EMBEDDED DYNAMICAL MEAN FIELD THEORY FUNCTIONAL (EDMFTF)



OF NEW JERSEY





Simons Collaboration on the Many Electron Problem



PART I EDMFTF

Projectors and locality of correlations

Rev. Mod. Phys. 78, 865 (2006)

G. Kotliar, S.Y. Savrasov, K Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti. **Phys. Rev. B 81, 195107 (2010)**, *K. Haule,* Chuck-Hou Yee, Kyoo Kim.

• Exact double-counting of DFT+EDMFTF (PRL 115, 196403 (2015)).

PART 2 EDMFTF

- Stationary free energy functional (within Embedded DMFT approach) for structural optimization, <u>PRLII5</u>, 256402 (2015).
- Implementation of Forces within E-DMFT functional for optimization of internal structural parameter, (Phys. Rev. B 49, 195146 (2016), K. Haule, G.L. Pascut)

PART I EDMFTF

• Projectors and locality of correlations

Theoretical Perspective - Correlated Matter



- Electrons have dual nature, partly itinerant and partly localized
- Need to incorporate the real space perspective: electrons are not just waves but are also particles — localization due to interaction.
- The electron density rho is not quite enough to describe the situation. We need to think of electron spectra Green's function.



Basic variable is Green's function: == dynamic analog of charge density

SOME FORMULAS FOR SLIDES

 $\text{Luttinger-Ward functional (1950):} \\ \Gamma[\{G\}] \stackrel{\Sigma}{=} \stackrel{G^{-1}}{=} \stackrel{G^{-1}}{\to} \stackrel{G^{-1}}{\to}$

AS FOR IS GENERAL CONTROL CON



(3)

$$\frac{GT[G]}{\delta G} = \frac{1}{G} - \Sigma - G \frac{\delta Z}{\delta G} + \frac{1}{G}$$

$$\frac{\Gamma[G] = Tr InG - Tr(\Sigma G) + \Phi[G]}{\Gamma[G] = Tr InG - Tr(\Sigma G) + \Phi[G]} = 0$$
(4)
Density Functional Topp(G) = 0
(5)

$$\frac{\delta \Gamma[G]}{\delta G} = \frac{1}{G} - \Sigma - G \frac{\delta \Sigma}{\delta G} + \frac{1}{G} + \frac{$$

DFT In the LW language

We can get DFT by $\Phi[{G}] \to E_H[\rho] + E_{XC}[\rho]$

The LW functional then leads to identical equations as DFT.

Exact DFT gives exact ground state energy and charge density. But in this approach DFT is an approximation for the Green's function, which gives an approximate spectra, i.e., bands. The spectra can then interpreted as physical within such approximation for $\Phi[\{G\}]$

The fact that bands are very reasonable within LDA/GGA is an indication that correlations are very local in real space.

Weaker statement : The picture in which correlations are treated as local is very good — convergence faster when considering local correlations rather than long range correlations.

An illustrative example of local correlations

Electron gas problem

Interaction can be tuned from short to long ranged by parameter lambda:





Functional Point of view

DMFT for lattice models: $\Phi[\{G_{ij}\}] \rightarrow \sum_{i} \Phi[\{G_{ii}\}]$ *i* is site or cluster



DMFT for continuous problems: $\Phi[\{G\}] \to \sum_{\mathbf{R}_i} \Phi[\{\hat{P}_{\mathbf{R}_i}G\}]$

Need to define projector to site (or cluster): $\hat{P}_{\mathbf{R}_i}$ DMFT is projector dependent approximation

How is correlation potential determined?



In DMFT we want to lift the restriction and compute all correlations local to a given site (not given point in space).



The continuous DMFT problem

DMFT approximation: $\Phi[{G}] \to \sum \Phi[{\hat{P}_{\mathbf{R}_i}G}]$ in continuum requires **discretization** of projector, $\hat{P}_{\mathbf{R}_{i}} \to P_{\mathbf{R}_{i}}(\alpha\beta) = \int d\mathbf{r} d\mathbf{r}' \mathcal{P}_{\mathbf{R}_{i}}(\alpha\beta;\mathbf{rr}')$ for example: $P_{\mathbf{R}_i}(\alpha\beta) = \int d\mathbf{r} d\mathbf{r}' \phi_\alpha(\mathbf{r} - \mathbf{R}_i) \phi_\beta^*(\mathbf{r}' - \mathbf{R}_i)$ where $\phi_{\alpha}(\mathbf{r} - \mathbf{R}_i) \equiv \langle \mathbf{r} | \phi_{\alpha}^i \rangle$ forms a basis on a given atom so that:

$$G_{local}^{i}(\mathbf{r},\mathbf{r}') = \sum_{\alpha\beta} \langle \mathbf{r} | \phi_{\alpha}^{i} \rangle \langle \phi_{\alpha}^{i} | G | \phi_{\beta}^{i} \rangle \langle \phi_{\beta}^{i} | \mathbf{r}' \rangle$$

$$quasi \ atomic \ orbitals$$

$$(locally \ complete \ set)$$

$$\alpha, \beta \ are \ orbitals$$

 α, β are orbital-spin indices

How local are correlations?

Correlations are local in large d (large connectivity z) where DMFT is exact -- Weiss mean field theory



What about finite D? What about 0?

H₂ molecule:



How local are correlations ? 0-D test of the single site DMFT.



Juho Lee, KH, <u>PRB</u> 91, 155144 (2015).

Error of total energy using LDA+DMFT <0.2%!

DFT+Embedded Dynamical Mean Field Theory Functional



$$\Gamma[G] = Tr \log G - \text{Tr}((G_0^{-1} - G^{-1})G) + E_{Vc}^{H+XC}[\rho] + \Phi_{V_{DMFT}}^{DMFT}[G_{local}] - \Phi_{V_{DMFT}}^{H+XC}[\rho_{local}]$$

$$\mathbf{G}_0^{-1} = [i\omega_n + \mu + \nabla^2 - V_{nuc}(\mathbf{r})]\delta(\mathbf{r} - \mathbf{r}')$$

$$\mathbf{non-interacting part of G}$$

$$\mathbf{Hartree + XC}_{functional}$$

$$\mathbf{G}_0^{-1} = [i\omega_n + \mu + \nabla^2 - V_{nuc}(\mathbf{r})]\delta(\mathbf{r} - \mathbf{r}')$$

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$$\mathbf{G}_0^{-1} = [i\omega_n + \mu + \nabla^2$$

 Γ [G] Is stationary and gives free energy of the system.

$$\frac{\delta\Gamma[G]}{\delta G} = 0 \quad \text{because} \quad G^{-1} - G_0^{-1} + V_{H+XC}\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') + \\ + \hat{P}\frac{\delta\Phi^{DMFT}[G_{local}]}{\delta G_{local}} - \hat{P}\frac{\delta\Phi^{DC}[\rho_{local}]}{\delta\rho_{local}}\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') = 0$$

Stationarity : 1st order error in G leads to 2nd order error in free energy.

Note: Migdal-Galitskii formula gives non-stationary total energies in DFT+DMFT.

Embedded Dynamical Mean Field Theory Functional



Note: The trick of mapping is used only to determine the exchange-correlation potential, while the *kinetic energy* and *Hartree* term are always treated exactly.

Quantum impurity model solved by Monte Carlo

Embedded Dynamical Mean Field Theory Functional

Details on extremization:

$$\frac{\delta}{\delta G}\Gamma[\{G\}] = \frac{\delta}{\delta G}(\operatorname{Tr}\log G - \operatorname{Tr}((G_0^{-1} - G^{-1})G)) + \frac{\delta E_{V_c}^{H+XC}[\rho]}{\delta G} + \frac{\delta \sum_{\mathbf{R}_i} \Phi_{V_{DMFT}}[\{\int d\mathbf{r} d\mathbf{r}' \phi_{\alpha}(\mathbf{r} - \mathbf{R}_i)G(\mathbf{rr}')\phi_{\beta}^*(\mathbf{r}' - \mathbf{R}_i)\}]}{\delta G} - \frac{\delta \sum_{\mathbf{R}_i} \Phi_{V_{DMFT}}^{H+XC}[\{\int d\mathbf{r} d\mathbf{r}' \phi_{\alpha}(\mathbf{r} - \mathbf{R}_i)\rho(\mathbf{rr}')\phi_{\beta}^*(\mathbf{r}' - \mathbf{R}_i)\}]}{\delta G}$$

