# 12 The GW+EDMFT Method

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### **1** Introduction

Dynamical mean-field theory (DMFT) [1] has been combined with density functional theory (DFT), commonly referred to as LDA+DMFT [2], which is regarded as a major breakthrough in the description of strongly correlated materials since it allows for a realistic description of the electronic structure of materials with strong dynamical correlations ranging from strongly correlated metals to Mott-Hubbard insulators. Starting from a DFT electronic structure as a background, a model Hamiltonian containing the strongly correlated manifold is constructed by making a tight-binding fit to the DFT bandstructure. This is usually done by using maximally localized Wannier functions [3]. The single-particle Hamiltonian is then complemented by a Hubbard-*U* term describing the onsite interaction between the strongly correlated states and a double-counting (DC) term is subtracted. This yields the DFT+DMFT Hamiltonian

$$\hat{H} = \sum_{\mathbf{R}i,\mathbf{R}'j,\sigma} \hat{c}^{\dagger}_{\mathbf{R}i\sigma} \left( H^{\mathrm{DFT}}_{\mathbf{R}i,\mathbf{R}'j,\sigma} - H^{\mathrm{DC}}_{\mathbf{R}i,\mathbf{R}'j,\sigma} \right) \hat{c}_{\mathbf{R}'j\sigma} + \frac{1}{2} \sum_{\mathbf{R},ijkl,\sigma\sigma'} \hat{c}^{\dagger}_{\mathbf{R}i\sigma} \hat{c}^{\dagger}_{\mathbf{R}j\sigma'} U^{\sigma\sigma'}_{ijkl} \hat{c}_{\mathbf{R}l\sigma'} \hat{c}_{\mathbf{R}k\sigma}, \quad (1)$$

where  $\mathbf{R}^i$  label an atomic site  $\mathbf{R}$  and an orbital i whereas  $\sigma$  labels the spin. The onsite effective Coulomb interaction U is defined as

$$U_{ijkl}^{\sigma\sigma'} = \int d^3r d^3r' \ \phi_i^{\sigma*}(\mathbf{r}) \phi_j^{\sigma'*}(\mathbf{r}') \ U(\mathbf{r}, \mathbf{r}') \ \phi_k^{\sigma}(\mathbf{r}) \phi_l^{\sigma'}(\mathbf{r}').$$
(2)

The problem defined by the Hamiltonian (1) can then be solved by using the DMFT machinery. By mapping the lattice problem to an impurity problem, this method captures the essential physics of local correlations responsible for many phenomena in strongly correlated systems, not accessible within the one-particle description. Notable among these are the Mott metal-to-insulator transition and Kondo effects, which are beyond static theories. The combination of DMFT and DFT opens up wider possibilities of investigating electronic properties of strongly correlated materials than is possible within earlier static theories such as the DFT+U scheme. It provides a coherent description of metal and insulator on equal footing.

The basic assumption of local self-energy, however, hinders applications of this scheme to phenomena involving long-range correlations such as charge disproportionation and superconductivity, as well as phenomena involving coupling to long-range charge and spin fluctuations. Even in iron, a seemingly simple system, it has been found that its Curie transition temperature  $T_{\rm C}$  from paramagnetic to ferromagnetic ordering is greatly overestimated when computed within DMFT. The predicted value of  $T_{\rm C}$  of iron by DMFT is 1900 K, almost twice as large as the experimental value of 1043 K [4]. It is likely that this large discrepancy has its origin in the long-range part of the self-energy. Also, there is evidence from photoemission experiments [5] for a strong momentum dependence of the mass renormalization of iron. Another phenomenon for which a nonlocal self-energy is expected to play an important role is the non-Fermi-liquid behavior in two dimensions arising from the van Hove singularity [6].

This local self-energy assumption is one of the fundamental problems inherent in DFT+DMFT. The other is the issue of double counting, inherited from the DFT+U scheme. There is also the problem of determining the Hubbard U, which is often treated as adjustable, making the

method less predictive when applied to real materials. In recent years, however, it has become possible to compute the Hubbard U for real materials using a number of methods such as the constrained LDA (cLDA) [7] or the constrained random-phase approximation (cRPA) [8]. There are thus two main aspects of DFT+DMFT which should be improved: the assumption of local self-energy and the problem of double counting. The assumption of the self-energy being local is expected to be valid when the correlated electrons are highly localized such that the local Coulomb interaction far exceeds the nonlocal one. As the correlated electrons become more itinerant, one anticipates that the nonlocal interaction becomes more significant and accordingly the importance of the nonlocal components of the self-energy increases. The second aspect, the double-counting problem, is a long-standing one, which was already recognized in the earlier LDA+U method. The problem arises from the nonlinear dependence on the density of the Kohn-Sham exchange-correlation energy, which renders it difficult, if not impossible, to remove the contribution from the density of the correlated electrons only.

One approach to address the above two issues is the combination of the GW method and extended DMFT (GW+EDMFT) [9,10], the former replaces the Kohn-Sham DFT. Being a Green function method, it is possible to remove the double-counting correction precisely. Moreover, the GW method was originally conceived to treat extended systems such as the electron gas so that it naturally incorporates nonlocal or long-range self-energies. Seen from the point of view of GW, the GW+EDMFT method can be regarded as a route of going beyond RPA by including vertex corrections in the form of local correlations.

# 2 GW method

In this section, the GW approximation (GWA) [11–13], which may be regarded as the background electronic structure in the GW+EDMFT method, is elaborated. The screened interaction, which is an essential part of the GW+EDMFT method, is discussed first in detail. The detailed derivations of the self-energy and response functions are given in Appendix A. In our notation, r is a combined variable for space and spin:  $r = (\mathbf{r}, \sigma)$ , and an integer represents space, spin, and time:  $n = (r_n, t_n)$ . Atomic units are used throughout where

$$m = \hbar = e = a_0 = 1. \tag{3}$$

#### 2.1 Screened interaction W

The screened interaction is an essential part of GWA as well as in the GW+EDMFT scheme. Due to the Coulomb repulsion and the exchange interaction, electrons tend to avoid each other, resulting in the formation of a screening hole around each electron. This leads to an effective screened interaction between an electron and the other electrons in the system, which is much smaller than the bare interaction, especially in metals. It is the density response functions that determine the screened interaction.

Within GWA, only the linear density response function is needed. Higher-order density response functions correspond to vertex corrections. The linear response theory can be established by applying an external time-dependent field  $V_{\text{ext}}$  to a system of electrons. The induced density to linear order in the applied field is given by

$$\rho_{\rm ind}(1) = \int d2 \ R(1,2) \, V_{\rm ext}(2) \,, \tag{4}$$

where R is the linear density response function. This induced density in turns generates an induced back potential, with v the bare interaction,

$$V_{\rm ind}(1) = \int d2 \ v(1-2) \ \rho_{\rm ind}(2) \,, \tag{5}$$

which screens the applied potential

$$V_{\rm tot}(1) = V_{\rm ext}(1) + V_{\rm ind}(1)$$
. (6)

The screened or total potential in terms of the linear response function is then given by

$$V_{\text{tot}}(1) = V_{\text{ext}}(1) + \int d2 \ v(1-2) \ \rho_{\text{ind}}(2)$$
  
=  $V_{\text{ext}}(1) + \int d2 \ d3 \ v(1-2) \ R(2,3) \ V_{\text{ext}}(3)$   
=  $\int d3 \Big[ \delta(1-3) + \int d2 \ v(1-2) \ R(2,3) \Big] V_{\text{ext}}(3) .$  (7)

The quantity in the square bracket can be identified as the inverse dielectric matrix

$$\varepsilon^{-1}(1,3) = \frac{\delta V_{\text{tot}}(1)}{\delta V_{\text{ext}}(3)} = \delta(1-3) + \int d2 \, v(1-2) \, R(2,3) \,. \tag{8}$$

Applying the above formula to the instantaneous Coulomb potential

$$v(1-2) = v(r_1 - r_2) \,\delta(t_1 - t_2) \tag{9}$$

as the applied perturbing field yields the screened Coulomb interaction

$$W(1,2) = \int d3 \,\varepsilon^{-1}(1,3) \,v(3-2) = v(1-2) + \int d3 \,d4 \,v(1-4) \,R(4,3) \,v(3-2) \,, \tag{10}$$

or in matrix form

$$W = \varepsilon^{-1}v = v + vRv.$$
<sup>(11)</sup>

Thus, W(1, 2) may be interpreted as a screened potential at space-time point 1 of a test charge located at space-time point 2. Fourier transforming Eq. (10) with respect to  $t = t_1 - t_2$  yields

$$W(r_1, r_2; \omega) = v(r_1 - r_2) + \int dr_3 dr_4 \ v(r_1 - r_4) R(r_4, r_3; \omega) v(r_3 - r_2) \,. \tag{12}$$

There are two limiting cases of interest. The static screened interaction  $W(r_1, r_2; \omega=0)$  can be seen as the time-average of  $W(r_1, r_2; t)$ , as follows from the definition of the Fourier transform. Equivalently, from Eq. (12), it is the screened interaction of the static bare interaction  $v(r_1-r_2)$  (without  $\delta(t_1-t_2)$ ). At the other end of the limit,  $W(r_1, r_2; \omega \to \infty)$  approaches the bare interaction  $v(r_1-r_2)$  since  $R(\omega)$  decays asymptotically as  $\sim 1/\omega^2$  as can be seen from Eq. (90). This simply means that at very high frequency the electrons cannot react sufficiently fast to the rapid variation of the applied potential so that screening is ineffective.

The screened interaction via the linear density response encodes the charge excitation spectrum of the system, as can be seen in Eq. (12). Thus, when the applied frequency matches a collective charge excitation (plasmon) energy of the system, a resonance peak in the imaginary part of W is expected to appear.

