

# First-Principles Approach to the Electronic Structure of Strongly Correlated Systems: Combining the *GW* Approximation and Dynamical Mean-Field Theory

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We propose a dynamical mean-field approach for calculating the electronic structure of strongly correlated materials from first principles. The scheme combines the *GW* method with dynamical mean-field theory, which enables one to treat strong interaction effects. It avoids the conceptual problems inherent to conventional “LDA + DMFT,” such as Hubbard interaction *parameters* and double-counting terms. We apply a simplified version of the approach to the electronic structure of nickel and find encouraging results.

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For systems with weak or moderate Coulomb correlations such as *sp* metals and semiconductors the *GW* method [1–3] is the tool of choice for the determination of excited states properties from first principles. In this method the effective screened interaction is treated at the RPA level and used to construct an approximation to the electronic self-energy. Applications to more strongly correlated systems with localized orbitals, however, indicate a need to go beyond the *GW* approximation (GWA). For example, in ferromagnetic nickel, the GWA [4] is successful at predicting the quasiparticle band narrowing, but neither improves the (too large) exchange splitting found in density-functional theory (DFT) within the local-density approximation (LDA) nor reproduces the 6 eV photoemission satellite [5].

Recently, a new approach to the electronic structure of strongly correlated materials has been developed. This approach, dubbed “LDA + DMFT,” combines the dynamical mean-field theory (DMFT) [6] of correlated electron models with DFT-LDA calculations [7]. It is also a Green’s function technique, but—unlike GWA—it does not treat the Coulomb interaction from first principles. Instead, an effective Hamiltonian involving Hubbard-like interaction *parameters* is used, as well as a “double-counting” correction term. The strength of DMFT is that the on-site electronic interactions are treated to all orders, by using a mapping onto a self-consistent quantum impurity problem. DMFT has led to remarkable success with materials in which the Mott phenomenon or the formation of local moments plays a key role, e.g., for the satellite structure in Ni [8].

The aim of this Letter is to propose a first-principles scheme for strongly correlated materials in which the *GW* treatment of the screened Coulomb interaction and self-energy is combined with an extended DMFT [9–12] calculation in a self-consistent manner [13]. This combined *GW* + (E)DMFT scheme does not make use of Hubbard interaction parameters and bypasses the need

for a double-counting correction when implemented in a self-consistent dynamical manner (in fact, using LDA is in principle no longer necessary then). In this work however, we implement a simplified version of this scheme for ferromagnetic nickel, which serves as a test for the feasibility of realistic calculations using this approach.

We consider the Hamiltonian for electrons in a solid interacting via the Coulomb potential  $V(\mathbf{r} - \mathbf{r}') = e^2/|\mathbf{r} - \mathbf{r}'|$ . The general strategy of our approach is to construct a functional of the one-electron Green’s function  $G(\mathbf{r}, \mathbf{r}'; \tau - \tau') \equiv -\langle T_\tau \psi(\mathbf{r}, \tau) \psi^\dagger(\mathbf{r}', \tau') \rangle$  and the screened Coulomb interaction  $W$  [14,15]. The screened Coulomb interaction is defined using the (connected) density-density response function:  $\chi(\mathbf{r}, \mathbf{r}'; \tau - \tau') \equiv \langle T_\tau [\hat{\rho}(\mathbf{r}, \tau) - n(\mathbf{r})][\hat{\rho}(\mathbf{r}', \tau') - n(\mathbf{r}')] \rangle$  as  $W = V - V \cdot \chi \cdot V$  where the dots denote spatial convolutions. Following [14,15] we introduce the free-energy functional

$$\Gamma[G, W] = \text{Tr} \ln G - \text{Tr}[(G_H^{-1} - G^{-1})G] - \frac{1}{2} \text{Tr} \ln W + \frac{1}{2} \text{Tr}[(V^{-1} - W^{-1})W] + \Psi[G, W]. \quad (1)$$