$$\frac{\delta}{\delta G} \Gamma[\{G\}] = G^{-1} - G_0^{-1} + \frac{\delta E_{V_c}^{H+XC}[\rho]}{\delta \rho} \delta(\mathbf{r} - \mathbf{r}') \delta(\tau - \tau') \qquad \left(\rho(\mathbf{r}) = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')G(\mathbf{r}\tau, \mathbf{r}'\tau')\right) + \sum_{\alpha\beta} \frac{\delta \Phi_{V_{DMFT}}[\{G_{local}^i\}]}{\delta G_{local\beta\alpha}^i} \phi_{\alpha}(\mathbf{r} - \mathbf{R}_i) \phi_{\beta}^*(\mathbf{r}' - \mathbf{R}_i) - \sum_{\alpha\beta} \frac{\delta \Phi_{V_{DMFT}}^{H+XC}[\{\rho_{local}\}]}{\delta \rho_{local\beta\alpha}} \phi_{\alpha}(\mathbf{r} - \mathbf{R}_i)\delta(\tau - \tau')\phi_{\beta}^*(\mathbf{r}' - \mathbf{R}_i)\}] = 0$$

finally

$$G^{-1} = G_0^{-1} - V_{H+XC}(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') - \langle \mathbf{r} | \phi_{\alpha}^i \rangle (\Sigma^{imp} - V^{DC})_{\alpha\beta} \langle \phi_{\beta}^i | \mathbf{r}' \rangle$$

where
$$\Sigma_{\alpha\beta}^{imp} = \frac{\delta \Phi_{V_{DMFT}}[\{G_{local}^{i}\}]}{\delta G_{local\beta\alpha}^{i}}$$
 and $V_{\alpha\beta}^{DC} = \frac{\delta \Phi_{V_{DMFT}}^{H+XC}[\{\rho_{local}\}]}{\delta \rho_{local\beta\alpha}}\delta(\tau - \tau')$

Embedded Dynamical Mean Field Theory Functional

$$G^{-1} = G_0^{-1} - \underbrace{V_{H+XC}(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')}_{DFT} - \underbrace{\langle \mathbf{r} | \phi_{\alpha}^i \rangle (\Sigma^{imp} - V^{DC})_{\alpha\beta} \langle \phi_{\beta}^i | \mathbf{r}' \rangle}_{quantum impurity S. embedded}$$
where $\Sigma_{\alpha\beta}^{imp} = \frac{\delta \Phi_{V_{DMFT}}[\{G_{local}^i\}]}{\delta G_{local\beta\alpha}^i}$ and $V_{\alpha\beta}^{DC} = \frac{\delta \Phi_{V_{DMFT}}^{H+XC}[\{\rho_{local}\}]}{\delta \rho_{local\beta\alpha}}\delta(\tau - \tau')$

On each correlated site \mathbf{R}_i we have to solve a $\Sigma_{\alpha\beta}^{imp} = \frac{\delta \Phi_{V_{DMFT}}[\{G_{local}^i\}]}{\delta G_{local\beta\alpha}^i}$ quantum impurity model with $\alpha\beta$ orbitals And **Embed** self-energies to continuum space by

Notice that once the projector is defined, embedding is uniquely given by the same matrix elements of $\mathcal{P}_{\mathbf{R}_i}(\alpha\beta;\mathbf{rr'}) = \phi_{\alpha}(\mathbf{r} - \mathbf{R}_i)\phi_{\beta}^*(\mathbf{r'} - \mathbf{R}_i)$

$$\hat{P}_i$$

$$\hat{P}_i$$

$$\hat{P}_i^{-1} \equiv \hat{E}_i$$

 $\langle \mathbf{r} | \phi^i_{\alpha} \rangle \Sigma^{imp}_{\alpha\beta} \langle \phi^i_{\beta} | \mathbf{r'} \rangle$

REQUIREMENT FOR STATIONARITY

Projector should not depend on the solution

Return to definition of the projector:

$$G_{local,\mathbf{R}_{i},\alpha\beta} = \int_{\mathbf{rr'}} \mathcal{P}_{\mathbf{R}_{i}}(\alpha\beta;\mathbf{rr'})G(\mathbf{rr'})$$

Return to saddle point Eq.: $\frac{\delta \Phi^{DMFT}[\{G_{local,\mathbf{R}_i}\}]}{\delta G(\mathbf{rr'})} = \sum_{\alpha\beta} \left(\frac{\delta G_{local,\mathbf{R}_i,\alpha\beta}}{\delta G(\mathbf{rr'})} \right) \frac{\delta \Phi[\{G_{local,\mathbf{R}_i}\}}{\delta G_{local,\mathbf{R}_i,\alpha\beta}} = \sum_{\alpha\beta} \mathcal{P}_{\mathbf{R}_i}(\alpha\beta;\mathbf{rr'}) \right)$ $\int \frac{\delta \Phi[\{G_{local,\mathbf{R}_{i},\alpha\beta}\}]}{\delta G_{local,\mathbf{R}_{i},\alpha\beta}}$ $\frac{\delta P}{\delta G} = 0 \quad \text{or} \quad \frac{\delta \phi_{\alpha}(\mathbf{r} - \mathbf{R}_{i})}{\delta \rho} = 0$ Here we implicitly assumed that otherwise more complicated terms would appear... Requirement: $\frac{\delta P}{\delta G} = 0$ is not satisfied with Wannier functions because they essentially depend on the charge, and hence on G.

REQUIREMENT FOR CAUSAL DMFT EQ.



If projection separable: $\mathcal{P}_{\mathbf{R}_{i}}(\alpha\beta;\mathbf{rr}') = \phi_{\alpha}(\mathbf{r}-\mathbf{R}_{i})\phi_{\beta}^{*}(\mathbf{r}'-\mathbf{R}_{i})$ Define: $\phi_{\alpha}^{*}(\mathbf{r}-\mathbf{R}_{i}) \equiv U(\alpha,\mathbf{r})$ Satisfied: $\hat{P}\hat{E}^{-1} = U[U^{\dagger}U]^{-1}U^{\dagger} = 1$

LDA+U projector

non-separable:

 $P^{0}(lm, lm', \mathbf{rr'}) = Y_{lm}(\hat{\mathbf{r}})\delta(r - r')Y_{lm'}(\hat{\mathbf{r}'})$

SUMMARY: PROJECT/EMBED

Phys. Rev. B 81, 195107 (2010), K. Haule, Chuck-Hou Yee, Kyoo Kim.



DMFT in continuum problem (Project/Embed):

Projection:
$$G_{local,\mathbf{R}_{i},\alpha\beta} \equiv \int d\mathbf{r} d\mathbf{r}' \mathcal{P}_{\mathbf{R}_{i}}(\alpha\beta;\mathbf{rr}')G(\mathbf{rr}')$$

 $\Sigma(\mathbf{rr'}) = \sum \mathcal{P}_{\mathbf{R}_i}(\alpha\beta;\mathbf{rr'})\Sigma_{local,\mathbf{R}_i,\alpha\beta}$

Embedding:

$$\begin{split} \mathbf{R}_{i,\alpha\beta} \\ G(\mathbf{r},\mathbf{r}') &= (\delta(\mathbf{r}-\mathbf{r}')(i\omega+\mu+\nabla^2-V_{ext}(\mathbf{r})-V_{xc}(\mathbf{r}))-\Sigma^{DMFT}(\mathbf{r},\mathbf{r}'))^{-1} \\ \frac{\delta P}{\delta G} &= 0 \quad \text{ and projector must be separable} \end{split}$$

Requirements:

PART I EDMFTF

- Projectors and locality of correlations
- Exact double-counting of DFT+EDMFTF



Some part of exchange/correlations counted in both approximations.

Important : Determines the oxidation state of correlated ion

only a degenerate Hubbard model approximation can "avoid" this problem

Metal-insulator transition in FeO (earth core)

High-spin to low spin transition occurs only in correct d⁶ valence state of iron.

high-spin : insulator low spin : metal





Isostructural MIT in FeO predicted by DFT-DMFT theory.

DMFT predicted MIT in FeO

Kenji Ohta, R. E. Cohen, et.al., PRL 108, 026403 (2012)

Importance of correct valence in RNiO₃

S=1

fluctuating m.

small effective

moment

Basic physical picture:

The chemical picture suggest Ni d⁷ configuration.

Bond disproportionation: Ni₁ is in d⁸ with S=1 while Ni₂ +two ligand holes $d^{8}L_{2}$ with total S=0.

"Site Selective Mott transition",

Hyowon Park, Andrew J. Millis, and Chris A. Marianetti, Phys. Rev. Lett. **109**, 156402 (2012) .

G. Sawatzky, arXiv 1608.01645

Other approaches postulate d⁸ and can not justify without experiment.

The exact double-counting gives Ni d⁸ configuration.

Haule & Pascut, ScientificReports7(2017)10375.