The correlation part of the screened interaction is the second term on the right-hand side of (12),

$$W^{c}(\omega) = v R(\omega) v.$$
(13)

Since the Coulomb interaction v is frequency independent, the spectral representation of  $W^c$  is identical in form to that of  $R(\omega)$  as in Eq. (91) of Appendix A

$$W^{c}(r_{1}, r_{2}; \omega) = \int_{-\infty}^{0} d\omega' \, \frac{B(r_{1}, r_{2}; \omega')}{\omega - \omega' - i\eta} + \int_{0}^{\infty} d\omega' \, \frac{B(r_{1}, r_{2}; \omega')}{\omega - \omega' + i\eta} \,, \tag{14}$$

where

$$B(\omega) = v S(\omega) v = -\frac{1}{\pi} \operatorname{sign}(\omega) \operatorname{Im} W(\omega), \qquad (15)$$

with S given in Eq. (92). Since  $S(\omega)$  is anti-symmetric, so is  $B(\omega)$ :

$$B(-\omega) = -B(\omega).$$
(16)

#### 2.2 Random-phase approximation

It is unrealistic to calculate the exact response functions for real systems since it would require computation of the exact excited states of the many-electron system. This necessitates approximations, as commonly the case in many-electron theory. A practical and highly successful approximation is the random-phase approximation (RPA) [14]. As discussed later, it can be understood as the time-dependent Hartree approximation since it takes into account only the change in the Hartree potential with respect to the time-dependent perturbing field.

In the Green function formalism, the response function is expressed as in Eq. (88), and using the identity  $\delta G = -G \, \delta G^{-1} G$  as in Eq. (84), we find

$$R(1,2) = i \int d3 \, d4 \, G(1,3) \, \frac{\delta G^{-1}(3,4)}{\delta V_{\text{ext}}(2)} G(4,1^+) \,. \tag{17}$$

Using  $\delta G^{-1}/\delta V_{\text{ext}}$  as in Eq. (85) we obtain

$$R(1,2) = -i \int d3 \, d4 \, G(1,3) \left( \left[ \delta(3-2) + \frac{\delta V_{\rm H}(3)}{\delta V_{\rm ext}(2)} \right] \delta(3-4) + \frac{\delta \Sigma(3,4)}{\delta V_{\rm ext}(2)} \right) G(4,1^+) \,. \tag{18}$$

RPA corresponds to neglecting the term  $\delta \Sigma / \delta V_{\text{ext}}$  in (18) yielding

$$R(1,2) = -i \int d3 \ G(1,3) \left[ \delta(3-2) + \frac{\delta V_{\rm H}(3)}{\delta V_{\rm ext}(2)} \right] G(3,1^+)$$
  
=  $P(2,1) + \int d3 \ d4 \ P(1,3) \ v(3-4) \ R(4,2) \ .$  (19)



**Fig. 1:** *Feynman diagrams for the screened interaction in the random-phase approximation and the self-energy within the GW approximation. Figures courtesy of Fredrik Nilsson.* 

where

$$P(1,2) = -iG(1,2)G(2,1^+).$$
(20)

Thus, in RPA only the change in the Hartree potential is taken into account and for this reason it can be regarded as the time-dependent Hartree approximation. In frequency space, the polarization P in RPA is given by

$$P(r_1, r_2; \omega) = -i \int \frac{d\omega'}{2\pi} G(r_1, r_2; \omega + \omega') G(r_2, r_1; \omega').$$
(21)

Replacing G by  $G^0$  given in Eq. (109) yields

$$P^{0}(r_{1}, r_{2}; \omega) = \sum_{n}^{\text{occ}} \sum_{n'}^{\text{unocc}} \left( \frac{\phi_{n}(r_{2})\phi_{n'}^{*}(r_{2})\phi_{n}^{*}(r_{1})\phi_{n'}(r_{1})}{\omega - (\varepsilon_{n'} - \varepsilon_{n} - i\delta)} - \frac{\phi_{n}(r_{1})\phi_{n'}^{*}(r_{1})\phi_{n}^{*}(r_{2})\phi_{n'}(r_{2})}{\omega + (\varepsilon_{n'} - \varepsilon_{n} - i\delta)} \right).$$
(22)

#### 2.3 GW self-energy

Within the GW approximation [11–13], the self-energy is given by

$$\Sigma(1,2) = iG(1,2) W(2,1), \qquad (23)$$

where W is the screened Coulomb interaction, usually calculated within RPA. Using the convolution theorem, the Fourier transform into frequency space yields

$$\Sigma(r_1, r_2; \omega) = i \int \frac{d\omega'}{2\pi} G(r_1, r_2; \omega + \omega') W(r_2, r_1; \omega') e^{i\eta\omega'}.$$
(24)

The screened interaction and the corresponding GW self-energy within RPA are shown in Fig. 1.

GWA has been found to be very successful in providing an accurate electronic structure for weakly to moderately correlated systems, whose valence electrons usually originate from s- or p-orbitals. Being based on perturbation theory, GWA is not expected to be able to describe the Mott metal-to-insulator transition in strongly correlated systems. To see the problem with the

GWA consider the expression for the imaginary part of the Green function, which is proportional to the spectral function:

$$\operatorname{Im} G(\omega) = \frac{\operatorname{Im} \Sigma(\omega)}{\left(\omega - \varepsilon - \operatorname{Re} \Sigma(\omega)\right)^2 + \left(\operatorname{Im} \Sigma(\omega)\right)^2}.$$
(25)

In order to open up a gap from a metallic state, Im  $\Sigma(\omega)$ , which is inversely proportional to the quasiparticle life-time, should become infinite around the Fermi level so that the quasiparticle weight present in the metal at the Fermi level is removed. The transition from a metal to a Mott insulator poses a fundamental difficulty because the GW self-energy varies as  $\omega^2$  around the Fermi level, characteristic of a Fermi liquid, as shown below. From Eq. (14) and using (16)

$$\frac{\partial}{\partial\omega}W^{c}(r_{1}, r_{2}; \omega)\Big|_{\omega=0} = -\int_{-\infty}^{0} d\omega' \frac{B(r_{1}, r_{2}; \omega')}{(-\omega' - i\eta)^{2}} - \int_{0}^{\infty} d\omega' \frac{B(r_{1}, r_{2}; \omega')}{(-\omega' + i\eta)^{2}} \\
= +\int_{0}^{\infty} d\omega' \frac{B(r_{1}, r_{2}; \omega')}{(\omega' - i\eta)^{2}} - \int_{0}^{\infty} d\omega' \frac{B(r_{1}, r_{2}; \omega')}{(-\omega' + i\eta)^{2}} = 0, \quad (26)$$

Since  $B(\omega)$  is proportional to Im  $W(\omega)$  and recognizing that B(0) = 0, it follows from Eqs. (113) and (114) that for small  $\omega$  the imaginary part of the self-energy behaves as

$$\Gamma(\omega) \propto \omega^2.$$
 (27)

This implies that the GW self-energy cannot not open up a Mott gap starting from a metallic band structure.

## **3** *GW*+EDMFT method

The problem with GWA in treating strongly correlated systems such as Mott insulators motivates the combination of GWA and extended DMFT (EDMFT). On the other hand, from the point of view of DMFT, GWA provides a route for taking into account nonlocal self-energy, missing in DMFT or EDMFT.

### 3.1 $G^0W^0$ +DMFT

It is instructive to first consider the combination of the one-shot GWA,  $\Sigma = iG^0W^0$ , with the standard DMFT. This combination may be seen as a simple extension of the DFT+DMFT scheme, in which the background DFT electronic structure is replaced by the GW one. Apart from improving the band structure, the double-counting problem inherent in the DFT+DMFT scheme is avoided. However, there are different ways of removing the double-counting term when self-consistency is not imposed. Either one removes the local projection of the lattice GW self-energy or one removes the impurity GW self-energy. These two double-counting forms are not strictly the same.

In the  $G^0W^0$ +DMFT scheme the self-energy is given by

$$\hat{\Sigma}(\omega) = \sum_{\mathbf{k}nn'} |\psi_{\mathbf{k}n}\rangle \, \Sigma_{nn'}^{GW}(\mathbf{k},\omega) \, \langle\psi_{\mathbf{k}n'}| + \sum_{mm'} |\phi_m\rangle \left(\Sigma_{mm'}^{imp}(\omega) - \Sigma_{mm'}^{DC}(\omega)\right) \langle\phi_{m'}| \,.$$
(28)

Here  $\psi_{\mathbf{k}n}$  are the DFT eigenstates and  $\phi_m$  the localized orbitals, e.g., the maximally localized Wannier orbitals constructed from the correlated states.  $\Sigma_{nn'}^{GW}$  is the GW self-energy obtained from a one-shot GW calculation based on the DFT band structure and  $\Sigma^{\text{imp}}$  the impurity selfenergy calculated with a dynamical U computed using the constrained RPA (cRPA) method.  $\Sigma_{nm'}^{\text{DC}}$  is the double-counting correction, defined as

$$\Sigma_{mm'}^{\rm DC}(\omega) = i \sum_{m_1m_2} \int \frac{d\omega'}{2\pi} G_{m_1m_2}^{\rm loc}(\omega + \omega') W_{mm_1m_2m'}^{\rm loc}(\omega') , \qquad (29)$$

where  $G_{m_1m_2}^{\text{loc}}$  is the local projection of the DFT Green function expressed in the localized orbitals. In Matsubara frequencies,

$$\Sigma_{mm'}^{\rm DC}(i\omega_n) = -\sum_{m_1m_2} \frac{1}{\beta} \sum_k G_{m_1m_2}^{\rm loc}(i\omega_n + i\nu_k) W_{mm_1m_2m'}^{\rm loc}(i\nu_k), \qquad (30)$$

where  $\omega_n$  and  $\nu_k$  are, respectively, the fermionic and bosonic Matsubara frequencies, and  $\beta$  is the inverse temperature. The two different ways of subtracting the double-counting term arise from the ambiguity in  $W^{\text{loc}}$  when it is not determined self-consistently. If  $W^{\text{loc}}$  is calculated from the impurity problem one has

$$W^{\rm loc}(\omega) = \left(1 - U(\omega)P^{\rm loc}(\omega)\right)^{-1}U(\omega),\tag{31}$$