In this expression  $G_H^{-1} = i\omega_n + \mu + \nabla^2/2 - v_c - v_H$  is the bare Green’s function of the solid including the Hartree potential  $v_H(\mathbf{r}) \equiv \int d\mathbf{r}' V(\mathbf{r} - \mathbf{r}')n(\mathbf{r}')$ .  $\Psi$  contains all contributions beyond Hartree. It is the sum of skeleton diagrams, irreducible with respect to both one-electron and interaction lines. A formal construction of this functional can be given (following [15]) by making a Hubbard-Stratonovich transformation and a Legendre transformation with respect to both  $G$  and  $W$ .  $\Psi$  can be expressed as  $\Psi[G, W] = i \int_0^1 d\alpha \int d\mathbf{r} d\tau \langle \phi(\mathbf{r}, \tau) \times [\psi^\dagger(\mathbf{r}, \tau)\psi(\mathbf{r}, \tau) - n(\mathbf{r})] \rangle$ , where  $\phi(\mathbf{r}, \tau)$  is an auxiliary bosonic field conjugate to the density fluctuation  $[\psi^\dagger(\mathbf{r}, \tau)\psi(\mathbf{r}, \tau) - n(\mathbf{r})]$ . Stationarity of  $\Gamma$  (corresponding

to equilibrium), yields the exchange and correlation self-energy  $\Sigma^{xc}$  and the polarization operator  $P$ :

$$\begin{aligned} \frac{\delta\Gamma}{\delta G} = 0 &\Rightarrow G^{-1} = G_H^{-1} - \Sigma^{xc}, & \Sigma^{xc} &= \frac{\delta\Psi}{\delta G}, \\ \frac{\delta\Gamma}{\delta W} = 0 &\Rightarrow W^{-1} = V^{-1} - P, & P &= -2 \frac{\delta\Psi}{\delta W}. \end{aligned} \quad (2)$$

The (self-consistent)  $GW$  approximation retains only the first order contribution to  $\Psi$  in the  $\alpha$  expansion, i.e., the exchange diagram  $\Psi_{GW} = -\frac{1}{2}\text{Tr}GWG$  [14], leading to  $\Sigma_{GW}^{xc} = -GW$  and  $P_{GW} = GG$ .

In order to proceed further, we need to specify a basis set. One-particle quantities like  $G$  or  $\Sigma$  are represented as  $G(\mathbf{r}, \mathbf{r}', i\omega_n) = \sum_{LL'\mathbf{R}\mathbf{R}'} \phi_L^{\mathbf{R}}(\mathbf{r}) G_{LL'}^{\mathbf{R}\mathbf{R}'}(i\omega_n) \phi_{L'}^{\mathbf{R}'}(\mathbf{r}')^* = \sum_{LL'\mathbf{k}} \phi_L^{\mathbf{k}}(\mathbf{r}) G_{LL'}(\mathbf{k}, i\omega_n) \phi_{L'}^{\mathbf{k}}(\mathbf{r}')^*$ , where  $\phi$  are localized basis functions [e.g., linearized muffin tin orbitals (LMTOs)] [16], centered at an atomic position  $R$  (and for simplicity assumed to be orthogonal). Two-particle quantities such as  $P$  or  $W$  are represented as  $W(\mathbf{r}, \mathbf{r}', i\nu_n) = \sum_{\alpha\beta\mathbf{R}\mathbf{R}'} B_\alpha^{\mathbf{R}}(\mathbf{r}) W_{\alpha\beta}^{\mathbf{R}\mathbf{R}'}(i\nu_n) B_\beta^{\mathbf{R}'}(\mathbf{r}')^*$ . Here  $B$ 's are linear combinations of  $\phi\phi$  and form an orthonormal set [2,17]. Note that the set  $\phi\phi$  is in general over-complete so that the number of  $B$ 's is smaller or equal to the number of  $\phi\phi$ . Matrix elements in products of LMTOs are then given by

$$W_{L_1L_2L_3L_4}^{\mathbf{R}\mathbf{R}'} \equiv \langle \phi_{L_1}^{\mathbf{R}} \phi_{L_2}^{\mathbf{R}} | W | \phi_{L_3}^{\mathbf{R}'} \phi_{L_4}^{\mathbf{R}'} \rangle = \sum_{\alpha\beta} O_{L_1L_2}^\alpha W_{\alpha\beta}^{\mathbf{R}\mathbf{R}'} O_{L_3L_4}^{\beta*} \quad (3)$$

