$

$$P^d = -iG^d\Gamma G^d, \tag{7}$$

$$\Gamma^d = 1 + \frac{\delta\Sigma^{\text{eff}}}{\delta G^d}G^d\Gamma^d G^d, \tag{8}$$

$$W = W^r + W^r P^d W, \tag{9}$$

$$G^d = g^d + g^d\Sigma^{\text{eff}}G^d. \tag{10}$$

As a convention, we denote a basis in the d subspace by Greek letters (α) and in the rest of the Hilbert space by small roman letters (n) whereas capital letters (N) denote the full Hilbert space. A small letter g is used to denote a noninteracting Green's function throughout this Letter. From $(\omega - h - \Sigma)G = 1$, where h is the Hartree Hamiltonian and Σ is the self-energy of the *full* Hilbert space, the effective self-energy Σ^{eff} is given by

$$\Sigma^{\text{eff}}_{\alpha\beta} = \Sigma_{\alpha\beta} + (h_{\alpha n} + \Sigma_{\alpha n})G^{rd}_{n\gamma}(G^d)^{-1}_{\gamma\beta}. \tag{11}$$

The first term on the right-hand side is confined to the d subspace and the second represents a coupling between the d and r subspaces. Thus, the effective self-energy $\Sigma^{\text{eff}}_{\alpha\beta}$ is not a simple projection of the full self-energy onto the d subspace $\Sigma_{\alpha\beta}$, but it also contains the effects of the off-diagonal matrix elements $\Sigma_{\alpha n}$ between the d and r subspaces as well as one-particle hoppings $h_{\alpha n}$ arising from the Hartree Hamiltonian. Through G^{rd} and $(G^d)^{-1}$ the effective self-energy depends implicitly on all components of the self-energy.

In contrast to the original Hedin's equations, instead of the bare Coulomb interaction v we have in (9) W^r, a frequency-dependent effective interaction among electrons residing in the chosen d subspace. This effective interaction is screened by P^d, the polarization propagator of the d electrons, yielding the fully screened interaction W of the full system. Γ^d is an *effective* vertex acting on the d subspace only and consequently different from the full vertex. If the subspace is isolated and identified as the Hilbert space of a Hubbard model and W^r is approximated by a local and static interaction (the Hubbard U), Σ^d is then the exact self-energy of the Hubbard model. The explicit expressions for the self-energies will be shown later. In addition to Σ^d the true effective self-energy Σ^{eff} contains a term, Σ^{rd}, which is the self-energy arising from the r subspace acting on the d subspace and a hopping term Σ^{drd} representing hybridization between the d and r subspaces. For a *given* W^r the effective self-energy is a functional of G^d only.

We now proceed with the derivation of the above set of equations and use the convention that repeated indices are summed and repeated variables are integrated, unless they appear on both sides of the equation. We define projection operators for the d and r subspaces as follows:

$$\Delta_i(1,2) = \chi_i(\mathbf{r}_1)\chi_i^*(\mathbf{r}_2)\delta(t_1 - t_2), \qquad (i = d, r). \tag{12}$$

Projecting the equation of motion of the full Green's function on to the d subspace on the right we obtain

$$\left[i\frac{\partial}{\partial t_1} - h(1)\right]\tilde{G}(1,2) - \tilde{\Sigma}(1,3)G^d(3,2) = \Delta_d(1,2), \tag{13}$$

where h is the Hartree Hamiltonian and

$$\tilde{G}(1,2) = G(1,3)\Delta_d(3,2) = G^{rd}(1,2) + G^d(1,2). \tag{14}$$

The self-energy $\tilde{\Sigma}$ is related to the full self-energy Σ according to

$$\tilde{\Sigma}(1,3)G^d(3,2) = \Sigma(1,4)\tilde{G}(4,2). \tag{15}$$

Projecting (13) on the left on the d and r subspaces, respectively, we obtain

$$i\frac{\partial}{\partial t_1}G^d(1,2) - \Delta_d(1,3)h(3)[G^{rd}(3,2) + G^d(3,2)] - \Delta_d(1,3)\tilde{\Sigma}(3,4)G^d(4,2) = \Delta_d(1,2), \tag{16}$$

$$i\frac{\partial}{\partial t_1}G^{rd}(1,2) - \Delta_r(1,3)h(3)[G^{rd}(3,2) + G^d(3,2)] - \Delta_r(1,3)\tilde{\Sigma}(3,4)G^d(4,2) = 0. \tag{17}$$