in which  $P^{\text{loc}}$  is the local projection of the RPA polarization within the correlated subspace and  $U(\omega)$  is obtained from cRPA. This definition of  $W^{\text{loc}}$  corresponds to the fully screened interaction of the impurity model computed within GWA. On the other hand,  $W^{\text{loc}}$  can also be defined to be the local projection of the lattice W onto the correlated subspace

$$W^{\rm loc}(\omega) = \sum_{\mathbf{k}} W(\omega, \mathbf{k}), \qquad (32)$$

which is not necessarily the same as the one in Eq. (29), when self-consistency is not imposed. In Fig. 2 the quasiparticle bandstructure and the k-integrated spectra of a prototypical correlated metal SrVO<sub>3</sub> within the  $G^0W^0$ +DMFT scheme are shown and compared with the results obtained from the one-shot  $G^0W^0$  and LDA+DMFT. The one-shot  $G^0W^0$  quasiparticle dispersion shows a band narrowing compared with that of LDA, as expected. The  $G^0W^0$  occupied band width of 0.8 eV, while a significant improvement over the LDA band width, is still too large compared with the value measured in photoemission experiment, which is about 0.6 eV [16]. In contrast, the LDA+DMFT calculations, which have been performed using a frequency-dependent U, result in an occupied band width of only 0.4 eV, which is significantly smaller than the experimental value as can be seen in Fig. 2. There is a very large band narrowing compared with the starting LDA bandstructure. This band narrowing is significantly stronger than in the standard LDA+DMFT with a static U [17]. One may then conclude that the frequency dependence of U effectively enhances correlations, which would correspond to a larger static U. The  $G^0W^0$ +DMFT scheme, on the other hand, brings the quasiparticle dispersion in close agreement with experiment with an occupied band width of slightly smaller than 0.6 eV. It is interesting to observe that a nonlocal self-energy tends to increase the band width while a local self-energy narrows the band width.



**Fig. 2:** Quasiparticle bandstructure (left) and k-integrated spectral function (right) of  $SrVO_3$  within the  $G^0W^0$ +DMFT scheme, compared with the results based on  $G^0W^0$  and LDA + DMFT with dynamic U. The figures are taken from Sakuma et al. [15].

#### **3.2 Extended DMFT**

In order to develop the full GW+EDMFT scheme it is necessary to extend DMFT in order to take into account long-range interactions. The  $G^0W^0$ +DMFT scheme described in the previous section does not take into account self-consistency in the local screened interaction. As in the original DMFT, self-consistency is only imposed on the local Green function, which may be regarded as a one-particle correlation function. The concept of DMFT, however, is more general and can be extended to higher-order correlation functions. A natural step beyond the original DMFT is to impose self-consistency on the local screened interaction  $W_{loc}$  or equivalently the local (charge) response function, which is a two-particle correlation function. This extended DMFT (EDMFT) [9] furnishes a means to solve an extended Hubbard model with nonlocal interactions using the framework of DMFT. As in the original DMFT, the auxiliary system is the And erson impurity model but in addition to the hybridization function (Weiss field)  $\Delta(\tau)$ , which determines the local Green function, another "Weiss field",  $\mathcal{U}(\tau)$ , is needed to determine the local screened interaction  $W_{\text{loc}}$ . In this case  $\mathcal{U}(\tau)$  plays the role of a dynamic effective impurity interaction in such a way that when it is screened by the impurity polarization reproduces at selfconsistency the local projection of the lattice screened interaction. Thus,  $\mathcal{U}(\tau)$  is analogous to  $\mathcal{G}(\tau)$ , which when "screened" by the impurity self-energy reproduces at self-consistency the local projection of the lattice Green function. It can then be seen that the impurity polarization is analogous to the impurity self-energy.

The effective impurity action is given by

$$S_{\rm imp} = -\int_0^\beta d\tau d\tau' \sum_\sigma d^*_\sigma(\tau) \,\mathcal{G}^{-1}(\tau - \tau') \,d_\sigma(\tau') + \int_0^\beta d\tau d\tau' \,n_\uparrow(\tau) \,\mathcal{U}(\tau - \tau') \,n_\downarrow(\tau') \,, \quad (33)$$

where

$$-\left(\frac{\partial}{\partial\tau}-\mu\right)\mathcal{G}(\tau-\tau')-\int_{0}^{\beta}d\tau''\Delta(\tau-\tau'')\mathcal{G}(\tau''-\tau')=\delta(\tau-\tau'),\tag{34}$$

or in frequency space

$$\mathcal{G}^{-1}(i\omega_n) = i\omega_n + \mu - \Delta(i\omega_n). \tag{35}$$

The self-consistency condition in EDMFT requires self-consistency in both the local Green function and the local screened interaction:

$$G_{\rm loc} = G_{\rm imp}, \quad W_{\rm loc} = W_{\rm imp}, \tag{36}$$

where

$$W_{\rm imp}(\omega) = \mathcal{U}(\omega) + \mathcal{U}(\omega)P_{\rm imp}(\omega)W_{\rm imp}(\omega), \qquad (37)$$

and  $W_{\rm loc}$  is the local projection of the lattice screened interaction W obtained from the equation

$$W(\omega) = U + UP(\omega)W(\omega), \tag{38}$$

where P is the lattice polarization. U is the bare interaction which may contain long-range (offsite) components. Comparison with the Dyson equation for the Green function,

$$G(\omega) = G^{0}(\omega) + G^{0}(\omega)\Sigma(\omega)G(\omega), \qquad (39)$$

shows that the polarization P is the analog of the self-energy whereas W is the analog of the Green function, whereas U plays the role of the bare propagator  $G^0$ . For the Hubbard model with onsite interactions we have  $U_{ij} = U\delta_{ij}$  which yields  $\mathcal{U} = U$  by construction and in this case EDMFT reduces to the regular DMFT. For a system with offsite interactions this is not the case. The effects of the long-range interactions are taken into account in the dynamically screened effective impurity interaction  $\mathcal{U}(\omega)$ . In real materials U will be the bare Coulomb interaction  $v(\mathbf{r}-\mathbf{r}')$ .

#### 3.2.1 Self-consistency loop in EDMFT

To initiate the self-consistency loop in EDMFT, the local polarization and the local self-energy are, respectively, set to  $P_{\rm imp}$  and  $\Sigma_{\rm imp}$ 

$$P_{\rm loc} = P_{\rm imp}, \quad \Sigma_{\rm loc} = \Sigma_{\rm imp}.$$
 (40)

The basic assumption of EDMFT, in which the lattice self-energy and the lattice polarization are approximated by their local values, is applied

$$P_{\mathbf{k}} = P_{\text{loc}}, \quad \Sigma_{\mathbf{k}} = \Sigma_{\text{loc}}. \tag{41}$$

From the Dyson equation the lattice Green function  $G_{\mathbf{k}}$  and the lattice screened interaction  $W_{\mathbf{k}}$  are computed

$$G_{\mathbf{k}} = G_{\mathbf{k}}^0 + G_{\mathbf{k}}^0 \Sigma_{\mathbf{k}} G_{\mathbf{k}} \,, \tag{42}$$

$$W_{\mathbf{k}} = U_{\mathbf{k}} + U_{\mathbf{k}} P_{\mathbf{k}} W_{\mathbf{k}} \,, \tag{43}$$

from which the local Green function and the local screened interaction are obtained

$$G_{\rm loc} = \sum_{\mathbf{k}} G_{\mathbf{k}}, \quad W_{\rm loc} = \sum_{\mathbf{k}} W_{\mathbf{k}}.$$
(44)

Applying the EDMFT self-consistency condition,

$$G_{\rm imp} = G_{\rm loc}, \quad W_{\rm imp} = W_{\rm loc}, \tag{45}$$

allows us to construct the fermionic Weiss field  $\mathcal{G}$  and the effective impurity interaction (bosonic Weiss field)  $\mathcal{U}$  from the Dyson equations

$$G_{\rm imp} = \mathcal{G} + \mathcal{G} \Sigma_{\rm imp} G_{\rm imp} \quad \rightarrow \quad \mathcal{G} = G_{\rm imp} \left( 1 + \Sigma_{\rm imp} G_{\rm imp} \right)^{-1}, \tag{46}$$

$$W_{\rm imp} = \mathcal{U} + \mathcal{U} P_{\rm imp} W_{\rm imp} \quad \rightarrow \quad \mathcal{U} = W_{\rm imp} \left( 1 + P_{\rm imp} W_{\rm imp} \right)^{-1}. \tag{47}$$

G and U provide an input for solving the impurity problem with the action defined in Eq. (33), yielding a new  $G_{imp}$  and the impurity charge susceptibility  $\chi_{imp}$ . A new impurity self-energy and a new impurity polarization are extracted as

$$\Sigma_{\rm imp} = \mathcal{G}^{-1} - G_{\rm imp}^{-1},\tag{48}$$

$$P_{\rm imp} = \chi_{\rm imp} \left( 1 + \mathcal{U} \, \chi_{\rm imp} \right)^{-1}. \tag{49}$$

The last equation is formally equivalent to

$$P_{\rm imp} = \mathcal{U}^{-1} - W_{\rm imp}^{-1}$$
(50)

but it is numerically preferable since it avoids possible singularities in  $\mathcal{U}^{-1}$  and  $W_{imp}^{-1}$ .

The loop is now continued until the EDMFT self-consistency condition in Eq. (36) is fulfilled. The concept of DMFT is quite general and it is possible to extend the self-consistency condition on local quantities to a three-particle correlation function, such as the one formulated in the TRILEX approach [18], and to higher-order correlation functions.