With two holes in the high-spin state, **Hund's coupling** induces strong correlations and Mott physics.

P21/n insulating



Double-Counting

$$\Phi[\{G\}] \rightarrow E_{V_{c}}^{H+XC}[\{\rho\}] + \sum_{\mathbf{R}_{i} \in corr} \Phi_{V_{DMFT}}[\{G_{local}^{i}\}] - \Phi_{V_{DMFT}}^{H+XC}[\{\rho_{local}^{i}\}]$$

$$E_{xact}$$
functional
approximated
$$LDA \text{ functional}$$

$$Sum \text{ of all skeleton diagrams}$$

$$local \text{ to correlated ions}$$

$$DC : \text{ intersection of DMFT}$$

$$and LDA \text{ approximation}$$

$$Hartree: E_{V_{c}}^{H}[\{\rho\}] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' V_{C}(|\mathbf{r} - \mathbf{r}'|)\rho(\mathbf{r})\rho(\mathbf{r}')$$

$$LDA - XC: \Phi_{V_{c}}[\{G\}] \rightarrow E_{XC}^{LDA}[\{\rho\}] = \int d\mathbf{r}\rho(\mathbf{r})\varepsilon_{V_{c}}^{XC}(\rho(\mathbf{r}))$$

$$\Phi^{DMFT} \text{ sums all Feyn. graphs (just like the exact } \Phi)$$

$$but uses only G_{local} and screened interaction instead of full G and V_{c}$$

$$V_{DMFT}(\mathbf{r} - \mathbf{r}') = \frac{e^{-\lambda |\mathbf{r} - \mathbf{r}'|}}{|\mathbf{r} - \mathbf{r}'|} \qquad \Phi[G_{local,\mathbf{R}_i}] = \frac{1}{2} + \frac{1}{2} + \frac{1}{2} + \frac{1}{4} + \frac{1}$$

Double-Counting of Hartree

Exact Hartree:
$$E_{V_C}^{H}[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' V_C(|\mathbf{r} - \mathbf{r}'|)\rho(\mathbf{r})\rho(\mathbf{r}')$$

LDA approximation
Exact Hartree Hartree term included
 $E_{V_C}^{H}[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' V_C(|\mathbf{r} - \mathbf{r}'|)\rho(\mathbf{r})\rho(\mathbf{r}')$

$$E_{DMFT}^{H}[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' (\hat{P}\rho(\mathbf{r})) (\hat{P}\rho(\mathbf{r}')) V_{DMFT}(\mathbf{r} - \mathbf{r}')$$

This approximation for Hartree term appears in Φ^{DMFT}

Double counting to subtract is the DMFT approximation for the Hartree term:

$$\Phi_{DMFT}^{DC,Hartree}[\{\rho\}] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' (\hat{P}\rho(\mathbf{r})) (\hat{P}\rho(\mathbf{r}')) V_{DMFT}(\mathbf{r}-\mathbf{r}')$$

Double-Counting of Exchange



Double-Counting of Exchange



Intersection of both approximations: apply **both approximations** to the functional



Double-Counting of Correlations



Summary for Double-Counting



EDMFTF Functional:

$$\Gamma[G] = Tr \log G - \text{Tr}((G_0^{-1} - G^{-1})G) + E_{Vc}^{H+XC}[\rho] + \Phi_{V_{DMFT}}^{DMFT}[G_{local}] - \Phi_{V_{DMFT}}^{H+XC}[\rho_{local}]$$

Double-counting functional:

DMFT approximation on LDA functional OR LDA approximation on DMFT functional :

$$\begin{split} \Phi_{V_{DMFT}}^{H+XC}[\rho_{local}] &= \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \rho_{local}(\mathbf{r}) V_{DMFT}(\mathbf{r} - \mathbf{r}') \rho_{local}(\mathbf{r}') + \\ &+ \int d\mathbf{r} \rho_{local}(\mathbf{r}) \epsilon_{xc}^{V_{DMFT}}[\rho_{local}(\mathbf{r})] \\ &\text{electron gas interacting with screened Coulomb interaction!} \\ &\text{only the DMFT charge density enters.} \end{split}$$

KH, PRL **II5**, 196403 (2015).

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- Implementation of Forces within E-DMFT functional for optimization of internal structural parameter, (Phys. Rev. B 49, 195146 (2016), K. Haule, G.L. Pascut)

DFT: The best Independent Particle Approach

Very useful : each electron interacts with an effective potential that mimics to some extent the effects of the other electrons.

We do not target the wave function, but rather the charge density, or the single-particle Green's function (spectral function). The two particle correlation function is almost always enough, rarely we need to go beyond the two particle correlation function.

Independent particle approach (within DFT) gives total energy and charge density very well, spectral should be unphysical, but in and simple metals and band insulators it is quite good.

DFT STRUCTURAL PREDICTIONS: SIZE OF THE IONS / UNIT CELL



crosses: experiment black lines: DFT

DFT predicts very accurately the size of unit cells in 3d&4d elements.

4f elements are essentially localized - no binding versus filling.

essentially atomic physics.

early 5f elements are quite itinerant, beyond Pu very localized=> DFT fails.

Conclusion: LDA/GGA pretty good in elemental solids, except in 4f,5f.



lattice constant in Mott insulating FeO iron(II) oxide FeO 80 non-magnetic 70 LDA 60 PBE Energy[meV] PBEsol 50 40 30 NaCl structure V_{exp} 20 Missing local 10 fluctuating moments 0⊾ 15 19 16 17 18 20 21 22 Overbinding problem 80 70 60 Energy[meV] Ordered moments 50 improve, not good 40 enough. 30 LDA-AFM V_{exp} 20 PBE-AFM **Even PBE** 10 **PBEsol-AFM** 0⊑ 15 $\frac{1}{22}$ underestimates volume. 19 16 17 18 20 21 Volume[Å³]




Can we consistently improve energies and spectra of correlated solids?

Spectra & structure of correlated oxides



green : LDA blue dots and color: exp.



SrVO3 is a correlated metal with m*/m~3. DFT predicts its volume quite right.

LDA+DMFT and the best DFT functional are in good agreement.

In many correlated metals DFT may not give good spectra, but the structure is quite good.

This is typically not the case in Mott insulator. DFT overbids, and does not account for subtle structural distortions, essential for the Mott transition.



DTF-DMFT: Haule & Birol, PRL 115, 256402 (2015).

non-perturbative treatment like DFT+DMFT.

ELECTRON-PHONON COUPLING IN FLUCTUATING MOMENT SYSTEMS

Change of the band structure due to displacement of the ions in the direction of a phonon mode.

$$g_{mn\nu}(\mathbf{k},\mathbf{q}) = \langle u_{m\mathbf{k}+\mathbf{q}} | \int d\mathbf{r}' \epsilon_{\mathrm{e}}^{-1}(\mathbf{r},\mathbf{r}',\omega) \Delta_{\mathbf{q}\nu} v^{\mathrm{en}}(\mathbf{r}') | u_{n\mathbf{k}} \rangle_{\mathrm{uc}}$$

DFT tends to underestimate g, because of RPA-overscreening



DFT+DMFT Prediction: Mandal, Cohen, & Haule, PRB 89, 220502(R) (2014).

ELECTRON-PHONON COUPLING IN FLUCTUATING MOMENT SYSTEMS



DMFT Prediction confirmed by experiment:

Pressure (GPa)

DFT+DMFT Prediction: Mandal, Cohen, & Haule, PRB 89, 220502(R) (2014). Experiment: S. Gerber, ...,Z.X. Shen et.al., Science (2017).

	Experiment (2017	7) DET+DMET (2014)) DFT	
) $DI^{T} DIVIT (2014)$		
z_{Se} (r.l.u.)	0.2653	0.27	0.2456	position of Se
f_{A1g} (THz)	5.30 ± 0.05	5.7	6.5 ± 0.3	A1g frequency
$\Delta E_{xz/yz}/\Delta z_{\rm Se}$ (meV/pm)	-13.0 ± 2.5	-10.3 to -13.4	-1.6 ± 0.2	e-ph coupling

Bands in correlated metals

Momentum resolved spectra of LiFeAs





DFT+DMFT for LiFeAs:



H Miao,....Haule, Kotliar, H. Ding, Phys. Rev. B 94, 201109(R) (2016).

Bands are sharp only very near the fermi level and only at low temperature (Fermi liquid).

Above the coherence temperature, electrons are better described as fluctuating moments, rather than plane waves. m*/m~3

DMFT predicted coherence incoherence crossover in Fe-SC (Hund's metals)

(Haule & Kotliar NJP 11, 25021 (2009))

Magnetism and moments in iron superconductors



DMFT predicts much better magnetic moments than DFT.