These two equations of motion can also be obtained by evaluating the equations of motion of the field operators $\hat{\psi}_d$ and $\hat{\psi}_r$.

Rewriting (17) in frequency space and working in the basis representation we solve for G^{rd}:

$$G^{rd}_{n\beta}(\omega) = g^r_{nm}(\omega)[h_{m\gamma} + \tilde{\Sigma}_{m\gamma}(\omega)]G^d_{\gamma\beta}(\omega), \tag{18}$$

where g^r is the noninteracting Green's function of the r subspace: $g^r = [\omega - h^r]^{-1}$. Substituting the above expression for G^{rd} in (16) yields the effective self-energy for G^d.

$$[\omega\delta_{\alpha\gamma} - h_{\alpha\gamma} - \Sigma^{\text{eff}}_{\alpha\gamma}(\omega)]G^d_{\gamma\beta}(\omega) = \delta_{\alpha\beta}, \tag{19}$$

$$\Sigma^{\rm eff}_{\alpha\gamma}(\omega) = \tilde{\Sigma}_{\alpha\gamma}(\omega) + \Sigma^{drd}_{\alpha\gamma}(\omega), \tag{20}$$

$$\Sigma^{drd}_{\alpha\gamma}(\omega) = h_{\alpha n} g^{r}_{nm}(\omega)[h_{m\gamma} + \tilde{\Sigma}_{m\gamma}(\omega)]. \tag{21}$$

Σ^{drd} is zero if we choose the orbitals to be eigenstates of the Hartree Hamiltonian since $h_{\alpha n} = 0$ and the effective self-energy is then simply given by $\tilde{\Sigma}$.

We have established the general form of the downfolded self-energy of the chosen d subspace, which we are now in the position to calculate. The crucial quantity is $\tilde{\Sigma}$, from which the effective self-energy can be readily obtained. From the Schwinger functional derivative technique [7] we have the following well-known relation that relates the self-energy to the functional derivative of the Green function with respect to the applied external field [1,2].

$$\Sigma(1,4)G(4,2) = iW(3,1)\frac{\delta G(1,2)}{\delta V(3)}. \tag{22}$$

Projecting the above relation and the Dyson equation on the right on to the d subspace, and using the relation in (15) yields, respectively,

$$\tilde{\Sigma}(1,4)G^{d}(4,2) = iW(3,1)\frac{\delta \tilde{G}(1,2)}{\delta V(3)}, \tag{23}$$

$$\tilde{G}(1,2) = \tilde{g}(1,2) + g(1,3)\tilde{\Sigma}(3,4)G^{d}(4,2), \tag{24}$$

where $\tilde{g} = g\Delta_{d}$ and g is the Hartree Green's function. We have defined V as the sum of the Hartree and external fields: $V = V_{H} + \varphi$, and the screened Coulomb interaction $W = (\delta V/\delta\varphi)v = \varepsilon^{-1}v$. The quantity $\tilde{\Sigma}$ is in fact the sum of Σ^{d} and Σ^{rd} appearing in (6), which are defined as follows:

$$\Sigma^{d} = iW\frac{\delta G^{d}}{\delta V}(G^{d})^{-1}, \qquad \Sigma^{rd} = iW\frac{\delta G^{rd}}{\delta V}(G^{d})^{-1}, \tag{25}$$

$$\tilde{\Sigma} = \Sigma^{d} + \Sigma^{dr}. \tag{26}$$

We note that $\Sigma^{d} \neq \langle d|\hat{\Sigma}|d\rangle$ and $\Sigma^{rd} \neq \langle r|\hat{\Sigma}|d\rangle$. The splitting of $\tilde{\Sigma}$ into Σ^{d} and Σ^{rd} is useful when making comparison with the conventional Hubbard model.