#### 3.3 GW+EDMFT

We are now in the position to construct the full GW+EDMFT scheme [9, 10]. The formal functional derivation is given in Appendix B. As in EDMFT, two propagators are now at disposal: the fermionic one-particle Green function G and the bosonic screened interaction W. These two propagators constitute the basic variables in the free-energy functional  $\Psi$  (see Appendix B). The lattice self-energy  $\Sigma$  and polarization P are no longer approximated by the corresponding local impurity quantities but they now in addition contain nonlocal components from the GW self-energy. The self-energy is then given by

$$\Sigma(\mathbf{k}, i\omega_n) = \Sigma_{\rm imp}(i\omega_n) + \Sigma_{\rm nonloc}^{GW}(\mathbf{k}, i\omega_n).$$
(51)

Similarly, the polarization is given by

$$P(\mathbf{k}, i\omega_n) = P_{\rm imp}(i\omega_n) + P_{\rm nonloc}^{GW}(\mathbf{k}, i\omega_n).$$
(52)

 $\Sigma_{\rm imp}$  and  $P_{\rm imp}$  are, respectively, the EDMFT impurity self-energy and polarization. The above two expressions are free from double counting since both  $\Sigma_{\rm nonloc}^{GW}$  and  $P_{\rm nonloc}^{GW}$  contain no local components, which have been subtracted out from the corresponding lattice quantities

$$\Sigma_{\text{nonloc}}^{GW}(\mathbf{k}, i\omega_n) = \Sigma^{GW}(\mathbf{k}, i\omega_n) - \Sigma_{\text{loc}}^{GW}(i\omega_n)$$
(53)

$$P_{\text{nonloc}}^{GW}(\mathbf{k}, i\omega_n) = P^{GW}(\mathbf{k}, i\omega_n) - P_{\text{loc}}^{GW}(i\omega_n) \,.$$
(54)

The lattice Green function and screened interaction are calculated from  $\Sigma$  and P using the Dyson equations

$$G = G^0 + G^0 \Sigma G, \tag{55}$$

$$W = v + vPW, (56)$$

where  $G^0$  is the Hartree Green function and v is the bare interaction.

#### **3.4** Multitier *GW*+EDMFT scheme

To apply the full GW+EDMFT scheme described in the previous sections to real materials, due to computational complexity it is inevitable to adopt approximations. Fortunately, in most correlated materials, for which the GW+EDMFT scheme is designed, it is usually the case that the electronic structure can be separated into correlated and noncorrelated bands. Although the separation is arbitrary, physical consideration imposes a strong constraint on the choice of correlated bands, which are typically partially filled narrow bands across or around the chemical potential originating from localized states of 3d- or 4f-orbitals.

In the multitier GW+EDMFT scheme [19, 20], the Hilbert space is decomposed into a correlated subspace and the rest. A concrete example is provided by the prototypical correlated metal SrVO<sub>3</sub> in which the narrow partially filled  $t_{2g}$  band is naturally chosen as the correlated subspace. GW+EDMFT self-consistency is performed only in the correlated subspace whereas contribution to the self-energy from the rest of the Hilbert space is accounted for by a one-shot GW. There is a physical motivation for this strategy. It is known that fully self-consistent GW alone tends to worsen the one-shot results (in comparison with experiment). Within the correlated subspace, however, the detrimental effects of self-consistency on the GW self-energy are expected to be counteracted by vertex corrections in the form of the EDMFT self-energy.

It is possible to introduce an intermediate subspace  $\mathcal{I}$ , containing an additional few bands around the Fermi level considered to be important for the low-energy physics. The intermediate subspace then includes a set of correlated bands crossing the Fermi energy and, in addition, a few more weakly correlated bands which hybridize with the correlated bands. The correlated subspace  $\mathcal{C}$  is spanned by the narrow bands that cannot be accurately described by GWA. In this case, the GW+EDMFT self-consistency is performed within the intermediate subspace  $\mathcal{I}$ . In Fig. 3 the different subspaces and tiers are illustrated. Together with the three subspaces there are three different tiers that work on the respective subspace. In tier III the full space is treated within the one-shot GWA ( $G^0W^0$ -approximation).



**Fig. 3:** Upper panel: Schematic figure of the subspaces in multitier GW+EDMFT, reproduced from Nilsson et al. [19]. Lower panel: Schematic figure of how the tiers work on the different subspaces, reproduced from Boehnke et al. [20].

In practice, one starts with a DFT bandstructure and performs a one-shot GW calculation on the entire Hilbert space (tier III) yielding a self-energy  $\Sigma^{G^0W^0}$ . A calculation of the bare effective interaction,  $U(\omega)$ , within the intermediate subspace is also performed using the constrained random-phase approximation (cRPA) method. The effective bare propagator ( $G_k^0$ ) and interaction ( $U_q$ ) on the intermediate subspace are then computed as

$$G_{\mathbf{k}}^{0-1} = \underbrace{\mathrm{i}\omega_n + \mu - \varepsilon_{\mathbf{k}}^{\mathrm{KS}}}_{G_{\mathrm{KS},\mathbf{k}}^{-1}} + V_{\mathrm{XC},\mathbf{k}} - \underbrace{\left(\Sigma_{\mathbf{k}}^{G^0W^0} - \Sigma_{\mathbf{k}}^{G^0W^0}\big|_{I}\right)}_{\Sigma_{\mathbf{k}}^{\mathrm{r}}}$$
(57)

$$U_{\mathbf{q}}^{-1} = v_{\mathbf{q}}^{-1} - \underbrace{\left(P_{\mathbf{q}}^{G^{0}G^{0}} - P_{\mathbf{q}}^{G^{0}G^{0}}\big|_{I}\right)}_{P_{\mathbf{q}}^{r}}.$$
(58)

Here  $\varepsilon_{\mathbf{k}}^{\mathrm{KS}}$  are the Kohn-Sham (KS) DFT eigenenergies,  $V_{\mathrm{XC},\mathbf{k}}$  the exchange-correlation potential,  $\Sigma_{\mathbf{k}}^{G^0W^0}$  the one-shot GW self-energy,  $v_{\mathbf{q}}$  the bare Coulomb interaction and  $P_{\mathbf{q}}^{G^0G^0}$  the RPA polarization computed from the Kohn-Sham bandstructure. The notation  $A|_I$  means that all internal sums when computing the quantity A are restricted to the intermediate subspace. U is the effective Coulomb interaction on the intermediate subspace computed using cRPA and  $G^0$  is the analogous downfolding for the single-particle Green function. The exchange-correlation potential is removed from the Kohn-Sham Green function and replaced by the  $G^0W^0$  self-energy. The exact double counting  $\Sigma^{G^0W^0}|_I$  is known and thus the scheme is free from the double-counting problem.



**Fig. 4:** Self-consistency flow chart of the multitier GW+EDMFT scheme. Reproduced from Nilsson et al. [19].

The bare propagators  $G^0$  and U are computed only once and used in subsequent self-consistent GW+EDMFT calculation within the intermediate subspace. At each iteration the GW selfenergy within the intermediate subspace (tier II) is computed, while the local part of the selfenergy within the correlated subspace C is computed using the EDMFT impurity solver (tier I). The two self-energies are then summed with the double counting removed. The screened interaction W is computed in a similar fashion in which the polarization plays the role of the self-energy. This yields the final equations for the single particle Green function G and screened interaction W

$$G_{\mathbf{k}}^{-1} = \underbrace{G_{\mathbf{k}}^{0-1}}_{\text{TIER III}} - \underbrace{\left( \sum_{\mathbf{k}}^{GW} \Big|_{I} - \sum^{GW} \Big|_{C,\text{loc}} + \Delta V_{\text{H}} \Big|_{I} \right)}_{\text{TIER II}} - \underbrace{\sum_{i=1}^{\text{imp}} \Big|_{C,\text{loc}}}_{\text{TIER II}} .$$
(59)

$$W_{\mathbf{q}}^{-1} = \underbrace{U_{\mathbf{q}}^{-1}}_{\text{TIER III}} - \underbrace{\left(P_{\mathbf{q}}^{GG}\big|_{I} - P^{GG}\big|_{C,\text{loc}}\right)}_{\text{TIER II}} - \underbrace{P^{\text{imp}}\big|_{C,\text{loc}}}_{\text{TIER I}}.$$
(60)

 $\Sigma_{\mathbf{k}}^{GW}$  is the self-energy computed within the self-consistent GWA and  $\Sigma^{imp}$  the local impurity self-energy computed within EDMFT.  $\Delta V_{\rm H}|_{I}$  is the change of the Hartree potential within the intermediate subspace.  $\Sigma^{GW}|_{C,\text{loc}}$  and  $P^{GG}|_{C,\text{loc}}$  are, respectively, the double counting for the self-energy and the polarization in tier II.



**Fig. 5:** LDA bandstructures of  $SrMoO_3$  and  $SrVO_3$  with  $t_{2g}$  bands crossing the Fermi level. *Figures courtesy of Fredrik Nilsson.* 

The impurity EDMFT self-energy and polarization provide local vertex corrections within the correlated subspace through, respectively,  $\Sigma^{\text{imp}}|_{C,\text{loc}}$  and  $P^{\text{imp}}|_{C,\text{loc}}$  at each iteration in the self-consistency cycle in tier II. This feature, a feedback between local and nonlocal correlations both on the single-particle and two particle level, is not accounted for in one-shot GW+DMFT. The self-consistency cycle is illustrated in Fig. 4.

#### 3.5 Applications to cubic perovskites SrMoO<sub>3</sub> and SrVO<sub>3</sub>

Application of the GW+EDMFT method to real materials is illustrated for the case of cubic perovskites SrMoO<sub>3</sub> and SrVO<sub>3</sub>. In Fig. 5 the LDA bandstructure for the two compounds are shown. Both compounds have an open *d*-shell, split into the  $t_{2g}$  and  $e_g$  components by the crystal field, with the  $e_g$  bands lying higher in energy and unoccupied. As can be seen in the figure, the  $t_{2g}$ -band of SrMoO<sub>3</sub> is wider than that of SrVO<sub>3</sub> since the 4*d*-orbitals of Mo are more spread than the 3*d*-orbitals of V. It is then expected that SrMoO<sub>3</sub> is less correlated than SrVO<sub>3</sub> and as such these compounds provide an interesting testing ground for GW+EDMFT.

LDA+DMFT calculations cannot reproduce the satellite structures of  $SrMoO_3$  unless U is chosen to be unrealistically large. This suggests that the satellites may be of different nature from the commonly assumed Hubbard bands and it was proposed that they could be plasmons arising from long-range charge fluctuations [21].