These are not Stoner magnets.

Have substantial local fluctuating moment at low energy.



Large fluctuating moment



Large fluctuating moment can not be explained by a purely itinerant model - property of Hundsness! The DMFT account for a dual nature of electrons in Hund's metals: itinerant and localized nature.

magnetic susceptibility in paramagnetic state

black dots : inelastic neutron scattering color : DFT-DMFT paramagnetic spin susceptibility



Magnetic susceptibility is the only quantity up to now which requires the two particle correlation function.

It is harder to compute.

DMFT can predict well neutron dynamical SF

Isostructural transition in elemental Cerium





Stationary Free Energy Functional



For numeric stability, the stationary DFT+DMFT functional $\delta\Gamma[G]/\delta G = 0$ is needed but the implementation is challenging.

$$\Gamma[G] = \operatorname{Tr} \log G - \operatorname{Tr}[(G_0^{-1} - G^{-1})G] + E^H[\rho] + E^{xc}[\rho] + \Phi^{\mathrm{DMFT}}[\hat{P}G] - \Phi^{\mathrm{DC}}[\hat{P}\rho] + E_{\mathrm{nuc-nuc}}$$
Can be computed from frequency

dependent band structure $\varepsilon_{\mathbf{k}\omega_n,i}$

Idea : Use the free energy F_{imp} of the auxiliary impurity problem:

 $F_{imp} = \operatorname{Tr} \log G_{imp} - \operatorname{Tr}(\Sigma_{imp}G_{imp}) + \Phi[G_{imp}]$

Very hard to compute

 $\Phi[G_{imp}] == \Phi[PG]$ and

 $J_{1/T}$

$$F_{solid} = F_{imp} + \operatorname{Tr}\log G - \operatorname{Tr}\log G_{imp} + E^H + E^{XC} + E_{nuc-nuc} - \Phi^{DC}[\{\hat{P}\rho\}]$$

For the impurity problem we can compute with CTQMC very precisely all necessary quantities

$$F_{imp} = \text{Tr}((\Delta + \varepsilon_{imp} - \omega_n \frac{d\Delta}{d\omega_n})G_{imp}) + E_{imp}^{potential} - T S_{imp}$$
$$S_{imp}(T) = S_{imp}(T_{>}) - \frac{E_{imp}(T_{>})}{T_{>}} + \frac{E_{imp}(T)}{T} - \int_{1/T_{>}}^{1/T} E_{imp}(1/\beta)d\beta$$

Details of the implementation in supplementary of PRL115, 256402 (2015).

Embedded Dynamical Mean Field

Stationary Embedded DMFT functional extremized in real space.

Free energy functional can be analytically differentiated to give forces on all atoms.

Can predict complex crystal structures

see : Phys. Rev. B 94, 195146 (2016).

Optimized Structure NdNiO₃



2-3 times better than the best of GGA or GGA+U.

Energetic of the Mott transition in NdNiO₃

- Paramagnetic Mott insulating state exists in NdNiO3 (we optimized it), but is metastable
- Magnetism stabilizes the insulating state, but is not driving the MIT. Fluctuating moments release their entropy by ordering and system gains some energy.



Force on all atoms from derivative of the functional







The Hellman-Feynman force

We need to differentiate the implemented expression for the free energy.

Free energy expression again

Recall: $\Gamma[\{G\}] = \operatorname{Tr} \log G - \operatorname{Tr}((G_0^{-1} - G^{-1})G) + E_{V_C}^{H+XC}[\rho] + \sum_{\mathbf{R}_i \in corr} \Phi_{V_{DMFT}}^{DMFT}[G_{loc}^i] - \Phi_{V_{DMFT}}^{DC}[\rho_{loc}^i]$

at the DMFT solution the Dyson Eq. is satisfied

$$G^{-1} = G_0^{-1} - V_{H+XC}(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') - \langle \mathbf{r} | \phi_{\alpha}^i \rangle (\Sigma^{imp} - V^{DC})_{\alpha\beta} \langle \phi_{\beta}^i | \mathbf{r}' \rangle$$

hence the free can be computed by

$$F = \operatorname{Tr}\log(G) - \operatorname{Tr}(V_{H+XC})\rho) + E^{H+XC}[\rho] + \sum_{\mathbf{R}_i} \left\{ -\operatorname{Tr}((\Sigma^{imp} - V^{DC})G^i_{loc}) + \Phi^{DMFT}[G^i_{loc}] - \Phi^{DC}[\rho^i_{loc}] \right\}$$

nuclear-nuclear energy is added; canonical ensemble needs +µ N

Free energy expression implementation

To implement $\operatorname{Tr}\log G$ we compute generalized Kohn-Sham orbitals:

$$\left\langle \psi_{j\mathbf{k}\omega_{n}} \right| \left(T + V_{nuc} + V_{H+XC} + \sum_{mm',\mathbf{R}_{\mu}} \left| \phi_{m}^{\mu} \right\rangle \left\langle \phi_{m}^{\mu} | \Sigma_{i\omega_{n}} - V_{DC} | \phi_{m'}^{\mu} \right\rangle \left\langle \phi_{m'}^{\mu} | \right) \left| \psi_{i\mathbf{k}\omega_{n}} \right\rangle = \delta_{ij} \varepsilon_{\mathbf{k}\omega_{n},i}$$

so that
$$\langle \psi_{j\mathbf{k}\omega_n}|G|\psi_{i\mathbf{k}\omega_n}\rangle = \frac{\delta_{ij}}{i\omega_n + \mu - \varepsilon_{\mathbf{k}\omega_n,i}}$$
, i.e., G is diagonalized

then

$$Tr \log(-G) = -Tr \log(-i\omega_n - \mu + \varepsilon_{\mathbf{k}\omega_n})$$
and

$$F = -\operatorname{Tr}\log(-i\omega_n - \mu + \varepsilon_{\mathbf{k}\omega_n}) - \operatorname{Tr}(V_{H+XC})\rho) + E^{H+XC}[\rho] + E_{nuc-nuc} + \mu N$$
$$+ \sum_{\mathbf{R}_i} \left\{ -\operatorname{Tr}((\Sigma - V_{DC})G^i_{loc}) + \Phi^{DMFT}[G^i_{loc}] - \Phi^{DC}[\rho^i_{loc}] \right\}$$

Derivative of the free energy

The free energy :



Derivative of the free energy



But: Functional is stationary, and $\delta\Sigma(\omega)$ cancel out.

K.H. & G.L. Pascut, Phys. Rev. B 49, 195146 (2016)

Derivative of the free energy

To prove that $\delta(\Sigma - V_{DC})$ cancels, we recall eigenvalue Eq.

$$\langle \psi_{i\mathbf{k}\omega_{n}} | \left(T + V_{nuc} + V_{H+XC} + \sum_{mm',\mathbf{R}_{\mu}} |\phi_{m}^{\mu}\rangle \langle \phi_{m}^{\mu}|\Sigma_{i\omega_{n}} - V_{DC}|\phi_{m'}^{\mu}\rangle \langle \phi_{m'}^{i}| \right) - \varepsilon_{\mathbf{k}\omega_{n},i} |\psi_{i\mathbf{k}\omega_{n}}\rangle = 0$$
which is satisfied for each atomic position \mathbf{R}_{μ} hence
$$\langle \psi_{i\mathbf{k}\omega_{n}} | \left(\delta(T + V_{nuc} + V_{H+XC}) + \sum_{mm',\mathbf{R}_{\mu}} \delta(|\phi_{m}^{\mu}\rangle \langle \phi_{m}^{\mu}|\Sigma_{i\omega_{n}} - V_{DC}|\phi_{m'}^{\mu}\rangle \langle \phi_{m'}^{i}|) \right) - \delta\varepsilon_{\mathbf{k}\omega_{n},i} |\psi_{i\mathbf{k}\omega_{n}}\rangle = 0$$
therefore
$$\delta\varepsilon_{\mathbf{k}\omega_{n},i} = \sum_{\mathbf{R}_{\mu},mm'} \langle \psi_{i\mathbf{k}\omega_{n}} | \phi_{m}^{\mu}\rangle \langle \phi_{m}^{\mu} | \delta(\Sigma - V_{DC}) | \phi_{m'}^{\mu}\rangle \langle \phi_{m'}^{\mu} | \psi_{i\mathbf{k}\omega_{n}}\rangle + \dots$$

