A first principles approach to correlated systems – Combining the GW method and dynamical mean field theory

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cond-mat/0207419

Outline

- Some motivations
- Combining GW and Dynamical Mean Field Theory (DMFT): a functional point of view
- Towards a first principles method for correlated materials – some open questions
- Combining GW and DMFT: a simplified implementation
- Application to ferromagnetic Nickel

LDA+DMFT

- LDA Hamiltonian in a localized basis
- Hubbard interaction term for correlated orbitals

$$H = \sum_{\{im\sigma\}} (H^{LDA}_{im,i'm'} - H^{double\ counting}_{im,i'm'}) a^{+}_{im\sigma} a_{i'm'\sigma}$$
$$+ \frac{1}{2} \sum_{imm'\sigma\ \text{(correl. orb.)}} U^{i}_{mm'} n_{im\sigma} n_{im'-\sigma}$$
$$+ \frac{1}{2} \sum_{im\neq m'\sigma\ \text{(correl. orb.)}} (U^{i}_{mm'} - J^{i}_{mm'}) n_{im\sigma} n_{im'\sigma}$$

- treat within DMFT

In nature ...

- *long-range* Coulomb interactions
- self-energies are *non-local*

Can we

- treat *long-range* Coulomb interactions in an ab initio way?
- avoid double counting correction terms?
- include non-local corrections to the self-energy?

$\rightarrow$ “GW+DMFT”

A functional point of view

[Chitra & Kotliar, 2001, Almbladh et al. 1999]

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

Free energy Γ is a functional of

- one-electron Green's function
  $G(\mathbf{r},\mathbf{r}';\tau-\tau') \equiv -\langle T_\tau\, \psi(\mathbf{r},\tau)\psi^\dagger(\mathbf{r}',\tau')\rangle$
- the screened Coulomb interaction
  $W = V - V\chi V$

G_H = bare (Hartree) Green's function

$\Psi[G,W] \sim \int_0^1 d\alpha \int d\mathbf{r}\, d\tau \langle \phi(\mathbf{r},\tau) : \psi^\dagger(\mathbf{r},\tau)\psi(\mathbf{r},\tau) :\rangle$

Approximations to $\Psi[G,W]$?

Extended DMFT ("E-DMFT"):
$\Psi_{DMFT}[G,W] = \Psi_{impurity}[G_{impurity}, W_{impurity}]$

GW:
$\Psi_{GW}[G,W] = -\frac{1}{2} TrGWG$

Combine E-DMFT for local part with GW for nonlocal part:
$\Psi_{GW+DMFT}[G,W] =$
$\Psi_{DMFT}[G,W] + \Psi_{GW}^{nonlocal}[G,W]$

NB: "local" = "onsite" is a basis-set dependent notion!

$$\Psi_{GW}^{\text{nonloc}} \sim \int d\tau \sum_{L_i} \sum_{\mathbf{R}\neq\mathbf{R}'} G^{\mathbf{R}\mathbf{R}'}_{L_1 L_1'} W^{\mathbf{R}\mathbf{R}'}_{L_1 L_2 L_1' L_2'} G^{\mathbf{R}'\mathbf{R}}_{L_2' L_2}$$

GW+DMFT: local part

$\Psi_{imp}[G_{imp}, W_{imp}]$ calculated from local impurity model:

$$S_{imp} = \int d\tau d\tau' \Big[-\sum c^\dagger_L(\tau) \mathcal{G}^{-1}_{LL'}(\tau - \tau') c_{L'}(\tau') \quad (\text{C}$$

$$+ \quad \sum : c^\dagger_{L_1}(\tau) c_{L_2}(\tau) : \mathcal{U}_{L_1 \ldots L_4}(\tau - \tau') : c^\dagger_{L_3}(\tau') c_{L}$$

$$G^{LL'}_{imp} = -\langle T_\tau c_L(\tau) c^+_{L'}(\tau') \rangle_S$$

$$\chi_{L_1 L_2 L_3 L_4} = \langle : c^\dagger_{L_1}(\tau) c_{L_2}(\tau) :: c^\dagger_{L_3}(\tau') c_{L_4}(\tau') : \rangle_S$$

$$W_{imp} = \mathcal{U} - \mathcal{U} \chi \mathcal{U}$$

GW+DMFT (contd)