**Fig. 6:** Spectral functions of  $SrMoO_3$  and  $SrVO_3$  calculated using GW+EDMFT. For  $SrMoO_3$  comparison is made with LDA and one-shot GW as well as with experiment. For  $SrVO_3$  comparison is made with one-shot and fully self-consistent GW. The lower figures show the imaginary part of the screened interaction. The figures are reproduced from Boehnke et al. [20] and Nilsson et al. [19]. The experimental spectra for  $SrMoO_3$  is taken from Wadati et al. [21].

Contrary to  $SrMoO_3$  the electronic structure of  $SrVO_3$  can be reproduced relatively well within LDA+DMFT with a suitable choice of U [17]. Experimentally, a significantly renormalized quasiparticle peak compared with the LDA bandwidth as well as upper and lower satellite structures can be observed.  $SrVO_3$  has therefore been commonly thought of as a strongly correlated metal with upper and lower Hubbard bands.

It has however been found that the spectral function of  $SrVO_3$  can be well reproduced within the cumulant expansion [22]. The cumulant expansion describes long-range charge fluctuations (plasmons) but cannot account for the strong local correlations giving rise to Hubbard bands. This raises the question concerning the physical origin of the satellites in  $SrVO_3$ , whether they are collective excitations or atomic in origin. This issue can neither be addressed by DMFT nor the cumulant method since the former accounts only local correlations but ignores long-range charge fluctuations whereas the latter is a theory constructed to describe a coupling between electrons and bosonic excitations such as plasmons and phonons. *GW*+EDMFT, on the other hand, accounts for these two aspects of local and nonlocal correlations and is therefore well suited to investigate the nature of the satellites in  $SrVO_3$ . titier GW+EDMFT method are shown. In these calculations the intermediate subspace (and hence the self-consistency) was restricted to the  $t_{2g}$ -bands. Information about collective charge excitations in Im W is manifested as peaks at the excitation energies. From Fig. 6 it is quite evident that these excitations are present in both SrMoO<sub>3</sub> and SrVO<sub>3</sub>. In SrMoO<sub>3</sub> these long-range charge fluctuations give rise to a shoulder structure in the spectral function. This conforms with the suggestion by Wadati *et al.* [21] that the shoulder structure is of plasmonic origin although the satellite position and weight are lower compared with experiment. However, oxygen vacancies can lead to an overestimation of the satellite weight [23] in these materials.

More revealing is the results for  $SrVO_3$ . The feedback from long-range screening included in the self-consistency cycle reduces the effective impurity interaction  $\mathcal{U}$  substantially. It is clear from Fig. 6 that the self-consistently computed  $\mathcal{U}$  for  $SrVO_3$  is much too small to account for the satellites as Hubbard bands, whose separation is approximately given by  $\mathcal{U}(0)$ . Similar to  $SrMoO_3$ , long-range charge fluctuations give rise to a peak in Im W which in turn yields plasmon satellites in the spectral function. Thus, these calculations strongly suggest that  $SrVO_3$ is a moderately correlated metal with plasmon satellites rather than Hubbard bands of atomic origin. It is worth noting that the quasiparticle renormalization in GW+EDMFT is slightly underestimated compared with experiment, which points to the important role of nonlocal vertex corrections beyond GW+EDMFT.

*GW*+EDMFT has also been applied to investigate screening from  $e_g$ -states and antiferromagnetic correlations in  $d^{(1,2,3)}$ -perovskites [24] as well as normal state of Nd<sub>1-x</sub>Sr<sub>x</sub>NiO<sub>2</sub> [25].

# Appendices

# A Derivation of the GW approximation

In this Appendix, the GW approximation is derived in detail. The derivations have been carried out for zero temperature, but extension to finite temperature is quite straightforward. In the occupation number representation, the electronic part of the Hamiltonian is given by

$$\hat{H} = \int dr \,\hat{\psi}^{\dagger}(r) \,h_0(r) \,\hat{\psi}(r) + \frac{1}{2} \int dr dr' \,\hat{\psi}^{\dagger}(r) \,\hat{\psi}^{\dagger}(r') \,v(r-r') \,\hat{\psi}(r') \,\hat{\psi}(r) \,, \tag{61}$$

where

$$h_0 = -\frac{1}{2}\nabla^2 + V(r) \text{ and } v(r-r') = \frac{1}{|\mathbf{r} - \mathbf{r}'|}.$$
 (62)

 $\hat{\psi}(r)$  is the field operator which annihilates an electron at r and  $\hat{\psi}^{\dagger}(r)$  is its conjugate which creates an electron at r. In our notation, r is a combined variable for space and spin:  $r = (\mathbf{r}, \sigma)$  and an integer represents space, spin, and time:  $n = (r_n, t_n)$ . Atomic units are used throughout, where

$$m = \hbar = e = a_0 = 1. \tag{63}$$

The atomic unit of energy is Hartree and one Hartree is equal to 2 Rydberg or about 27.2 eV.

#### A.1 Equation of motion

We will define and work out the self-energy using the equation of motion approach. From the equation of motion of the field operator

$$i\frac{\partial\psi_{\rm H}(1)}{\partial t_1} = \left[\hat{\psi}_{\rm H}(1), \hat{H}\right],\tag{64}$$

one obtains

$$\left(i\frac{\partial}{\partial t_1} - h_0(1)\right)G(1,2) + i\int d3 \ v(1-3) \ G^{(2)}(1,2,3,3^+) = \delta(1-2),\tag{65}$$

where

$$v(1-2) = v(r_1 - r_2) \,\delta(t_1 - t_2) \,, \tag{66}$$

and  $G^{(2)}$  is the two-particle Green function defined as

$$G^{(2)}(1,2,3,4) = (-i)^2 \langle \Psi_0 | T \ \hat{\psi}_{\rm H}(1) \ \hat{\psi}_{\rm H}^{\dagger}(2) \ \hat{\psi}_{\rm H}(3) \ \hat{\psi}_{\rm H}^{\dagger}(4) | \Psi_0 \rangle.$$
(67)

The notation  $3^+$  means  $(r_3, t_3^+)$ .

The equation of motion of the two-particle Green function will in turn generate the three-particle Green function and so forth resulting in a hierarchy of ever increasing order of Green functions. The mass operator M is introduced to truncate the hierarchy:

$$i \int d3 \, v(1-3) \, G^{(2)}(1,2,3,3^+) = -\int d3 \, M(1,3) \, G(3,2) \,. \tag{68}$$

The self-energy  $\Sigma$  is defined to be the mass operator with the mean-field Hartree potential  $V_{\rm H}$  removed

$$\Sigma(1,2) = M(1,2) - V_{\rm H}(1)\,\delta(1-2)\,,\tag{69}$$

where

$$V_{\rm H}(1) = \int d3 \, v(1-3)\rho(3) \,. \tag{70}$$

The equation of motion of the Green function then becomes

$$\left(i\frac{\partial}{\partial t_1} - h(1)\right)G(1,2) - \int d3\,\Sigma(1,3)\,G(3,2) = \delta(1-2)\,,\tag{71}$$

where h includes the Hartree potential

$$h = h_0 + V_{\rm H}.$$
 (72)

From the equation of motion in Eq. (71) the inverse Green function is given by

$$G^{-1}(1,2) = \left(i\frac{\partial}{\partial t_1} - h(1)\right)\delta(1-2) - \Sigma(1,2).$$
(73)

This expression will be used to construct an iterative equation for the self-energy.

#### A.2 Self-energy

Instead of following the conventional diagrammatic approach of many-body perturbation theory (see, e.g., Fetter and Walecka [26]), the self-energy can be conveniently derived using the functional derivative method [11, 12]. By applying a time-dependent probing field  $\varphi(rt)$  the response of the Green function with respect to this probing field can be related to the self-energy. Once derivatives are taken, the field is set to zero. In the presence of an external field it is suitable to work in the interaction representation and define the Green function as

$$iG(1,2) = \frac{\langle \Psi_0 | T \ \hat{S} \hat{\psi}_{\mathrm{D}}(1) \ \hat{\psi}_{\mathrm{D}}^{\dagger}(2) | \Psi_0 \rangle}{\langle \Psi_0 | \hat{S} | \Psi_0 \rangle}$$
(74)

where  $\hat{\psi}_{\rm D}(1)$  is the field operator in the interaction (Dirac) picture

$$\hat{\psi}_{\mathrm{D}}(rt) = \mathrm{e}^{i\hat{H}t}\,\hat{\psi}(r)\,e^{-i\hat{H}t}.\tag{75}$$

It is to be noted that the above field operators are independent of the probing field  $\varphi(rt)$ . The operator  $\hat{S}$  is defined as

$$\hat{S} = \hat{U}_{\rm D}(\infty, -\infty),\tag{76}$$

where

$$\hat{U}_{\rm D}(t,t') = T \,{\rm e}^{-i\int_{t'}^t d\tau \hat{\phi}(\tau)}$$
(77)

is the time-evolution operator in the interaction picture, with

$$\hat{\phi}(t) = \int d^3 r \,\hat{\rho}_{\rm D}(rt) \,\varphi(rt) \,, \tag{78}$$

where  $\hat{\rho}_{\rm D}(rt)$  is the density operator. The definition of G in Eq. (74) reduces to the usual definition when  $\varphi = 0$ .