....+derivative of the projector+DFT terms

hence

$$\operatorname{Tr}\left(\frac{\delta\varepsilon_{\mathbf{k}\omega_{n}}}{i\omega+\mu-\varepsilon_{\mathbf{k}\omega_{n}}}\right) = \operatorname{Tr}\left(|\psi_{i\mathbf{k}\omega_{n}}\rangle\frac{1}{i\omega+\mu-\varepsilon_{\mathbf{k}\omega_{n}}}\langle\psi_{i\mathbf{k}\omega_{n}}|\sum_{\mathbf{R}_{\mu},mm'}|\phi_{m}^{\mu}\rangle\langle\phi_{m}^{\mu}|\delta(\Sigma-V_{DC})|\phi_{m'}^{\mu}\rangle\langle\phi_{m'}^{\mu}|\right) + \dots$$

$$= \operatorname{Tr}\left(G\sum_{\mathbf{R}_{\mu},mm'}|\phi_{m}^{\mu}\rangle\langle\phi_{m}^{\mu}|\delta(\Sigma-V_{DC})|\phi_{m'}^{\mu}\rangle\langle\phi_{m'}^{\mu}|\right) + \dots$$

$$\mathbf{F}_{\mu}^{Puly} = -\operatorname{Tr}\left(\frac{1}{i\omega_{n}+\mu-\varepsilon_{\mathbf{k}\omega_{n}}}\frac{\delta\varepsilon_{\mathbf{k}\omega_{n}}}{d\mathbf{R}_{\mu}}\right) + \operatorname{Tr}\left(\rho\frac{\delta V_{KS}}{\delta\mathbf{R}_{\mu}}\right) + \operatorname{Tr}\left(G_{loc}\frac{\delta\Sigma-\delta V_{DC}}{\delta\mathbf{R}_{\mu}}\right) = \operatorname{Tr}\left(G_{loc}\delta(\Sigma-V_{DC})|\phi_{m'}^{\mu}\rangle\langle\phi_{m'}^{\mu}|\right) + \dots$$

Final results for forces in a mixed basis set



derivative of the DMFT projector, because the DMF basis moves with the atom

depends on the "non-trivial" DMFT density matrices

$$\widetilde{\rho} \equiv \frac{1}{\beta} \sum_{i\omega_n} B^R_{\omega_n} \frac{1}{i\omega_n + \mu - \varepsilon_{\mathbf{k}\omega_n}} B^L_{\omega_n}$$
$$\widetilde{(\rho\varepsilon)} \equiv \frac{1}{\beta} \sum_{i\omega_n} B^R_{\omega_n} \frac{\varepsilon_{\mathbf{k}\omega_n}}{i\omega_n + \mu - \varepsilon_{\mathbf{k}\omega_n}} B^L_{\omega_n}$$

Success: Forces do not depend on $\Phi[G]$ or $\delta\Sigma/\delta G = \delta^2\Phi/\delta G^2$

which are hard to compute.

K.H. & G.L. Pascut, Phys. Rev. B 49, 195146 (2016)

Forces more stable than free energies





 $\delta^2 \Phi[G] / \delta G^2$

hard to compute (needed for dynamical matrix)

Electronic structure package: EDMFTF



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Can be downloaded: http://hauleweb.rutgers.edu/tutorials

tutorials available

- Projection & Embedding instead of downloading in the full potential (APW+lo,LAPW) basis.
- Continuous time quantum Monte Carlo, OCA, NCA...
- Stationary implementation of free energy
- Forces on all atoms \bullet
- Structural optimization within DFT+DMFT

Database:

http://hauleweb.rutgers.edu/

<> hauleweb.rutgers.edu ITGERS Center for Computational Materials Theory School of Arts and Sciences

DFT + Embedded DMFT Functional^{*}

Developed by Kristjan Haule at Rutgers University, ©Copyright 2007-2016.

- What is DFT+Embedded DMFT Functional
- Installation
- Overview Tutorial on single band Hubbard model
- Tutorial 1 on MnO
- Tutorial 2 on FeSe, structural optimization, and spectral function plot
- Tutorial 3 on SrVO₃
- Tutorial 4 on LaVO₃
- Tutorial 5 on elemental Cerium
- Tutorial 6 on Sr₂IrO₄
- FAQ



Download the DMFT-Wien2K source code

dmft_w2k.tgz (version 2012)

dmft_w2k.tgz (version 2015)

Electronic structure package: EDMFTF

- Package needs wien2k for part of the DFT calculation.
- Composed of many Python, C++, Fortran90 executables linked by Python scripts.
- Two main Python executables:
 - *init_dmft.py* (initialization)
 - *run_dmft.py* (DFT+DMFT scheduler
- Several post processing tools, such as:
 - maxent_run.py
 - *x_dmft.py dmft1* (calculates DOS)
 - x_dmft.py dmft2 (recalculates electronic charge)
 - *x_dmft.py dmftp* (calculates A(k,w)
 - *wakplot.py* (displays A(k,w))
 - *dmftopt* (calculates optical conductivity or transport)

EXECUTION FLOW : DFT PART



FLOW: DFT+DMFT COMBINED



$$\sum_{\Sigma^{DMFT}(\omega), \varepsilon_{\mathbf{k},i}^{DFT}, \psi_{\mathbf{k},i}^{DFT} \to G^{DMFT}(\mathbf{r}), \Delta(\omega)}$$
input output
$$\Sigma(\omega), \varepsilon_{\mathbf{k},i}^{DFT}, \psi_{\mathbf{k},i}^{DFT}(\mathbf{r}) \longrightarrow G_{local}(\omega), \Delta(\omega), E_{imp}, n_{local}$$
1) Construct projector: $P(\mathbf{rr}', \mathbf{R}_{\mu}, mm') = \langle \mathbf{r} | \phi_{m}^{\mu} \rangle \langle \phi_{m'}^{\mu} | \mathbf{r}' \rangle$
where $\langle \mathbf{r} | \phi_{m}^{\mu} \rangle = u_{l}(|\mathbf{r} - \mathbf{R}_{\mu}|)Y_{lm}(\widehat{\mathbf{r} - \mathbf{R}_{\mu}})$
2) Embed self-energy: $\overline{\Sigma}_{ij}(\mathbf{k}, \omega) = \sum_{\mathbf{R}_{\mu}} \langle \psi_{\mathbf{k},i}^{DFT} | \phi_{m}^{\mu} \rangle (\Sigma_{mm'}^{\mu}(\omega) - V_{DC}^{\mu}) \langle \phi_{m'}^{\mu} | \psi_{\mathbf{k},j}^{DFT} \rangle$
3) Calculate local Green's function, hybridization, imp. levels:
$$G_{local,mm'}^{\mu} = \sum_{\mathbf{k},ij} \langle \phi_{m}^{\mu} | \psi_{\mathbf{k},i}^{DFT} \rangle (\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega))_{ij}^{-1} \langle \psi_{\mathbf{k},j}^{DFT} | \phi_{m'}^{\mu} \rangle = \left(\frac{1}{\omega - E_{imp}^{\mu} - \Sigma^{\mu}(\omega) - \Delta^{\mu}(\omega)}\right)_{mm'}$$
symmetrization over all group operations is performed

x_dmft.py dmft2 : $\Sigma^{DMFT}(\omega), \varepsilon^{DFT}_{\mathbf{k},i}, \psi^{DFT}_{\mathbf{k},i} \to \rho^{val}(\mathbf{r}), E_{valence}$ input output $\Sigma(\omega), \varepsilon_{\mathbf{k},i}^{DFT}, \psi_{\mathbf{k},i}^{DFT}(\mathbf{r}) \longrightarrow \rho_{val}^{DMFT}(\mathbf{r}), E_{valence}, F_{valence}, \mathbf{F}^{\mathbf{R}_{\mu}}$ 1) Construct projector: $P(\mathbf{rr}', \mathbf{R}_{\mu}, mm') = \langle \mathbf{r} | \phi_m^{\mu} \rangle \langle \phi_{m'}^{\mu} | \mathbf{r}' \rangle$ where $\langle \mathbf{r} | \phi_m^\mu \rangle = u_l (|\mathbf{r} - \mathbf{R}_\mu|) Y_{lm} (\mathbf{r} - \mathbf{R}_\mu)$ 2) Embed self-energy: $\bar{\Sigma}(\mathbf{r},\mathbf{r}') = \sum \langle \mathbf{r} | \phi_m^{\mu} \rangle \left(\Sigma_{mm'}^{\mu}(i\omega) - V_{DC}^{\mu} \right) \langle \phi_{m'}^{\mu} | \mathbf{r}' \rangle$ 3) Solve the Dyson Eq.: $(-\nabla^2 + V_{KS} + \bar{\Sigma}) |\psi_{\mathbf{k},i\omega_n,i}\rangle = |\psi_{\mathbf{k},i\omega_n,i}\rangle \varepsilon_{\mathbf{k},i\omega_n,i}^{DMFT}$ $\mathsf{Or} \quad (\varepsilon_{\mathbf{k},i_1}^{DFT} \delta_{i_1,i_2} + \langle \psi_{\mathbf{k},i_1}^{DFT} | \bar{\Sigma} | \psi_{\mathbf{k},i_2}^{DFT} \rangle) \, \langle \psi_{\mathbf{k},i_2}^{DFT} | \psi_{\mathbf{k},i\omega,i} \rangle = \langle \psi_{\mathbf{k},i_1}^{DFT} | \psi_{\mathbf{k},i\omega_n,i} \rangle \, \varepsilon_{\mathbf{k},i\omega_n,i}^{DMFT}$ 4) Determine the chemical potential: $N_{val} = T \sum_{i\omega = \mathbf{k}, i} \frac{1}{i\omega + \mu - \varepsilon_{\mathbf{k}, i\omega_n, i}}$ 5) Calculate DMFT electronic charge in space: $\rho_{val}^{DMFT}(\mathbf{r}) = \sum \psi_{\mathbf{k},i}^{DFT}(\mathbf{r}) T \sum \left[(i\omega + \mu - \varepsilon_{\mathbf{k}}^{DFT} - \bar{\Sigma}_{\mathbf{k}}(\omega))^{-1} \right]_{ij} \psi_{\mathbf{k},j}^{DFT^*}(\mathbf{r})$ 6) Calculate DMFT free energy and forces on all atoms symmetrization over group operations not performed