$$S = \int d\tau d\tau' \left[ - \sum c_L^\dagger(\tau) \mathcal{G}_{LL'}^{-1}(\tau - \tau') c_{L'}(\tau') + \frac{1}{2} \sum : c_{L_1}^\dagger(\tau) c_{L_2}(\tau) : \mathcal{U}_{L_1L_2L_3L_4}(\tau - \tau') : c_{L_3}^\dagger(\tau') c_{L_4}(\tau') : \right], \quad (6)$$

where the sums run over all orbital indices  $L$ ,  $c_L^\dagger$  is associated with orbital  $L$ , and the double dots denote normal ordering (taking care of Hartree terms). In analogy to the Kohn-Sham representation of the local density, this can be viewed as a *representability* assumption, namely, that the local components of  $G$  and  $W$  can be obtained from (6) with suitably chosen values of the auxiliary (Weiss) functions  $\mathcal{G}$  and  $\mathcal{U}$ . This construction defines the (frequency-dependent) interactions  $\mathcal{U}_{L_1L_2L_3L_4}(\omega)$ , for a specific material, in a unique manner (for a given basis set). Note that  $\mathcal{U}_{L_1L_2L_3L_4}$  must correspond to an interaction matrix  $\mathcal{U}_{\alpha\beta}$  in the  $B^\alpha$  basis via a transformation identical to (3). Taking derivatives of (4) as in (2) it is seen that the complete self-energy and polarization operators read

$$\begin{aligned} \Sigma^{xc}(\mathbf{k}, i\omega_n)_{LL'} &= \Sigma_{GW}^{xc}(\mathbf{k}, i\omega_n)_{LL'} - \sum_{\mathbf{k}} \Sigma_{GW}^{xc}(\mathbf{k}, i\omega_n)_{LL'} \\ &+ [\Sigma_{\text{imp}}^{xc}(i\omega_n)]_{LL'}, \end{aligned} \quad (7)$$

$$\begin{aligned} P(\mathbf{q}, i\nu_n)_{\alpha\beta} &= P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} - \sum_{\mathbf{q}} P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} \\ &+ P^{\text{imp}}(i\nu_n)_{\alpha\beta}. \end{aligned} \quad (8)$$

with the overlap matrix  $O_{L_1L_2}^\alpha \equiv \langle \phi_{L_1} \phi_{L_2} | B^\alpha \rangle$ . We note that in general we cannot obtain  $W_{\alpha\beta}$  from  $W_{L_1L_2L_3L_4}$ , while the converse is true.

The functionals  $\Gamma[G, W]$  and  $\Psi[G, W]$  can thus be viewed as functionals of the matrix elements  $G_{L_1L_2}^{\mathbf{R}\mathbf{R}'}(i\omega_n)$  and  $W_{\alpha\beta}^{\mathbf{R}\mathbf{R}'}(i\omega_n)$ . The main idea behind the present work is that the dependence of the  $\Psi$  functional upon the *off-site* components ( $\mathbf{R} \neq \mathbf{R}'$ ) of  $G^{\mathbf{R}\mathbf{R}'}$  and  $W^{\mathbf{R}\mathbf{R}'}$  can be treated within the  $GW$  approximation, while the dependence on the *on-site* components ( $\mathbf{R} = \mathbf{R}'$ ) requires a more accurate treatment for strongly correlated systems. We thus approximate the functional  $\Psi$  as

$$\Psi = \Psi_{GW}^{\text{nonloc}}[G^{\mathbf{R}\mathbf{R}'}, W^{\mathbf{R}\mathbf{R}'}] + \Psi_{\text{imp}}[G^{\mathbf{R}\mathbf{R}'}, W^{\mathbf{R}\mathbf{R}'}]. \quad (4)$$