We will now take the functional derivative of G with respect to  $\varphi$ . In order to do this it is only necessary to work out the functional derivative of  $\hat{S}$  with respect to  $\varphi$  since the field operators in the Dirac picture do not depend on  $\varphi$ :

$$\frac{\delta\hat{S}}{\delta\varphi(3)} = \frac{\delta}{\delta\varphi(3)} T \,\mathrm{e}^{-i\int d4\,\hat{\rho}_{\mathrm{D}}(4)\,\varphi(4)} = -i\,T\,\hat{S}\,\hat{\rho}_{\mathrm{D}}(3)\,. \tag{79}$$

We obtain from Eq. (74)

$$i\frac{\delta G(1,2)}{\delta\varphi(3)} = -\frac{i\langle\Psi_{0}|T\,\hat{S}\,\hat{\rho}_{\rm D}(3)\,\hat{\psi}_{\rm D}(1)\,\hat{\psi}_{\rm D}^{\dagger}(2)|\Psi_{0}\rangle}{\langle\Psi_{0}|\hat{S}|\Psi_{0}\rangle} + \frac{i\langle\Psi_{0}|T\,\hat{S}\,\hat{\psi}_{\rm D}(1)\,\hat{\psi}_{\rm D}^{\dagger}(2)|\Psi_{0}\rangle\langle\Psi_{0}|T\,\hat{S}\,\hat{\rho}_{\rm D}(3)]|\Psi_{0}\rangle}{\langle\Psi_{0}|\hat{S}|\Psi_{0}\rangle^{2}}.$$
(80)

The two-particle Green function in the interaction picture is defined according to

$$G^{(2)}(1,2,3,4) = (-i)^2 \frac{\langle \Psi_0 | T \, \hat{S} \, \hat{\psi}_{\rm D}(1) \, \hat{\psi}_{\rm D}^{\dagger}(2) \, \hat{\psi}_{\rm D}(3) \, \hat{\psi}_{\rm D}^{\dagger}(4) | \Psi_0 \rangle}{\langle \Psi_0 | \hat{S} | \Psi_0 \rangle} \,, \tag{81}$$

and obtain

$$\frac{\delta G(1,2)}{\delta \varphi(3)} = iG(1,2)\rho(3) - G^{(2)}(1,2,3,3^+).$$
(82)

This relation is fundamental because it expresses the two-particle Green function, and hence the self-energy through Eqs. (68) and (69), as a linear response of the one-particle Green function with respect to the probing field.

A formal expression for the self-energy can now be written down by substituting  $G^{(2)}$  in Eq. (68) with the expression in Eq. (82)

$$\Sigma(1,2) = i \int d3 \, d4 \, v(1-3) \frac{\delta G(1,4)}{\delta \varphi(3)} G^{-1}(4,2)$$
  
=  $-i \int d3 \, d4 \, v(1-3) \, G(1,4) \frac{\delta G^{-1}(4,2)}{\delta \varphi(3)},$  (83)

where in the second line the following identity has been used

$$G G^{-1} = 1 \to \delta G G^{-1} + G \delta G^{-1} = 0 \to \delta G = -G \delta G^{-1} G.$$
(84)

From the equation of motion in (71) one finds, keeping in mind that the probing field  $\varphi$  is present in h,

$$\frac{\delta G^{-1}(4,2)}{\delta\varphi(3)} = -\left(\delta(4-3) + \frac{\delta V_{\rm H}(4)}{\delta\varphi(3)}\right)\delta(4-2) - \frac{\delta\Sigma(4,2)}{\delta\varphi(3)}.$$
(85)

This expression together with Eq. (83) yields an *iterative* equation for the self-energy, which facilitates an expansion of the self-energy in powers of the Coulomb interaction. Since

$$V_{\rm H}(4) = \int d5 \, v(4-5)\rho(5) \,, \tag{86}$$

we have

$$\frac{\delta V_{\rm H}(4)}{\delta \varphi(3)} = \int d5 \, v(4-5) \frac{\delta \rho(5)}{\delta \varphi(3)} = \int d5 \, v(4-5) \, R(5,3) \,, \tag{87}$$

where R is the time-ordered linear density response function

$$R(1,2) = \frac{\delta\rho(1)}{\delta\varphi(2)} = -i\frac{\delta G(1,1^+)}{\delta\varphi(2)}.$$
(88)

From Eq. (80) we find, with  $2 \to 1^+$  and after setting  $\varphi = 0 \to \hat{S} = 1$ , the time-ordered linear density response function is given by

$$iR(1,2) = \langle \Psi_0 | T \,\hat{\rho}(1) \,\hat{\rho}(2) | \Psi_0 \rangle - \rho(1) \,\rho(2) \,.$$
(89)

The spectral representation can be obtained by inserting a complete set of N-electron eigenstates of the Hamiltonian and performing a Fourier transformation yielding

$$R(r_1, r_2; \omega) = \sum_n \left( \frac{\langle \Psi_0 | \Delta \hat{\rho}(r_1) | \Psi_n \rangle \langle \Psi_n | \Delta \hat{\rho}(r_2) | \Psi_0 \rangle}{\omega - (E_n - E_0) + i\eta} - \frac{\langle \Psi_0 | \Delta \hat{\rho}(r_2) | \Psi_n \rangle \langle \Psi_n | \Delta \hat{\rho}(r_1) | \Psi_0 \rangle}{\omega + (E_n - E_0) - i\eta} \right),$$
(90)

where

$$\Delta \hat{\rho}(r) = \hat{\rho}(r) - \rho(r) \,.$$

For analytical treatment it is useful to write the density response function in terms of its spectral representation as

$$R(r_1, r_2; \omega) = \int_{-\infty}^0 d\omega' \frac{S(r_1, r_2; \omega')}{\omega - \omega' - i\eta} + \int_0^\infty d\omega' \frac{S(r_1, r_2; \omega')}{\omega - \omega' + i\eta},$$
(91)

where S is the spectral function given by

$$S(r_1, r_2; \omega) = \sum_n \left\langle \Psi_0 \middle| \Delta \hat{\rho}(r_1) \middle| \Psi_n \right\rangle \left\langle \Psi_n \middle| \Delta \hat{\rho}(r_2) \middle| \Psi_0 \right\rangle \delta(\omega - E_n + E_0) , \qquad (92)$$

defined to be anti-symmetric in frequency,

$$S(-\omega) = -S(\omega) \,.$$

The above spectral function is valid for systems with time-reversal symmetry (no magnetic field) for which

$$\left\langle \Psi_0 \middle| \Delta \hat{\rho}(r_1) \middle| \Psi_n \right\rangle \left\langle \Psi_n \middle| \Delta \hat{\rho}(r_2) \middle| \Psi_0 \right\rangle = \left\langle \Psi_0 \middle| \Delta \hat{\rho}(r_2) \middle| \Psi_n \right\rangle \left\langle \Psi_n \middle| \Delta \hat{\rho}(r_1) \middle| \Psi_0 \right\rangle.$$
(93)

The density response functions are central quantities in the calculation of the self-energy. Iteration of Eq. (83) together with (85) generates increasingly higher order density response functions, which determine the self-energy. The iteration starts by setting  $\delta \Sigma / \delta \varphi = 0$  yielding the well-known GW approximation.

#### A.3 Self-energy expansion in the screened interaction

It is possible to reformulate the iterative equation for the self-energy in Eqs. (83) and (85) in terms of the screened interaction W instead of the bare interaction v [11,12]. Instead of varying the bare probing field  $\varphi$ , one varies the total screened potential  $V_{\text{tot}}$  in Eq. (6).

The linear density response function is reformulated by introducing the polarization function P, defined as the linear response function with respect to the *total* potential  $V_{tot}$ 

$$P(1,2) = \frac{\delta\rho(1)}{\delta V_{\text{tot}}(2)}.$$
(94)

In terms of the polarization function, the induced density becomes

$$\rho_{\rm ind}(1) = \int d2 \ P(1,2) \ V_{\rm tot}(2) \ . \tag{95}$$

Equating Eqs. (4) and (95) and using Eq. (6) for  $V_{tot}$  and Eq. (5) for  $V_{ind}$  one finds

$$R(1,2) = P(1,2) + \int d3 \, d4 \, P(1,4) \, v(4-3) \, R(3,2) \,. \tag{96}$$

The response function R is related to the polarization function as

$$R(1,2) = \frac{\delta\rho(1)}{\delta\varphi(2)} = \int d3 \, \frac{\delta\rho(1)}{\delta V_{\text{tot}}(3)} \frac{\delta V_{\text{tot}}(3)}{\delta\varphi(2)} = \int d3 \, P(1,3) \, \varepsilon^{-1}(3,2) \tag{97}$$

and Eq. (56) for the screened interaction can be written as

$$W(1,2) = v(1-2) + \int d3 \, d4 \, v(1-3) \, P(3,4) \, W(4,2) \,. \tag{98}$$

Using the chain rule for functional derivatives, the iterative equation for the self-energy can now be written in terms of the screened interaction W

$$\frac{\delta G^{-1}(4,2)}{\delta \varphi(3)} = \int d5 \, \frac{\delta G^{-1}(4,2)}{\delta V_{\rm tot}(5)} \frac{\delta V_{\rm tot}(5)}{\delta \varphi(3)} = -\int d5 \left( \delta(5-4) \, \delta(4-2) + \frac{\delta \Sigma(4,2)}{\delta V_{\rm tot}(5)} \right) \varepsilon^{-1}(5,3), \tag{99}$$

where the second line has been obtained from the definition of  $\varepsilon^{-1}$  in Eq. (8) and from Eq. (73). Substituting the above expression for  $\delta G^{-1}/\delta \varphi$  into Eq. (83) yields

$$\Sigma(1,2) = iG(1,2) W(2,1) + i \int d4 \, d5 \, G(1,4) W(5,1) \, \frac{\delta \Sigma(4,2)}{\delta V_{\text{tot}}(5)} \,, \tag{100}$$

where W is given in Eq. (56). This iterative equation clearly displays the expansion of the selfenergy in powers of the screened interaction W as the equation is iterated. The first term is the well-known GW approximation.