EDMFTF initialization: Exercise MnO

In Wien2k directory name has to be equal to struct file-name ("case"). In EDMFTF this is not needed anymore, hence we can create directory with any name.

Assume Wien2k run successfully finished in directory "case" = MnO. We will initialize EDMFTF in the same directory, because we will need energy files (for finding energy of bands).

> srun -n2 --pty bash # start interactive shell on compute node

> module load edmftf

(loads executables, paths, etc)

Two ways to initialize EDMFTF:

- 1) execute init_dmft.py and answer the questions.
- append all options in the command line as arguments to the init_dmft.py script. We will use method 2.

> cd MnO directory with Wien2k DFT run.

> init_dmft.py -ca l -ot d -qs 7

EDMFTF initialization



There are 2 atoms in the unit cell: 1 Mn

20

You have chosen the following atoms to be correlated: 1 Mn

For each atom, specify correlated orbital(s) (ex: d,f): You have chosen to apply correlations to the following orbitals: 1 Mn-1 d

Specify qsplit for each correlated orbital (default = 0): Qsplit Description

- 0 average GF, non-correlated
- 1 |j,mj> basis, no symmetry, except time reversal (-jz=jz)
- -1 |j,mj> basis, no symmetry, not even time reversal (-jz=jz)
- 2 real harmonics basis, no symmetry, except spin (up=dn)
- -2 real harmonics basis, no symmetry, not even spin (up=dn)
- 3 t2g orbitals
- -3 eg orbitals
- 4 |j,mj>, only I-1/2 and I+1/2
- 5 axial symmetry in real harmonics
- 6 hexagonal symmetry in real harmonics
- 7 cubic symmetry in real harmonics
- 8 axial symmetry in real harmonics, up different than down
- 9 hexagonal symmetry in real harmonics, up different than down
- 10 cubic symmetry in real harmonics, up different then down
- 11 |j,mj> basis, non-zero off diagonal elements
- 12 real harmonics, non-zero off diagonal elements
- 13 J_eff=1/2 basis for 5d ions, non-magnetic with symmetry
- 14 J_eff=1/2 basis for 5d ions, no symmetry

You have chosen the following qsplits:

1 Mn-1 d: 7

Atom 1 in MnO.struct is treated by DMFT

The "d" orbital is correlated by DMFT

"d" orbitals have cubic symmetry and we will use real harmonics (to avoid QMC sign problem)



Range (in eV) of hybridization taken into account in impurity problems; default -10.0, 10.0: -10.0,10.0

Very large 20eV window of bands is used to construct DMFT projector. *Rule of thumb : should be more than [-U,U].*

Perform calculation on real; or imaginary axis? (i/r): (default=i)

QMC works on the imaginary axis.

EDMFTF initialization

Which files were created by this initialization?

- projectorw.dat (contains the radial dependence of the orbital)
- MnO.indmfl (connects DMFT orbitals with the Kohn-Sham states)
- MnO.indmfi (connects DMFT orbitals with the impurity solver)

.indmfl file

connects DMFT orbitals with the Kohn-Sham states



.indmfl file

5 15 1 5# hybridization band index nemin and nemax, re1 0.025 0.025 200 -3.000000 1.000000 # matsubara, broadening-corr, bro1# number of correlated atoms11271# iatom, nL, locrot274L, qsplit, cix#====================================	enormalize for interstitials, projection type badening-noncorr, nomega, omega_min, omega_max (in eV) lated orbitals ====================================	
 1 5 2 # cix-num, dimension, num-independent-components # # independent components are 'eg' 't2g' # # Sigind follows 1 0 0 0 0 	correlated set (cix) if of dimension 5 and has two self-energies, i.e., eg and t2g	
0 1 0 0 0 0 0 2 0 0 0 0 0 2 0 0 0 0 0 2 # # Transformation matrix follows	How is self-energy matrix constructed from the two self-energy components	
$ \begin{array}{c} z^2 \\ x^2 - y^2 \\ xz \\ yz \\ xy \end{array} \begin{array}{c} 0.0000000 & 0.0000000 \\ 0.70710679 & 0.0000000 \\ 0.0000000 & 0.0000000 \\ 0.00000000 & 0.0000000 \\ 0.00000000 & 0.0000000 \\ 0.00000000 & 0.0000000 \\ 0.00000000 & 0.0000000 \\ 0.00000000 & 0.000000 \\ 0.00000000 & 0.000000 \\ 0.00000000 & 0.000000 \\ 0.00000000 & 0.000000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.0000 \\ 0.00000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.00000000 & 0.000 \\ 0.00000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.0000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.000000 & 0.000 \\ 0.00000 & 0.000 \\ 0.000000 & 0.000 \\ 0.00000 & 0.000 \\ 0.00000 & 0.000 \\ 0.00000 & 0.000 \\ 0.00000 & 0.000 \\ 0.00000 & 0.0000 \\ 0.00000 & 0.000 \\ 0.0000 & 0.000 \\ 0.0000 & 0.000 \\ 0.0000 & 0.000 \\ 0.0000 & 0.000 \\ 0.0000$	000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000 0.0000000	
L=2,m=-1 L=2,m=-1 Transformation matrix from	spheric harmonics to the orbitals used in the DMFT.	
$\begin{array}{c c} z \\ z \\ z \\ d_{z^2} \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{x^2-y^2} \end{array} \\ \begin{array}{c} z \\ d_{xz} \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} z \\ d_{yz} \end{array} \\ \end{array} $		
.indmfi file

connects DMFT orbitals with the impurity solver



EDMFTF initialization

Need two more files to start EDMFTF calculation:

1) params.dat (files with parameters — se below)

params.dat is a python file, which can use python expressions. Can be checked for syntactic correctness by "*python params.dat*"

> cp \$RESULTS/MnO/DMFT/params.dat .

2) sig.inp (guess for initial self-energy)

text file with columns corresponding to real and imaginary part of self-energy. first column is frequency, following by the real and imaginary part of Sigma for each orbital specified in *case.indmfi*



creates empty self-energy sig.inp with goods guess for Vdc and sigma(∞)