Combine local self-energy and polarization

$$\Sigma^{xc}_{imp} \equiv \delta\Psi_{imp}/\delta G_{imp} = \mathcal{G}^{-1} - G^{-1}_{imp}$$
$$P_{imp} \equiv -2\delta\Psi_{imp}/\delta W_{imp} = \mathcal{U}^{-1} - W^{-1}_{imp}$$

with non-local self-energy and polarization:

$$\begin{aligned}
\Sigma^{xc}(\mathbf{k}, i\omega_n)_{LL'} &= \Sigma^{xc}_{GW}(\mathbf{k}, i\omega_n)_{LL'} \\
&- \sum_{\mathbf{k}} \Sigma^{xc}_{GW}(\mathbf{k}, i\omega_n)_{LL'} + [\Sigma^{xc}_{imp}(i\omega_n)] \\
P(\mathbf{q}, i\nu_n)_{\alpha\beta} &= P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} \\
&- \sum_{\mathbf{q}} P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} + P^{imp}(i\nu_n)_{\alpha\beta}
\end{aligned}$$

Self-consistency condition

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

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

Update Weiss field and impurity interaction:

$$\mathcal{G}^{-1} = G_{loc}^{-1} + \Sigma_{imp}$$

$$\mathcal{U}^{-1} = W_{loc}^{-1} + P_{imp}$$

Iterate until self-consistency ...

Self-consistency loop

Impurity model :

$$\mathcal{G}(\tau), \mathcal{U}(\tau)$$

$$G_{imp} \equiv -\langle T_\tau \, c c^\dagger \rangle_S \;\rightarrow\; \Sigma^{xc}_{imp} = \mathcal{G}^{-1} - G^{-1}_{imp}$$

$$W_{imp} = \mathcal{U} - \mathcal{U}\chi\mathcal{U} \qquad P_{imp} = \mathcal{U}^{-1} - W^{-1}_{imp}$$

$$\uparrow \qquad\qquad \downarrow$$

Update

$$\mathcal{G}^{-1} = G^{-1}_{loc} + \Sigma_{imp}$$

$$\mathcal{U}^{-1} = W^{-1}_{loc} + P_{imp}$$

Combine :

$$\Sigma = \Sigma_{imp} + \Sigma^{nonlocal}_{GW}$$

$$P = P_{imp} + P^{nonlocal}_{GW}$$

$$\uparrow \qquad\qquad \downarrow$$

$$\text{Self} - \text{consistency}$$

$$G_{loc} = \sum_{\mathbf{k}} [{G_H}^{-1} - \Sigma^{xc}]^{-1}$$

$$W_{loc} = \sum_{\mathbf{q}} [V^{-1}_{\mathbf{q}} - P]^{-1}$$

Challenges and questions

- Global self-consistency?
- Treat all orbitals – localized and delocalized – on equal footing?
- Choice of orbitals? Hamiltonian? Downfolding?
- How to solve the dynamical impurity model?
  $\rightarrow$ P.Sun & G.Kotliar
  $\rightarrow$ S. Florens, work in progress
  here: static approximation

A simplified implementation

Non-selfconsistent GW + local Σ from static impurity model

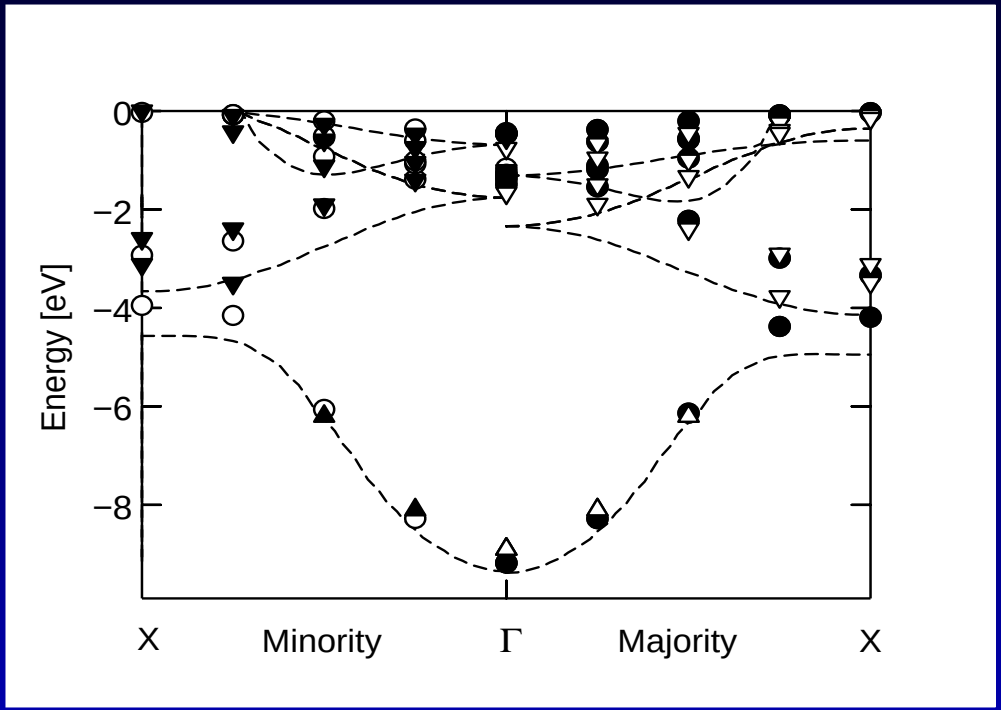
$$G^{\sigma}_{loc}(i\omega_n) = \sum_{\mathbf{k}} [{G_H}^{-1}(\mathbf{k}, i\omega_n) - (\Sigma^{xc}_{GW})_{non-loc} - (\Sigma_{imp,\sigma} - \frac{1}{2}\mathbf{Tr}_{\sigma}\Sigma_{imp,\sigma}(0) + V^{loc}_{xc})\,]^{-1}$$