To calculate $\tilde{\Sigma}$ we need to take the functional derivative of $\tilde{G}$ with respect to the probing field. However, $\tilde{G}$ cannot be inverted and consequently the usual procedure of using the identity $\delta(GG^{-1}) = 0$ does not apply. We therefore must calculate $\delta\tilde{G}/\delta V$ in Eq. (23) directly using the expression for $\tilde{G}$ in (24). This yields

$$\begin{aligned}\tilde{\Sigma}(1,2) = {} & \Sigma^{gW}(1,3)\tilde{G}(3,4)(G^{d})^{-1}(4,2)\\ & + iW(3,1)g(1,4)\frac{\delta\tilde{\Sigma}(4,2)}{\delta V(3)}\\ & + iW(3,1)g(1,4)\tilde{\Sigma}(4,5)G^{d}(5,6)\Gamma^{d}(6,2,3),\end{aligned} \tag{27}$$

where

$$\Sigma^{gW}(1,2) = iW(2,1)g(1,2). \tag{28}$$

The effective vertex of the d subspace is given by

$$\Gamma^{d}(1,2,3) = -\frac{\delta(G^{d})^{-1}(1,2)}{\delta V(3)}. \tag{29}$$

Using (19) for $(G^{d})^{-1}$ the equation for the effective vertex Γ^{d} is given by

$$\begin{aligned}\Gamma^{d}(1,2,3) = {} & \Delta_{d}(1,3)\Delta_{d}(3,2) + \Delta_{d}(1,1')\frac{\delta\Sigma^{\rm eff}(1',2')}{\delta G^{d}(4,5)}\\ & \times \Delta_{d}(2',2)G^{d}(4,6)\Gamma^{d}(6,7,3)G^{d}(7,5).\end{aligned} \tag{30}$$

The right-hand side has been obtained by using the chain rule $\delta\Sigma^{\rm eff}/\delta V = (\delta\Sigma^{\rm eff}/\delta G^{d})(\delta G^{d}/\delta V)$ and the identity $\delta G^{d} = -G^{d}\delta(G^{d})^{-1}G^{d}$. Equation (27) is the desired equation to determine the effective self-energy.

Finally, the polarization propagator of the d subspace, $P^{d} = \delta\rho^{d}/\delta V$, where ρ^{d} is the electron density in the d subspace, is given by

$$P^{d}(1,2) = -iG^{d}(1,3)\Gamma^{d}(3,4,2)G^{d}(4,1^{+}). \tag{31}$$

Defining the rest of the polarization as $P^{r} = P - P^{d}$, it is straightforward to verify that with

$$W^{r} = v + vP^{r}W^{r} = [1 - vP^{r}]^{-1}v. \tag{32}$$

Equation (9) is equivalent to $W = v + vPW$. This completes our derivation of the closed set of equations for G^{d} and the effective self-energy, which is schematically given in Eqs. (6)–(10), and in detail in Eqs. (9), (19), (20), (30), and (31). This set of equations may be regarded as a set of downfolded Hedin's equations for a *given* W^{r}. This quantity may be calculated, for example, within the constrained random-phase approximation (CRPA) scheme [6]. For a given screened interaction W^{r}, the self-energies and hence the effective self-energy are functionals of G^{d} only, as can be seen from (27).

