Recent Progress in ab-initio Treatment of Correlations in Solids within Green Function Methods
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Many Electron Collaboration
Annual Meeting

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Support
Simons Foundation
DOE CMS
Many Body Problem in Solids

Test the locality of various many body approximations with different projectors. Use of all electron LAPW basis. Use Matsubara formalism.
GW+DMFT: some references

- Chitra and Kotliar PRB 63, 115110 (2001)
Motivation I

• Combining the ideas of using “free electron” as a reference (DFT) for extended orbitals with that of a local “dressed atom” (DMFT) for the more localized ones, was very successful.

• LDA+DMFT has had many successes across the periodic table.

• Understand why it works from a diagramatically controlled perspective.

• Suggest improvements.
The light, SP (or SPD) electrons are extended describe them by LDA

The heavy, d (or f) electrons are more localized so we treat them with DMFT.

Take for the one electron Hamiltonian the LDA Kohn Sham Hamiltonian with a shift of the heavy electron (double counting corrections, dc) in the spirit of the Anderson Impurity Model

The U matrix estimated (from constrained LDA), other methods were suggested later (i.e. constrained RPA)

Solve for Greens function using DMFT

Later the method was made charge self consistent and derived from a functional to obtain total energies. (Chitra 2000, Savrasov 2003)

More accurate prescription for the dc was proposed (Haule 2015)
LDA+DMFT Across the Periodic Table

1) Heavy Fermion

2) Pu and Cm

3) Iron pnictides

4) Cuprates

5) Ruthenates

8) Iridates

\[ \Sigma(k, \omega) \approx \Sigma(k) + \sum_{\vec{R}, \alpha \beta \in L} |\vec{R}\alpha\rangle \Sigma_{\alpha R, \beta R}(\omega) \langle \vec{R}\beta| \]

Motivation II: correlated semiconductor FeSb2 gigantic thermopower
Bentien et. al. EPL 80 17008 (2007). Material is not well described by LDA+DMFT. This differs from FeSi which is one of LDA+DMFT success stories.

Spectral weight $N_{\text{eff}}$ does not recover until 2 eV.
DFT – GW  Non interacting electrons as a reference.
Density Functional Theory (Kohn Hohenberg, Kohn Sham 1964)

\[
\Gamma [\rho (r)] = \Gamma_{\text{univ}} [\rho (r)] + \int dr V_{\text{cryst}} (r) \rho (r)
\]

\[-\nabla^2 + V_{KS} (r) [\rho] \psi_{kj} = \varepsilon_{kj} \psi_{kj}\]

\[
\rho (r) = \sum_{\varepsilon_{kj} < 0} \psi_{kj}^* (r) \psi_{kj} (r)
\]

Starting point for perturbation theory in the screened Coulomb interactions (Hedin 1965) \(G_0 W_0\) Hybertsen and Louie (1985)

\[
G^{-1} = G^{-1}_{0 \; KS} \; - \; V_{xc}
\]

\[
G^{-1}_{0 \; KS} = i\omega_n + \mu + \nabla^2 - V_{KS} (r) [\rho]
\]

\[
V_{KS} = V_{\text{Hartree}} + V_{\text{cryst}} + V_{\text{xc}}.
\]

S. Louie in Topics in Computational Physics World Scientific (1997)

M.vanSchilfgaarde,T.Kotani,S.Faleev PRL.96226402 (2006) self consist on Vxc
\[
H^A_H = -\nabla^2 + V^A_H(r)[\rho] + V_{\text{cryst}}
\]

Many body theory in $W$ and $G$.

$$\Gamma[G,W] = \text{TrLn}G - \text{Tr}[G^{-1}_0 - G^{-1}]G - \frac{1}{2} \text{TrLn}W + \frac{1}{2} \text{Tr}[V^{-1}_C - W^{-1}]W + E_{\text{hartree}} + \Phi[G,W]$$

$$\Phi_{GW}(G,W) = G^{(rr')} + G^{(rr')}$$

R. Chitra, and G. Kotliar \ arXiv:cond-mat/9911223

Introduce projector $G_{\text{loc}}$ $W_{\text{loc}}$

$$\Phi_{\text{DMFT}}(G,W) = G_{\text{loc}} + W_{\text{loc}}$$

See also Almbladh et. al.

DMFT summation of ALL the local diagrams.
Band Gaps of Semiconductors

A. Kutepov: PRB 95, 195120 (2017)
GW+DMFT loop and its linearization

LqsGW+DMFT (S. Choi et. al. submitted to CPC(2018))

\[ G^{-1}(k, i\omega_n) = G^{-1}_{MF}(k, i\omega_n) - f_k \tilde{\Sigma}_{imp}(i\omega_n) f_k^\dagger \]

\[ \tilde{\Sigma}_{imp} = \tilde{\mathcal{G}}^{-1} - \tilde{G}_{imp}^{-1} \]

\[ \tilde{W}_{imp} = \tilde{\mathcal{U}} \tilde{\mathcal{U}}^{\dagger} \tilde{\Sigma}_{imp} \tilde{\mathcal{U}} \]

\[ \tilde{P}_{imp} = \tilde{\mathcal{U}}^{-1} \tilde{W}_{imp}^{-1} \]

\[ \tilde{\mathcal{G}}^{-1} = \tilde{G}_{loc}^{-1} + \tilde{\Sigma}_{imp} \]

\[ \tilde{U}^{-1} = \tilde{W}_{loc}^{-1} + \tilde{P}_{imp} \]

\[ G^{-1}(k, i\omega_n) = G^{-1}_0(k, i\omega_n) - \Sigma(k, i\omega_n) \]

\[ W^{-1}(k, i\omega_n) = V^{-1}(k) P(k, i\omega_n) \]

\[ \tilde{G}_{loc}(i\omega_n) = \frac{1}{N_k} \sum_k f_k^\dagger G(k, i\omega_n) f_k \]

\[ \tilde{W}_{loc}(i\omega_n) = \frac{1}{N_k} \sum_k b_k^\dagger W(k, i\omega_n) b_k \]
LQSGW+DMFT  LaCuO₄  Choi et. al. npj Quantum Materials 1, 16001 (2016).