	paran	ns.dat	$run_lapw : \rho^{DFT}(\mathbf{r})$ $run_dmft.py = = (x \mid apw0 : V_{ext}(\mathbf{r}) \ \rho(\mathbf{r}) \rightarrow V_{KS}(\mathbf{r})$ $\times mixer: \ \rho^{val} + \rho^{core}, \rho^{old}(\mathbf{r}) \rightarrow \rho^{new}(\mathbf{r}) \qquad \times \ lapw1$ $\times core : V_{KS}(\mathbf{r}), V_{ext}(\mathbf{r}) \rightarrow \rho_{core}(\mathbf{r}), E_{core} \qquad \times \ lapws$: $V_{KS}(\mathbf{r}), V_{ext}(\mathbf{r}) \rightarrow \varepsilon_{\mathbf{k},i}^{DFT}, \psi_{\mathbf{k},i}^{DFT}$ \odot : adds spin-orbit
solver = 'CTQMC' DCs = 'exactd' max_dmft_iterations = ' max_lda_iterations = 1 finish = 10	 # impurity solv # exact double 1 # number of 00 # number of # number of 	e-counting with dielectric constant approx. iteration of the dmft-loop only f iteration of the LDA-loop enly f iterations of full charge loop (1 = no charge	$\begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \end{array} \end{array} \\ \begin{array}{c} \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \end{array} \\ \begin{array}{c} \end{array} \\ \end{array} $	$ \begin{array}{c} \begin{array}{c} \text{-dmft.py dmftl:} \\ , \varepsilon_{\mathbf{k},i}^{DFT}, \psi_{\mathbf{k},i}^{DTT} \rightarrow G^{DMFT}(\mathbf{r}), \Delta(\omega) \end{array} \end{array} $
ntail = 300 # 0 cc = 5e-5 # 1 ec = 5e-5 # 1 recomputeEF = 0	on imaginary ax the charge dens the energy prec # Recompute	s, number of points in the tail of the logarithn ity precision to stop the LDA+DMFT run sion to stop the LDA+DMFT run EF in dmft2 step. If recomputeEF = 0, it fixed	To stop the loop ne QMC precision (n d the chemical potential. Good for insulators	ed sufficient nany cores)
<pre># Impurity problem num iparams0={"exe" "U" "J" "CoulombF" "beta" "beta" "svd_Imax" "M" "mode" "nom" "tsample" "GlobalFlip" "warmup" "nf0" }</pre>	nber 0 : ["ctqmc" : [9.0 : [1.14 : ["'Ising" : [38.68 : [25 : [5e6 : ["SH" : [100 : [30 : [500000 : [1e5 : [5.0	 , "# Name of the executable"], , "# Coulomb repulsion (F0)"], , "# Coulomb repulsion (F0)"], , "# Form of Coulomb repulsion. 'Full' allow, , "# Form of Coulomb repulsion. 'Full' allow, , "# Inverse temperature T=116K"], , "# We will use SVD basis to expand G, w. , "# Total number of Monte Carlo steps"], , "# Total number of Monte Carlo steps"], , "# We will use self-energy sampling, and , "# Number of Matsubara frequency point, , "# How often to record measurements"], , "# How often to try a global flip"], , "# Warmup number of QMC steps"], , "# Nominal occupancy nd for double-could 	Careful: allowed on For metals recorn ws rotational invariant form of C.I."], with this cutoff"], d Hubbard I tail"], ts sampled"],	y for insulators. nputeEF=1.

params.dat continue



- U Hubbard U
- J Hund's interaction
- CoulombF form of the Coulomb interaction : Ising==density-density Slater form, Full==rotationally invariant Slater form.
 See for details : http://htttpi.ka/http://http://http://http://http://http://http://http://http
- beta 1/T[eV] or 11604/T[K]
- *M* number of MC-steps on single core. If it runs on more cores, better results & equal run-time.
- mode S==computing self-energy by Schwinger equation (rather than Dyson) H==high frequency computed by Hubbard-I form.
- nom number of Matsubara points keeps in the mesh (the rest in log mesh of ntail~100) (usually 3*beta)
- tsample every 30 MC steps we measure self-energy
- GlobalFlip is attempted every 5e5 steps
- warmup first 1e5 steps are not used to meassure
- nf0 Mn valence, used for double-counting. Here only for the initial guess (from DFT), will use exactDC later.

Final steps in initialization

Increase number of k-points for more precise DMFT frequency dependence.

> x kgen -f MnO 2000

Optional : Rerun LDA with new k-mesh.

> run lapw -NI

Optional : Create new directory with only necessary files for DFT+DMFT

> mkdir LDA; mv * LDA/

> mkdir DMFT; cd DMFT

> dmft_copy.py ../LDA

basic parameters params.dat KS-lattice to DMFT connection MnO.indmfl MnO.indmfi impurity to DMFT connection projectorw.dat radial dependent projector sig.inp self-energy MnO.struct crystal structure MnO.clmsum charge density MnO.in0 input to lapw0 MnO.in1 input to lapw1 W2k files MnO.in2 input to lapw2 MnO.inm input to mixer MnO.klist list of k-points MnO.scf2 input to lapw2/dmft2

- DMFT files, which we already discussed

calculation

Final steps in initialization

For parallel execution we need to create special file "mpi_prefix.dat"

> echo "mpirun -np 2 -x OMP_NUM_THREADS=I" > mpi_prefix.dat

- If this file does not exists, the run will be serial.
- np specified number of available cores (unfortunately only 2 can be afforded here)
- OMP_NUM_THREADS=1 switches off open_mp when mpi is used. If a lot of cores are available, one can use combination of open_MP and MPI.
- mpi_prefix.dat is used for the ctqmc impurity solver. If dmft1, dmft2, lapw1, lapwso require different parallelization, one can specify also mpi_prefix.dat2.

It appears that multithreading makes code slower on this machine, so please turn it off.

Finally run EMDFTF code by executing

> run_dmft.py >& nohup.dat

> export OMP_NUM_THREADS=I

Monitor the run

> less info.iterate	check the summary file		
<pre>> less dmft_info.out</pre>	see what/how is executed		
> less ':log'	check execution log		
> less MnO.dayfile	execution log+convergence		
> grep ':CHARGE' MnO.dayfile	charge converged by iterations		
> plot -u1:3,1:5 -x:10 MnO.dlt1	plot hybridization function		
> plot -x:20 -g -u1:3,1:5 imp.0/Gf.out.?.1	plot the DMFT output G		
> plot -x:20 -g -u1:3,1:5 imp.0/Sig.out	plot the impurity output Sigma		
>plot -g -u1:9,1:10 info.iterate	plot lattice & imp. occupancy		

> less info.iterate

summary file



> less dmft_info.out

details of every step and its execution (a lot of information)

iparams0 = {'nom': [100, '# Number of Matsubara frequency points sampled'], 'tsample': [30, '# How often to record measurements'], 'Ncout': [500000, '# How often to print out info'], 'J': [1.14, '# Coulomb J'], 'M': [500000.0, '# Total number of Monte Carlo steps per core'], 'CoulombF': ["'Ising'", "# Density-density form. Can be changed to 'Full' "], 'beta': [38.68, '# Inverse temperature'], 'U': [9.0, '# Coulomb repulsion (F0)'], 'GlobalFlip': [500000, '# How often to try a global flip'], 'sderiv': [0.02, '# Maximum derivative mismatch accepted for tail concatenation'], 'Nmax': [500, '# Maximum perturbation order allowed'], 'exe': ['ctqmc', '# Name of the executable'], 'warmup': [500000.0, '# Warmup number of QMC steps'], 'OCA_G': [False, "# Don't compute OCA diagrams for speed"], 'aom': [3, '# Number of frequency points used to determin the value of sigma at nom']}

DCs = exactd

```
Znucs= {1: 25}
iSiginds= {0: [[1, 0, 0, 0, 0], [0, 1, 0, 0, 0], [0, 0, 2, 0, 0], [0, 0, 0, 2, 0], [0, 0, 0, 0, 2]]}
#<lapw0>: wien2k_14/x -p -f MnO lapw0
#<lapw1>: x_dmft.py -p lapw1
DmftEnvironment: mpi_prefix.dat exists -- running in parallel mode.
 mpirun -np 2
..... running: mpirun -np 2 ./lapw1 lapw1.def
case=MnO, nom=100, ntail=300, insig=sig.inp, outsig=sig.inp[1-n]
s_00= [38.22, 38.22]
Edc= [38.22, 38.22]
..... Creating logarithmic mesh
..... Going over all correlated blocks
icix= 1 colsp= [1, 2] colsm= []
icl = 1
icl = 2
Running ---- dmft1 ----
#<dmft1>: /usr/bin/time mpirun -np 2 ./dmft dmft1.def >> dmft1_info.out
#<sjoin>: sjoin.py -m 1.0
Running ----- impurity solver -----
Taking care of high frequency
Eimp= [-38.16593522 -38.26510458]
Edc= [ 38.22 38.22]
ncorr= 5.01358294333
#<sgather>: sgather.py
----- DMFT part of the step # 0 done ------
case=MnO, nom=100, ntail=300, insig=sig.inp, outsig=sig.inp[1-n]
s_oo= [38.26987655713145, 38.35976476628431]
Edc= [37.425774429186845, 37.431910457048744]
..... Creating logarithmic mesh
----- Preparing Charge calculation ------
#<prep-dmft2>: x_dmft.py -d --mode c -m 0 -x 1.0 -w 1.0 -p dmft2 >> ksum_info.out 2>&1
#<ssplit>: ssplit.py -n 100 -t 300
#<dmft2>: mpirun -np 2 ./dmft2 dmft2.def >> dmft2_info.out
#<lcore>: x -f MnO lcore
#<mixer>: x -f MnO mixer
----- LDA(charge) step 0 0 done ------
#<lapw0>: x -p -f MnO lapw0
#<lapw1>: x_dmft.py -p lapw1
..... running: mpirun -np 2 ./lapw1 lapw1.def
----- Preparing Charge calculation ------
#cprep-dmft2>: x dmft.py -d --mode c -m 0 -x 1.0 -w 1.0 -p dmft2 >> ksum info.out 2>&1
#<ssplit>: ssplit.py -n 100 -t 300
#<dmft2>: mpirun -np 2 ./dmft2 dmft2.def >> dmft2_info.out
#<lcore>: x -f MnO lcore
#<mixer>: x -f MnO mixer
----- LDA(charge) step 1 0 done ------
```

which steps have finished when?