In this expression, the first term corresponds to the  $GW$  functional restricted to off-site components of  $G$  and  $W$  (i.e., associated with distinct spheres  $\mathbf{R} \neq \mathbf{R}'$ ), namely,

$$\begin{aligned} \Psi_{GW}^{\text{nonloc}} &= -\frac{1}{2} \int d\tau \sum_{L_1 \dots L_2} \sum_{\mathbf{R} \neq \mathbf{R}'} G_{L_1L_1'}^{\mathbf{R}\mathbf{R}'}(\tau) W_{L_1L_2L_1'L_2'}^{\mathbf{R}\mathbf{R}'}(\tau) \\ &\times G_{L_2L_2'}^{\mathbf{R}\mathbf{R}'}(-\tau) \end{aligned} \quad (5)$$

with  $W_{L_1L_2L_1'L_2'}^{\mathbf{R}\mathbf{R}'}$  given by (3). All the dependence on the on-site components is gathered into  $\Psi_{\text{imp}}$ . Following (extended) DMFT, this on-site part of the functional is generated [18] from a local *quantum impurity problem* (defined on a single atomic site), with effective action:

Hence, the off-site part of the self-energy is taken from the GWA, while the on-site part is calculated to all orders from the dynamical impurity model. This treatment thus goes beyond EDMFT, where the lattice self-energy and polarization are just taken to be their impurity counterparts. The second term in (7) subtracts the on-site component of the  $GW$  self-energy thus avoiding double counting. As explained below, at self-consistency this term can be rewritten as

$$\sum_{\mathbf{k}} \Sigma_{GW}^{xc}(\tau)_{LL'} = - \sum_{L_1L_1'} W_{LL_1L_1'L_1}^{\text{imp}}(\tau) G_{L_1L_1'}(\tau), \quad (9)$$

so that it precisely subtracts the contribution of the  $GW$  diagram to the impurity self-energy. Similar considerations apply to the polarization.

We now outline the iterative loop which determines  $\mathcal{G}$  and  $\mathcal{U}$  self-consistently (and also the full self-energy and polarization):

(i) The impurity problem (6) is solved, for a given choice of  $\mathcal{G}_{LL'}$  and  $\mathcal{U}_{\alpha\beta}$ : the ‘‘impurity’’ Green’s function  $G_{\text{imp}}^{LL'} \equiv -\langle T_\tau c_L(\tau) c_{L'}^\dagger(\tau') \rangle_S$  is calculated, together with

the impurity self-energy  $\Sigma_{\text{imp}}^{xc} \equiv \delta\Psi_{\text{imp}}/\delta G_{\text{imp}} = \mathcal{G}^{-1} - G_{\text{imp}}^{-1}$ . The two-particle correlation function  $\chi_{L_1 L_2 L_3 L_4} = \langle : c_{L_1}^\dagger(\tau) c_{L_2}(\tau) :: c_{L_3}^\dagger(\tau') c_{L_4}(\tau') : \rangle_S$  must also be evaluated.

(ii) The impurity effective interaction is constructed:

$$W_{\text{imp}}^{\alpha\beta} = \mathcal{U}_{\alpha\beta} - \sum_{L_1 \dots L_4} \sum_{\gamma\delta} \mathcal{U}_{\alpha\gamma} O_{L_1 L_2}^\gamma \chi_{L_1 L_2 L_3 L_4} [O_{L_3 L_4}^\delta]^* \mathcal{U}_{\delta\beta}. \quad (10)$$

Here all quantities are evaluated at the same frequency [19]. The polarization operator of the impurity problem is then obtained as  $P_{\text{imp}} \equiv -2\delta\Psi_{\text{imp}}/\delta W_{\text{imp}} = \mathcal{U}^{-1} - W_{\text{imp}}^{-1}$ , where the matrix inversions are performed in the  $B^\alpha$  basis.

(iii) From (7) and (8) the full  $\mathbf{k}$ -dependent Green's function  $G(\mathbf{k}, i\omega_n)$  and effective interaction  $W(\mathbf{q}, i\nu_n)$  can be constructed. The self-consistency condition is obtained by requiring that the on-site components of these quantities coincide with  $G_{\text{imp}}$  and  $W_{\text{imp}}$ . This is done by computing the on-site quantities

$$G_{\text{loc}}(i\omega_n) = \sum_{\mathbf{k}} [G_H^{-1}(\mathbf{k}, i\omega_n) - \Sigma^{xc}(\mathbf{k}, i\omega_n)]^{-1}, \quad (11)$$

$$W_{\text{loc}}(i\nu_n) = \sum_{\mathbf{q}} [V^{-1}(\mathbf{q}) - P(\mathbf{q}, i\nu_n)]^{-1}, \quad (12)$$