- O-p and La-f orbitals

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**DOS**

$LDA + DMFT$  
LQSGW+DMFT  
$EXP$

$La_2CuO_4$

$NiO$

$E - E_F (eV)$
Role of frequency dependence in U
FeSb2 low temperature spectral functions. Comparison among methods.
Recent photoemission results Chikina et al. (Radovic’s group at PSI)
Optical Conductivity FeSb$_2$

Quasi-one dimensional structure (not visible in the structure).

Surprise in the phonon spectra.
Conclusion

• Combining Hedin’s program of perturbation theory in G and W, with locality DMFT ideas.
• Showed some encouraging results
• Systematics in the gaps of semiconductors for the weak correlations side (also in total energies not presented)
• Need to complete and explore fully self consistent GW+DMFT in the large energy window to make precise correspondence to LDA+DMFT.
• Interesting physics in FeSb2. Quasi-one dimensionality in a three dimensional material.
• Thermopower not fully understood. Role of vacancies, phonon drag M. Battiato et. al. PRL 236603 (2015)
Table 1
Band gaps (eV) of selected semiconductors and insulators in comparison with the experimental data. We also include the error (%) relative to the experiment. For the materials with known effects of the electron-phonon and/or the spin-orbit interaction (Si, SiC, C, GaAs, MgO, ZnS, ZnSe, LiF, BN, AlP), the corresponding effects were excluded from the raw experimental data to facilitate the comparison with the calculations. Experimental data have been cited from Refs. [17,24,15,25–27].

<table>
<thead>
<tr>
<th>Material</th>
<th>[15,24]</th>
<th>[16]</th>
<th>[17]</th>
<th>Present work</th>
<th>Exp.</th>
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</thead>
<tbody>
<tr>
<td>Si</td>
<td>1.23</td>
<td>1.41</td>
<td>1.47</td>
<td>1.41(14.8%)</td>
<td>1.22</td>
</tr>
<tr>
<td>SiC</td>
<td>2.14</td>
<td>2.88</td>
<td>2.90</td>
<td>3.08(22.7%)</td>
<td>2.51</td>
</tr>
<tr>
<td>C</td>
<td>6.52</td>
<td>6.18</td>
<td>6.40</td>
<td>6.71(14.1%)</td>
<td>5.88</td>
</tr>
<tr>
<td>GaAs</td>
<td>1.93</td>
<td>1.85</td>
<td>1.75</td>
<td>2.08(23.1%)</td>
<td>1.69</td>
</tr>
<tr>
<td>ZnO</td>
<td>3.87</td>
<td>3.8</td>
<td>4.61</td>
<td>4.47(24.2%)</td>
<td>3.60</td>
</tr>
<tr>
<td>MgO</td>
<td>9.16</td>
<td>9.29</td>
<td>9.42</td>
<td>9.42(18.0%)</td>
<td>7.98</td>
</tr>
<tr>
<td>ZnS</td>
<td>4.04</td>
<td>4.15</td>
<td>4.19</td>
<td>4.19(6.3%)</td>
<td>3.94</td>
</tr>
<tr>
<td>ZnSe</td>
<td>3.08</td>
<td></td>
<td>3.17</td>
<td>3.17(5.7%)</td>
<td>3.00</td>
</tr>
<tr>
<td>LiF</td>
<td></td>
<td>15.9</td>
<td></td>
<td>16.63(14.8%)</td>
<td>14.48</td>
</tr>
<tr>
<td>NaCl</td>
<td></td>
<td></td>
<td></td>
<td>9.81(15.4%)</td>
<td>8.5</td>
</tr>
<tr>
<td>BN</td>
<td>7.14</td>
<td>7.51</td>
<td>7.06</td>
<td>7.06(7.0%)</td>
<td>6.6</td>
</tr>
<tr>
<td>AlP</td>
<td>2.90</td>
<td>3.10</td>
<td>2.80</td>
<td>2.80(13.4%)</td>
<td>2.47</td>
</tr>
<tr>
<td>NiO</td>
<td>4.8</td>
<td>4.97</td>
<td>4.47</td>
<td>4.47(3.9%)</td>
<td>4.3</td>
</tr>
<tr>
<td>Cu₂O</td>
<td>2.36</td>
<td>2.65</td>
<td>2.42</td>
<td>2.42(10.0%)</td>
<td>2.20</td>
</tr>
<tr>
<td>TiO₂</td>
<td>3.78</td>
<td>4.22</td>
<td>3.80</td>
<td>3.80(22.6%)</td>
<td>3.1</td>
</tr>
<tr>
<td>Sr TiO₃</td>
<td>4.19</td>
<td></td>
<td>4.01</td>
<td>4.01(21.5%)</td>
<td>3.3</td>
</tr>
<tr>
<td>CeO₂</td>
<td>~5</td>
<td></td>
<td></td>
<td>5.83(70.1%)</td>
<td>3-3.5</td>
</tr>
</tbody>
</table>


Explore Window Dependence

La$_2$CuO$_4$

DOS

PDOS

$E-E_F$(eV)

$E-E_F$(eV)
LQSGW+DMFT validation on NiO

• **Role of frequency dependent U**

- a proper choice of static U gives similar spectral function to Exp and full LQSGW+DMFT