### A.4 GW approximation

Fourier transformation of

$$\Sigma(1,2) = iG(1,2) W(2,1)$$
(101)

yields, using convolution theorem,

$$\Sigma(r_1, r_2; \omega) = i \int \frac{d\omega'}{2\pi} G(r_1, r_2; \omega + \omega') W(r_2, r_1; \omega') e^{i\eta\omega'}.$$
(102)

Using the spectral representation of G,

$$G(r_1, r_2; \omega) = \int_{-\infty}^{\mu} d\omega' \, \frac{A^{\rm h}(r_1, r_2; \omega')}{\omega - \omega' - i\delta} + \int_{\mu}^{\infty} d\omega' \, \frac{A^{\rm e}(r_1, r_2; \omega')}{\omega - \omega' + i\delta} \,, \tag{103}$$

one finds for the exchange part

$$\Sigma^{\mathbf{x}}(r_1, r_2) = iv(r_1 - r_2) \int \frac{d\omega'}{2\pi} \int_{-\infty}^{\mu} d\omega_1 \frac{A^{\mathbf{h}}(r_1, r_2; \omega_1) e^{i\eta\omega'}}{\omega + \omega' - \omega_1 - i\delta} = -v(r_1 - r_2) \int_{-\infty}^{\mu} d\omega_1 A^{\mathbf{h}}(r_1, r_2; \omega_1) d\omega_1 A^{\mathbf{h}}$$

The factor  $\exp(i\eta\omega')$  ensures that the contour is closed along a semicircle above the real axis so that only poles below the chemical potential corresponding to occupied states are picked up. For the correlation part we find, using the spectral representation of  $W^c$  in Eq. (14), dropping the space variables for clarity,

$$\Sigma^{c}(\omega) = \int_{-\infty}^{\mu} d\omega_{1} \int_{0}^{\infty} d\omega_{2} \frac{A^{h}(\omega_{1})B(\omega_{2})}{\omega + \omega_{2} - \omega_{1} - i\delta} - \int_{\mu}^{\infty} d\omega_{1} \int_{-\infty}^{0} d\omega_{2} \frac{A^{e}(\omega_{1})B(\omega_{2})}{\omega + \omega_{2} - \omega_{1} + i\delta}.$$
 (105)

The spectral representation of the correlation part of the self-energy has the same form as that of the Green function and can be written as

$$\Sigma^{c}(r_{1}, r_{2}; \omega) = \int_{-\infty}^{\mu} d\omega' \frac{\Gamma^{h}(r_{1}, r_{2}; \omega')}{\omega - \omega' - i\delta} + \int_{\mu}^{\infty} d\omega' \frac{\Gamma^{e}(r_{1}, r_{2}; \omega')}{\omega - \omega' + i\delta},$$
(106)

where

$$\Gamma^{\rm h}(\omega) = -\frac{1}{\pi} \operatorname{Im} \Sigma^{\rm c}(\omega) \Theta(\mu - \omega), \qquad (107)$$

$$\Gamma^{\rm e}(\omega) = -\frac{1}{\pi} \operatorname{Im} \Sigma^{\rm c}(\omega) \,\Theta(\omega - \mu) \,. \tag{108}$$

If we use a noninteracting Green function

$$G^{0}(r_{1}, r_{2}; \omega) = \sum_{n}^{\text{occ}} \frac{\phi_{n}(r_{1})\phi_{n}^{*}(r_{2})}{\omega - \varepsilon_{n} - i\delta} + \sum_{n}^{\text{unocc}} \frac{\phi_{n}(r_{1})\phi_{n}^{*}(r_{2})}{\omega - \varepsilon_{n} + i\delta},$$
(109)

corresponding to

$$A^{\rm h}(r_1, r_2; \omega) = \sum_n^{\rm occ} \phi_n(r_1) \, \phi_n^*(r_2) \, \delta(\omega - \varepsilon_n) \,, \tag{110}$$

$$A^{\mathrm{e}}(r_1, r_2; \omega) = \sum_{n}^{\mathrm{unocc}} \phi_n(r_1) \, \phi_n^*(r_2) \, \delta(\omega - \varepsilon_n) \,, \tag{111}$$

where the one-particle states  $\{\phi_n\}$  with eigenvalues  $\{\varepsilon_n\}$  are usually chosen to be the Kohn-Sham orbitals or Bloch states, then the exchange potential reduces to the familiar expression

$$\Sigma^{\mathbf{x}}(r_1, r_2) = -v(r_2 - r_1) \sum_{n=1}^{\infty} \phi_n(r_1) \phi_n^*(r_2).$$
(112)

The spectral functions of the correlation part of the self-energy become

$$\Gamma^{\rm h}(r_1, r_2; \omega \le \mu) = \sum_n^{\rm occ} \phi_n(r_1) \, \phi_n^*(r_2) \, B(r_2, r_1; \varepsilon_n - \omega) \, \Theta(\varepsilon_n - \omega) \,, \tag{113}$$

$$\Gamma^{\rm e}(r_1, r_2; \omega > \mu) = \sum_n^{\rm unocc} \phi_n(r_1) \, \phi_n^*(r_2) \, B(r_2, r_1; \omega - \varepsilon_n) \, \Theta(\omega - \varepsilon_n) \,. \tag{114}$$

The correlation part of the self-energy can then be obtained from the spectral representation in Eq. (106).

The above expression shows that collective charge excitations (plasmons) embodied in  $B(\omega)$ , proportional to Im W, are transferred to Im  $\Sigma$ . Through the Dyson equation these plasmon excitations are coupled to the Green function and appear as satellites in the spectral function. This explicitly describes a clear picture of the coupling between the electrons and the collective charge excitations in the system, which is illustrated in the Feynman diagram in Fig. 1. An added electron or hole represented by the Green function line interacts with the system and induces a collective charge excitation or a plasmon which is reabsorbed at a later time. This coupling to the plasmon excitation renormalizes the noninteracting Green function via the self-energy, resulting in a heavier effective mass and the transfer of quasiparticle weight to the satellite region.

## **B** Functional derivation of *GW*+EDMFT

The Hamiltonian of a many-electron system moving in some external potential  $v_{\mathrm{ext}}$  is given by

$$\hat{H} = \hat{H}_0 + \hat{V},\tag{115}$$

where

$$\hat{H}_0 = \int dr \,\hat{\psi}^{\dagger}(r) \,h_0(r) \,\hat{\psi}(r) \,, \tag{116}$$

$$h_0(r) = -\frac{1}{2} \bigtriangledown^2 + v_{\text{ext}}(r),$$
 (117)

$$\hat{V} = \frac{1}{2} \int dr dr' \,\hat{\psi}^{\dagger}(r) \,\hat{\psi}^{\dagger}(r') \,v(r-r') \,\hat{\psi}(r') \,\hat{\psi}(r) \,.$$
(118)

r labels position and spin variables:  $r = (\mathbf{r}, \sigma)$  and  $v(r-r') = 1/|\mathbf{r}-\mathbf{r}'|$ . The interaction term of the Hamiltonian can be rewritten as

$$\hat{V} = \frac{1}{2} \int dr dr' \,\hat{\rho}(r) \,v(r-r') \big(\hat{\rho}(r') - \delta(r-r')\big),\tag{119}$$

where the second term is the unphysical self-interaction term.  $\hat{V}$  can be expressed in terms of the density fluctuation operator,

$$\Delta \hat{\rho}(r) = \hat{\rho}(r) - \rho(r), \qquad (120)$$

as

$$\hat{V} = \Delta \hat{V} + \int dr \, V_{\rm H}(r) \hat{\rho}(r) - E_{\rm H} - \frac{1}{2} \int dr dr' \, \delta(r - r') \, v(r - r') \, \hat{\rho}(r), \tag{121}$$

where

$$\Delta \hat{V} = \frac{1}{2} \int dr dr' \,\Delta \hat{\rho}(r) \,v(r-r') \,\Delta \hat{\rho}(r'), \tag{122}$$

$$V_{\rm H}(r) = \int dr' \, v(r - r') \, \rho(r'), \tag{123}$$

and

$$E_{\rm H} = \frac{1}{2} \int dr dr' \,\rho(r) \,v(r-r') \,\rho(r'). \tag{124}$$

The Hartree potential  $V_{\rm H}$  can be incorporated into the one-particle part of the Hamiltonian  $\hat{H}_0$ and the Hartree energy  $E_{\rm H}$  can be absorbed into the chemical potential. The self-interaction term can also be included in  $\hat{H}_0$  and it will eventually cancel out of physical observables. In the functional integral formalism, one evaluates at each time slice  $\epsilon = \beta/M$  [27]

$$e^{-\epsilon(\hat{H}-\mu N)} = e^{-\epsilon(\hat{H}_0-\mu\hat{N}+\hat{V})} \sim e^{-\epsilon(\hat{H}'_0-\mu\hat{N})} e^{-\epsilon\Delta\hat{V}},$$
(125)

where  $\hat{H}'_0$  includes the one-particle residue  $\hat{V} - \Delta \hat{V}$ . Using the Hubbard-Stratonovich transformation, which is essentially a Gaussian integral formula for operators, one has

$$\int \prod_{i} \left( \frac{d\theta_{i}}{\sqrt{2\pi}} \right) e^{-\frac{1}{2}\theta A\theta + \theta \hat{J}} = \frac{1}{\sqrt{\det A}} e^{\frac{1}{2}\hat{J}A^{-1}\hat{J}}.$$
(126)

Applying this operator identity one obtains

$$e^{-\epsilon\Delta\hat{V}} = e^{-\epsilon\frac{1}{2}\int dr dr'\Delta\hat{\rho}(r)v(r-r')\Delta\hat{\rho}(r')}.$$
(127)

Associating  $\hat{J} \to i \Delta \hat{\rho}$ ,  $A^{-1} \to \epsilon v$  and  $\theta \to \epsilon \phi$ , one finds

$$e^{-\epsilon\Delta\hat{V}} = \left[\det(\epsilon v^{-1})\right]^{1/2} \int \frac{d\phi}{\sqrt{2\pi}} e^{-\epsilon\frac{1}{2}\int dr dr'\phi(r)v^{-1}(r-r')\phi(r') + i\epsilon\int dr\phi(r)\Delta\hat{\rho}(r)}, \qquad (128)$$

where  $\phi$  is an auxiliary bosonic field which is real (not a Grassmann variable). The partition function is given by

$$Z = \int \mathcal{D}[\psi^* \psi] e^{-S(\psi^*, \psi)}, \qquad (129)$$

where a correspondence between the annihilation and creation field operators and the Grassmann variables has been made as follows:

$$\hat{\psi}(x) \to \psi(x), \ \hat{\psi}^{\dagger}(x) \to \psi^*(x).$$
 (130)