and using them to update the Weiss field  $\mathcal{G}$  and the impurity model interaction  $\mathcal{U}$  according to  $\mathcal{G}^{-1} = G_{\text{loc}}^{-1} + \Sigma_{\text{imp}}$  and  $\mathcal{U}^{-1} = W_{\text{loc}}^{-1} + P_{\text{imp}}$ . This cycle is iterated until self-consistency for  $\mathcal{G}$  and  $\mathcal{U}$  is obtained (as well as for  $G$ ,  $W$ ,  $\Sigma^{xc}$ , and  $P$ ). Eventually, self-consistency over the local electronic density can also be implemented (in a similar way as in LDA + DMFT [20,21]) by recalculating  $\rho(\mathbf{r})$  from the Green's function at the end of the convergence cycle above. This new density is used as an input of a new  $GW$  calculation, and convergence over this external loop must be reached. While implementing self-consistency within the GWA is known to worsen spectra [22], we expect a more favorable situation in the proposed  $GW$  + DMFT scheme since part of the interaction effects are treated to all orders.

The implementation of the proposed approach in a dynamical and self-consistent manner is a major challenge. Here, we only demonstrate its potential within a simplified implementation, illustrated on nickel. The main simplifications made are (i) the DMFT local treatment is applied only to the  $d$  orbitals; (ii) the  $GW$  calculation is done only once, in the form [2]:  $\Sigma_{GW}^{xc} = -G_{\text{LDA}} \cdot W[G_{\text{LDA}}]$ , from which the nonlocal part of the self-energy is obtained; (iii) we replace the dynamical impurity problem by its static limit, solving the impurity model (6) for a frequency-independent  $\mathcal{U} = \mathcal{U}(\omega = 0)$ . Instead of the Hartree Hamiltonian we start from a one-electron Hamiltonian in the form:  $H_{\text{Hartree}} + V_{xc,\sigma}^{\text{local}} - \frac{1}{2} \text{Tr} \Sigma_{\sigma}^{\text{imp}}(0)$  where  $V_{xc,\sigma}^{\text{local}}$  is the  $\mathbf{R} = \mathbf{R}'$  component of

$V_{xc}$  in the LMTO basis. The nonlocal part of this Hamiltonian coincides with that of the Hartree Hamiltonian while its local part is derived from LDA, with a double-counting correction of the form proposed in [8] in the DMFT context. With this choice the self-consistency condition (11) reads

$$G_{\text{loc}}^\sigma(i\omega_n) = \sum_{\mathbf{k}} \left[ G_H^{-1}(\mathbf{k}, i\omega_n) - (\Sigma_{GW}^{xc})_{\text{nonloc}} - \left( \Sigma_{\text{imp},\sigma} + V_{xc}^{\text{loc}} - \frac{1}{2} \text{Tr}_{\sigma} \Sigma_{\text{imp},\sigma}(0) \right) \right]^{-1}. \quad (13)$$

We have performed finite temperature  $GW$  and LDA + DMFT calculations (within the LMTO atomic sphere approximation [16] with 29 irreducible  $\mathbf{k}$  points) for ferromagnetic nickel (lattice constant 6.654 a.u.), using  $4s4p3d4f$  states, at the Matsubara frequencies  $i\omega_n$  corresponding to  $T = 630$  K, just below the Curie temperature. The resulting self-energies are inserted into Eq. (13), which is then used to calculate a new Weiss field according to  $\mathcal{G}^{-1} = G_{\text{loc}}^{-1} + \Sigma_{\text{imp}}$ . The Green's function  $G_{\text{loc}}^\sigma(\tau)$  is recalculated from the impurity effective action by Quantum Monte Carlo (QMC) and analytically continued using the maximum entropy algorithm. Comparison of the resulting spectral function (Fig. 1) with the LDA + DMFT results in [8] shows that the good description of the satellite structure, exchange splitting, and band narrowing is indeed retained within the (simplified)  $GW$  + DMFT scheme. We have also calculated the quasiparticle band structure, from the poles of (13), after linearization of  $\Sigma(\mathbf{k}, i\omega_n)$  around the Fermi level [23]. Figure 2 shows a comparison of  $GW$  + DMFT with the LDA and experimental band structure.  $GW$  + DMFT correctly yields the bandwidth reduction compared to the (too large) LDA value and renormalizes the bands in a ( $\mathbf{k}$ -dependent) manner.