It is instructive to compute the effective self-energy in our formalism within the GWA, where the vertex correction $\delta\tilde{\Sigma}/\delta V$ is neglected and $\Gamma^{d} = 1$, in order to verify that the present formalism reproduces the definition of $\tilde{\Sigma}$ in (15). From (27)

$$\begin{aligned}\tilde{\Sigma}(1,4)G^{d}(4,2) = {} & \Sigma^{gW}(1,5)\tilde{G}(5,2) + iW(3,1)g(1,4)\\ & \times \tilde{\Sigma}(4,5)G^{d}(5,3)G^{d}(3,2).\end{aligned} \tag{33}$$

$g(1,4)\tilde{\Sigma}(4,5)G^{d}(5,3)$ on the right-hand side is a renormalization correction to $\tilde{g}$ in $\tilde{G}$ in (24). This together with W forms a self-energy correction to Σ^{gW}. Utilizing the relation in (15) we thus obtain

$$\tilde{\Sigma}(1,4)G^{d}(4,2) = \Sigma^{GW}(1,5)\tilde{G}(5,2), \tag{34}$$

where

$$\Sigma^{GW}(1,2) = iW(2,1)G(1,2), \tag{35}$$

in agreement with the expression in (15) when the full self-energy is approximated by the GWA.

The decomposition of the effective self-energy into Σ^d and Σ^{rd} allows for some general applications. One possible application is to devise a scheme beyond the GWA for the d subspace only, while keeping the contribution from the r subspace at the GW level. Thus, the d subspace, which is typically much smaller than the full Hilbert space, can be treated in a tractable way at a more sophisticated level than the GWA by including vertex corrections on the chosen d subspace only. The formalism also provides a route for numerical simplification of the GWA, by focusing on a certain subspace, which can be treated with high accuracy, while treating the rest of the subspace in an approximate scheme. For example, to understand the electronic properties of correlated materials may require accurate knowledge of the electronic structure around the Fermi level to within a few tens of meV. Full GW calculations for these complex systems with such high accuracy are not realistic but by focusing on the much smaller d subspace the computations may become feasible.

The set of equations for the downfolded self-energy provides a general framework for constructing theoretical models in the Green function language. It offers an alternative to model Hamiltonians, which cannot readily take into account frequency-dependent interactions. Rather than first mapping the full Hamiltonian to a model Hamiltonian with a static U and then solving the model using the Green function technique, the present formalism allows for a direct route to the self-energy with a fully frequency-dependent effective interaction. When W^r is approximated by a local and static value and Σ^{rd} and Σ^{drd} are neglected, it becomes evident that the set of equations may be interpreted as equivalent to the Hubbard model, provided the d subspace is chosen to be the same as the Hilbert space of the Hubbard model. Consequently, these two self-energy terms have no counterparts in the Hubbard model. Two ingredients are therefore missing in the Hubbard model, namely, the frequency dependence of U, which can be important [6], and the effects of the energy dependence of the self-energy arising from the r subspace. The present formalism can be used to improve the Hubbard or the Anderson impurity model by providing better parameters and by including the effects of the self-energy arising from the r subspace and the effects of the frequency-dependent W^r within, for example, the GWA.

In real applications, the choice of the d subspace is not always clear-cut. An example of this is provided by transition metals, where one would usually choose the $3d$ bands as the d subspace. However, due to the hybridization with the $4s$ and $4p$ bands, the $3d$ bands are not completely isolated. In this case, one possible solution is to first construct a set of Wannier orbitals following the postprocessing procedure of Marzari and Vanderbilt [8] or the preprocessing procedure of Andersen [9]. From this set of Wannier orbitals an isolated set of bands can be calculated forming a well-defined d subspace [10].

In conclusion, we have derived from the full many-body Hamiltonian a closed set of equations for the effective self-energy acting on a subspace of the full Hilbert space. The effective frequency-dependent interaction or the Hubbard U appears naturally in this formalism. This frequency-dependent interaction is usually not accounted for in conventional model Hamiltonians. The formalism provides a general framework for handling complex systems in which the main correlation effects are concentrated on a certain subspace, such is the case in many correlated materials characterized by partially filled narrow bands.

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