The label x represents position, spin and imaginary time variables:  $x = (r, \tau) = (\mathbf{r}, \sigma, \tau)$ . The Coulomb interaction is written as

$$v(x-x') = v(r-r')\,\delta(\tau-\tau').$$
(131)

The action is given by

$$S(\psi^*, \psi) = \int dx \, \psi^*(x) \left(\frac{\partial}{\partial \tau} - \mu + h_0(x)\right) \psi(x) + \frac{1}{2} \int dx dx' \, \psi^*(x) \, \psi^*(x') \, v(x-x') \, \psi(x') \, \psi(x).$$
(132)

Using the Hubbard-Stratonovich transformation described above the action can be naturally decomposed as a sum of three terms

$$S(\psi^*, \psi, \phi) = S_{\rm F}(\psi^*, \psi) + S_{\rm B}(\phi) + S_{\rm BF}(\psi^*, \psi, \phi),$$
(133)

where

$$S_{\rm F}(\psi^*,\psi) = \int dx \,\psi^*(x) \left(\frac{\partial}{\partial \tau} - \mu + h_0(x) + V_H(x)\right) \psi(x),\tag{134}$$

$$S_{\rm B}(\phi) = \frac{1}{2} \int dx dx' \,\phi(x) \, v^{-1}(x - x') \,\phi(x'), \tag{135}$$

$$S_{\rm BF}(\psi^*, \psi, \phi) = -i \int dx \,\phi(x) \big(\psi^*(x)\psi(x) - \rho(x)\big).$$
(136)

 $S_{\rm F}$  and  $S_{\rm B}$  correspond, respectively, to the fermion and boson mean-field actions.  $S_{\rm BF}$  is the interaction term between the bosonic and fermionic fields. Implicit in this last term is the presence of the Coulomb interaction, which can be brought out by making a transformation  $\phi = v \tilde{\phi}$ , yielding a more intuitive expression

$$S_{\rm BF}(\psi^*,\psi,\widetilde{\phi}) = \int dx dx' \,\widetilde{\phi}(x') \, v(x'-x) \big(\psi^*(x)\psi(x) - \rho(x)\big). \tag{137}$$

The partition function after the Hubbard-Stratonovich transformation becomes

$$Z = \int \mathcal{D}[\psi^*, \psi, \phi] e^{-S_{\rm F}(\psi^*, \psi) - S_{\rm B}(\phi)} e^{-S_{\rm BF}(\psi^*, \psi, \phi)}.$$
 (138)

Using the coupling constant integration technique, the contribution to the thermodynamic potential  $\Omega = -\ln Z$  from the  $S_{\rm BF}$  coupling term can be calculated

$$Z_{\alpha} = \int \mathcal{D}[\psi^*, \psi, \phi] e^{-S_{\mathrm{F}}(\psi^*, \psi) - S_{\mathrm{B}}(\phi)} e^{-\alpha S_{\mathrm{BF}}(\psi^*, \psi, \phi)}.$$
(139)

$$\frac{\partial \Omega_{\alpha}}{\partial \alpha} = -\frac{\partial \ln Z_{\alpha}}{\partial \alpha} = -\frac{1}{Z_{\alpha}} \frac{\partial Z_{\alpha}}{\partial \alpha} = \left\langle S_{\rm BF}(\psi^*, \psi, \phi) \right\rangle_{\alpha},\tag{140}$$

$$\Omega - \Omega_0 = \int_0^1 d\alpha \frac{\partial \Omega_\alpha}{\partial \alpha} = \int_0^1 d\alpha \left\langle S_{\rm BF}(\psi^*, \psi, \phi) \right\rangle_\alpha.$$
(141)

Introducing source potentials  $J_{\rm F}$  and  $J_{\rm B}$  which couple, respectively, to the fermion and boson fields one finds

$$Z[J_{\rm F}, J_{\rm B}] = \int \mathcal{D}[\psi^*, \psi, \phi] \, \mathrm{e}^{-S_{\rm F}(\psi^*, \psi) - S_{\rm B}(\phi) - \psi^* J_{\rm F} \psi + \frac{1}{2} \phi J_{\rm B} \phi} \, \mathrm{e}^{-S_{\rm BF}(\psi^*, \psi, \phi)}.$$
(142)



**Fig. 7:** The GW+EDMFT approximation to the  $\Psi$ -functional. The first diagram corresponds to GWA and the second diagram the double counting. Lower-case indexes are restricted to the same unit cell, and thus correspond to diagrams accounted for in the impurity problem. The figure is taken from Boehnke et al. [20].

Consider first the case when  $S_{\rm BF} = 0$ , i.e., no coupling between the bosonic and fermionic fields.  $S_{\rm F}$  can be associated with a thermodynamic potential  $\Omega_{\rm F}$  and its Legendre transform

$$\Gamma_F[G] = \Omega_F[J_F] - J_F G = \text{Tr } \ln G - \text{Tr } (G_0^{-1} G - 1),$$
(143)

where

$$G_0^{-1}(x,x') = -\left(\frac{\partial}{\partial\tau} - \mu + v_{\text{ext}}(x) + V_{\text{H}}(x)\right)\delta(x-x').$$
(144)

For the bosonic part,

$$Z_{\rm B}[J_{\rm B}] = \int \mathcal{D}[\phi] \, \mathrm{e}^{-\frac{1}{2}\phi(v^{-1} - J_{\rm B})\phi} = \left(\det(v^{-1} - J_{\rm B})\right)^{-1/2},\tag{145}$$

where the Gaussian integral formula for real variables has been used. Defining the boson propagator

$$W(x,y) = \left\langle \phi(x)\phi(y) \right\rangle = \frac{2}{Z_{\rm B}} \frac{\delta Z_{\rm B}}{\delta J_{\rm B}(x,y)} = 2 \frac{\delta \ln Z_{\rm B}}{\delta J_{\rm B}(x,y)}.$$
(146)

and using the identity

$$\ln \det M = \operatorname{Tr} \, \ln M, \tag{147}$$

one finds

$$\ln Z_{\rm B}[J_{\rm B}] = -\frac{1}{2} \ln \det \left( v^{-1} - J_{\rm B} \right) = -\frac{1}{2} \operatorname{Tr} \ln \left( v^{-1} - J_{\rm B} \right), \tag{148}$$

$$W[J_{\rm B}] = 2 \frac{\delta \ln Z_{\rm B}[JB]}{\delta J_{\rm B}} = \left(v^{-1} - J_{\rm B}\right)^{-1} \to J_{\rm B}[W] = v^{-1} - W^{-1}, \tag{149}$$

$$\Omega_{\rm B}[J_{\rm B}] = -\ln Z_{\rm B}[J_{\rm B}] = \frac{1}{2} \mathrm{Tr} \, \ln W^{-1} = -\frac{1}{2} \mathrm{Tr} \, \ln W.$$
(150)

One then obtains

$$\Gamma_{\rm B}[W] = \Omega_{\rm B}[J_{\rm B}] + \frac{1}{2} \text{Tr} (J_{\rm B}W) = -\frac{1}{2} \text{Tr} \ln W + \frac{1}{2} \text{Tr} (v^{-1}W - 1), \qquad (151)$$

which shows that  $\Gamma_{\rm B}$ , the Legendre transform of  $\Omega_{\rm B}$ , is indeed a functional of W.

Taking into account the coupling term  $S_{\rm BF}$  yields

$$\Gamma[G,W] = \Gamma_{\rm F}[G] + \Gamma_{\rm B}[W] + \Psi[G,W], \qquad (152)$$

where from Eq. (141)

$$\Psi[G,W] = \int_0^1 d\alpha \left\langle S_{\rm BF}(\psi^*,\psi,\phi) \right\rangle_\alpha.$$
(153)

Functional derivative with respect to G yields the Dyson equation for G

$$\frac{\delta\Gamma[G,W]}{\delta G} = \frac{\delta\Gamma_{\rm F}[G]}{\delta G} + \frac{\delta\Psi[G,W]}{\delta G}$$
(154)

$$-J_{\rm F} = G^{-1} - G_0^{-1} + \frac{\delta \Psi[G, W]}{\delta G},$$
(155)

with the fermion self-energy

$$\Sigma[G, W] = \frac{\delta \Psi[G, W]}{\delta G} + J_{\rm F}.$$
(156)

Similarly, functional derivative with respect to the boson propagator W yields

$$\frac{\delta\Gamma[G,W]}{\delta W} = \frac{\delta\Gamma_{\rm B}[W]}{\delta W} + \frac{\delta\Psi[G,W]}{\delta W}$$
(157)

$$\frac{1}{2}J_{\rm B} = -\frac{1}{2}W^{-1} + \frac{1}{2}v^{-1} + \frac{\delta\Psi[G,W]}{\delta W},$$
(158)

with the boson self-energy or polarization

$$P[G,W] = -2\frac{\delta\Psi[G,W]}{\delta W} + J_{\rm B}.$$
(159)

For  $J_{\rm F} = J_{\rm B} = 0$ , which correspond to the extrema of  $\Gamma[G, W]$  we find the Dyson equations for G and W

$$\frac{\delta\Gamma[G,W]}{\delta G} = 0 \to G = G_0 + G_0 \Sigma G, \tag{160}$$

$$\frac{\delta\Gamma[G,W]}{\delta W} = 0 \to W = v + v P W.$$
(161)

The EDMFT approximation corresponds to an onsite approximation to G and W

$$\Psi^{\text{EDMFT}} = \Psi[G_{\text{RR}}, W_{\text{RRRR}}].$$
(162)

GWA, on the other hand, corresponds to the first-order diagram in  $\Psi$ 

$$\Psi^{GW} = -\frac{1}{2} \operatorname{Tr} GWG \,. \tag{163}$$

This yields the total GW+EDMFT functional in Fig. 7

$$\Psi^{GW+\text{EDMFT}} = \Psi^{GW} + \Psi^{\text{EDMFT}} + \frac{1}{2} \text{Tr}(G_{\mathbf{RR}} W_{\mathbf{RRRR}} G_{\mathbf{RR}}),$$
(164)

where the last term is the double-counting term.

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