Nonlocal part: correct Hartree by GW

Local part: correct LDA by DMFT

Simplified GW+DMFT

Ni band structure

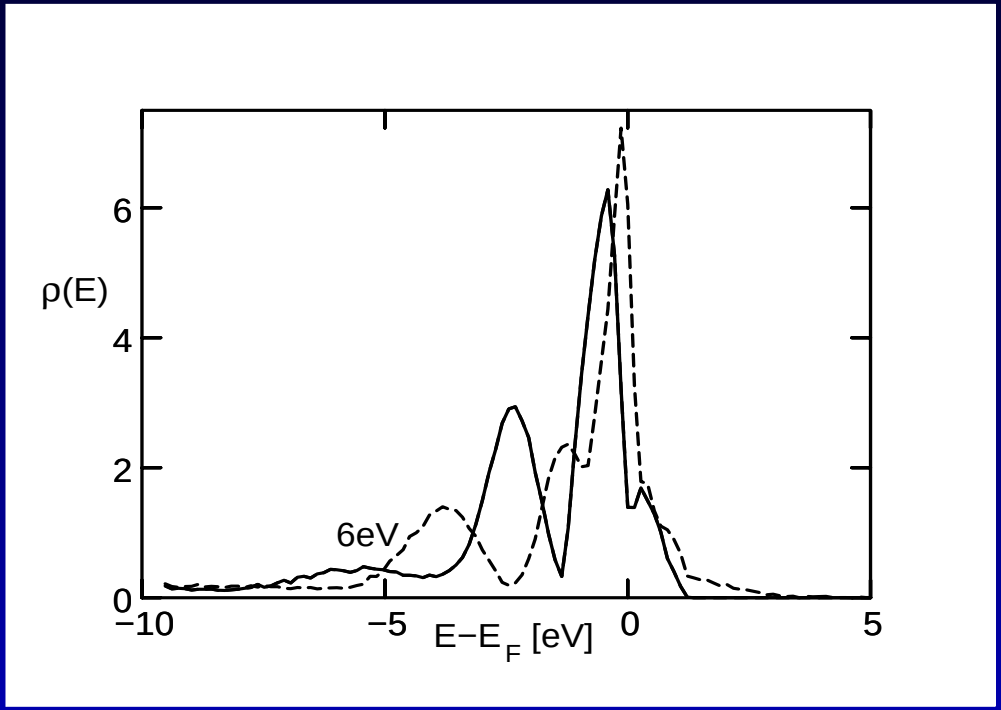


Circles: GW+DMFT
Dashed: LDA
Triangles: photoemission data

(Bünemann et al. 2002, Mårtensson et al., 1984)

Simplified GW+DMFT:

Spectral function of Ni



Majority and minority spins

Conclusion and perspectives

Combination of GW and DMFT ...

- describes long-range interactions in an ab initio way
- includes non-local self-energy effects

Questions and perspectives:

- Dynamical impurity models in realistic context?
- Effects of self-consistency?
- Choice of orbitals, Hamiltonian?

An exciting new fi eld with many open problems!