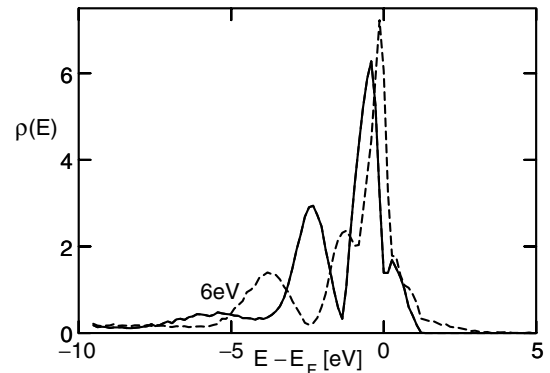


FIG. 1. Partial density of states of  $d$  orbitals of nickel (solid [dashed] lines give the majority [minority] spin contribution) as obtained from the combination of  $GW$  and DMFT (see text). For comparison with LDA and LDA + DMFT results, see [8]; for experimental spectra, see [5].

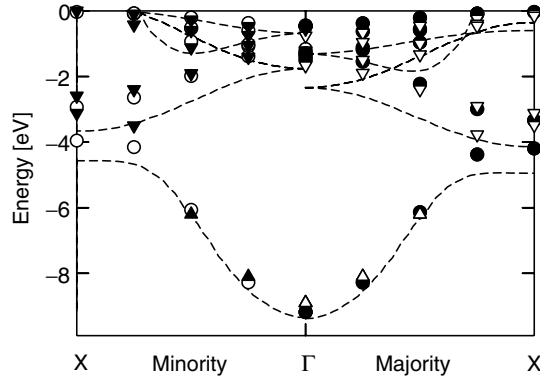


FIG. 2. Band structure of Ni (minority and majority spins) from  $GW + DMFT$  as described in the text (dots) in comparison to the LDA band structure (dashed lines) and experiments [24] (triangles down) and [5] (triangles up).

We now discuss further the simplifications made in our implementation. Because of the static approximation (iii), we could not implement self-consistency on  $W_{loc}$  [Eq. (12)]. We chose the value of  $\mathcal{U}(\omega = 0)$  ( $\approx 3.2$  eV) by calculating the correlation function  $\chi$  and ensuring that Eq. (10) is fulfilled at  $\omega = 0$ , given the  $GW$  value for  $W_{loc}(\omega = 0)$  ( $\approx 2.2$  eV for Ni [25]). The resulting impurity self-energy  $\Sigma_{imp}$  is then much smaller than the local component of the  $GW$  self-energy (or than  $V_{xc}^{loc}$ ), especially at high frequencies. It is thus essential to choose the second term in (7) to be the on-site component of the  $GW$  self-energy rather than the right-hand side of Eq. (9). For the same reason, we included  $V_{xc}^{loc}$  in Eq. (13) (i.e., we implemented a mixed scheme which starts from the LDA Hamiltonian for the local part and thus still involves a double-counting correction). We expect that these limitations can be overcome in a self-consistent implementation with a frequency-dependent  $\mathcal{U}(\omega)$  [hence fulfilling Eq. (9)].

In conclusion, we have proposed an *ab initio* approach to the electronic structure of strongly correlated materials, which combines  $GW$  and DMFT. The scheme aims at avoiding the conceptual problems inherent to LDA + DMFT methods, such as double-counting corrections and the use of Hubbard parameters assigned to correlated orbitals. A full implementation of the  $GW + DMFT$  scheme is a major goal for future research, which requires further work on impurity models with  $\omega$ -dependent interactions [26–28] as well as studies of the various self-consistency schemes.

During completion of this work, we learned about Ref. [28] in which a  $GW$  correction to the EDMFT scheme has been successfully implemented for an extended Hubbard model. We thank G. Kotliar for providing a copy of this work prior to publication. We are grateful, for comments and helpful discussions, to S. Florens, G. Kotliar, P. Sun, and to A. Lichtenstein (who also shared with us his QMC code). This work has benefitted from the hospitality of the MPI-FKF Stuttgart (for which we

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