> less :log

Wed May 23 23:56:53 EDT 2018> (x) -f MnO lapw0 Wed May 23 23:56:54 EDT 2018> lapw1 Wed May 23 23:56:55 EDT 2018> dmft1 Wed May 23 23:56:56 EDT 2018> impurity Wed May 23 23:59:38 EDT 2018> dmft2 Wed May 23 23:59:39 EDT 2018> (x) -f MnO Icore Wed May 23 23:59:39 EDT 2018> (x) -f MnO mixer Wed May 23 23:59:39 EDT 2018> (x) -f MnO lapw0 Wed May 23 23:59:40 EDT 2018> lapw1 Wed May 23 23:59:41 EDT 2018> dmft2 Wed May 23 23:59:42 EDT 2018> (x) -f MnO Icore Wed May 23 23:59:42 EDT 2018> (x) -f MnO mixer Wed May 23 23:59:42 EDT 2018> (x) -f MnO lapw0 Wed May 23 23:59:43 EDT 2018> lapw1 Wed May 23 23:59:44 EDT 2018> dmft2 Wed May 23 23:59:44 EDT 2018> (x) -f MnO Icore Wed May 23 23:59:44 EDT 2018> (x) -f MnO mixer Wed May 23 23:59:44 EDT 2018> (x) -f MnO lapw0 Wed May 23 23:59:46 EDT 2018> lapw1

> less MnO.dayfile

Wed May 23 23:56:53 2018 30/30 to go cycle 0 (23:56:53)>lapw0 >lapw1 (23:56:54)>dmft1 (23:56:55) >impurity (23:56:56) (23:59:38)>dmft2 (23:59:39)>lcore (23:59:39)>mixer :ENERGY convergence: 0.00275430000011 :CHARGE convergence: 0.269067 :EF convergence: 0

Some output files





Some output files

Check difference between impurity and lattice occupancy

> plot -g -ul:9,l:10 info.iterate

Check convergence of electronic charge

> grep ':CHARGE' MnO.dayfile

Check what impurity solver is doing

>less imp.0/nohup_imp.out.000



:CHARGE	convergence:	1.0
:CHARGE	convergence:	0.272381
:CHARGE	convergence:	0.208401
:CHARGE	convergence:	0.094013
:CHARGE	convergence:	0.075578
:CHARGE	convergence:	0.071506
:CHARGE	convergence:	0.014747
:CHARGE	convergence:	0.005258
:CHARGE	convergence:	0.001651
:CHARGE	convergence:	0.00165
:CHARGE	convergence:	0.00068
:CHARGE	convergence:	0.00045
:CHARGE	convergence:	0.000309
:CHARGE	convergence:	0.000188
:CHARGE	convergence:	0.000108
:CHARGE	convergence:	6.2e-05
:CHARGE	convergence:	2e-05
:CHARGE	convergence:	0.085869
:CHARGE	convergence:	0.075714
:CHARGE	convergence:	0.055726
:CHARGE	convergence:	0.026821
:CHARGE	convergence:	0.014785
:CHARGE	convergence:	0.010197
:CHARGE	convergence:	0.001906
:CHARGE	convergence:	0.001064
:CHARGE	convergence:	0.000376
:CHARGE	convergence:	0.000176
:CHARGE	convergence:	2.1e-05

Postprocessing : maxent

> mkdir maxent; cd maxent

create new directory for analytic continuation

> cp ../sig.inp.10.1 .

copy the latest self-energy

alternatively, take average over previous run:

saverage.py \$RESULTS/MnO/sig.inp.1?.1

> cp \$RESULTS/MnO/maxent/maxent_params.dat .

need several parameters

params={'statistics': 'fermi',	# fermi/bose
'Ntau' : 300,	# Number of time points
'L' : 20.0,	# cutoff frequency on real axis
'x0' : 0.005,	# low energy cut-off
'bwdth' : 0.004,	# smoothing width
'Nw' : 300,	# number of frequency points on real axis
'gwidth' : 2*15.0,	# width of gaussian
'idg' : 1,	# error scheme: idg=1 -> sigma=deltag ; idg=0 -> sigma=deltag*G(tau)
'deltag' : 0.004,	# error
'Asteps' : 4000,	# anealing steps
'alpha0' : 1000,	# starting alpha
'min_ratio' : 0.001,	# condition to finish, what should be the ratio
'iflat' : 1,	# iflat=0 : constant model, iflat=1 : gaussian of width gwidth, iflat=2 : input using file model.dat
'Nitt' : 1000,	# maximum number of outside iterations
'Nr' : 0,	# number of smoothing runs
'Nf' : 40,	# to perform inverse Fourier, high frequency limit is computed from the last Nf points
}	

> maxent_run.py sig.inp.10.1

run maximum entropy

Postprocessing : maxent



Check the resulting self-energy on real axis

> plot -g -u1:3,1:5 -x-20:20 Sig.out

the gap

Postprocessing : Density of states

> mkdir ../onreal; cd ../onreal

> dmft_copy.py ../

Create another directory for real axis DOS

copy converged results on imaginary axis

If you do not have your own results, use: dmft_copy.py \$RESULTS/MnO

>cp ../maxent/Sig.out sig.inp

copy maxent real-axis self-energy to this directory

edit the second line of MnO.indmfl file and change the flag matsubara to 0

5 15 1 5	# hybridization band index nemin and nemax, renormalize for interstitials, projection type
10.025 0.025 200) -3.000000 1.000000 # matsubara, broadening-corr, broadening-noncorr, nomega, omega_min,
omega_max (in e'	$\vee)$
1	# number of correlated atoms
1 1 0	# iatom, nL, locrot
2 7 1	# L, qsplit, cix
changed file:	
5 15 1 5	# hybridization band index nemin and nemax, renormalize for interstitials, projection type
00.025 0.025 200) -3.000000 1.000000 # matsubara, broadening-corr, broadening-noncorr, nomega, omega_min,
omega_max (in e	$\vee)$
1	# number of correlated atoms
1 1 0	# iatom, nL, locrot
2 7 1	# L, qsplit, cix

Postprocessing : Density of states



Check with Experiment



Should be careful with direct comparison(surface issues, matrix elements effects, low resolution for inverse Ph). Nevertheless, the agreement good, and far superior than LDA+U or GW.



Postprocessing : A(k,w)

> cp \$RESULTS/MnO/onreal/MnO.klist_band .

copy k-list path can be created by xcrysden

> x_dmft.py lapw1 --band

> x dmft.py dmftp

recomputed KS bands on the current k-path.

edit the second line of **MnO.indmfl** file and change **omega_min, omega_max** to -6 6 . We use 200 frequency points

5 15 1 5	# hybridization band index nemin and nemax, renormalize for interstitials, projection type
0 0.025 0.025 200	-3.000000 1.000000 # matsubara, broadening-corr, broadening-noncorr, nomega, omega_min,
omega_max (in e\	/)
1	# number of correlated atoms
1 1 0	# iatom, nL, locrot
2 7 1	# L, qsplit, cix
changed file:	
5 15 1 5	# hybridization band index nemin and nemax, renormalize for interstitials, projection type
0 0.025 0.025 200	-6.000000 6.000000 # matsubara, broadening-corr, broadening-noncorr, nomega, omega_min,
omega_max (in e	/)
1	# number of correlated atoms
1 1 0	# iatom, nL, locrot
2 7 1	# L, qsplit, cix

computed and store DMFT bands (eigvals.dat) on the same k-path

We did not change EF during self-consistent run, therefore "EF.dat" file was not yet created. For metals "EF.dat" should exist already. Check "mu" from **info.iterate** and copy it into "EF.dat"

> echo 6.530817 > EF.dat



Exercise 2: FeSe example (iron superconductor)



Add "params.dat" and "sig.inp"

> cp \$RESULTS/FeSe/DMFT/params.dat .

	solver = 'CTQMC' # impurity solver DCs = 'nominal' # double counting scheme max_dmft_iterations = 1 # number of iteration of the dmft-loop only max_lda_iterations = 100 # number of iteration of the LDA-loop only finish = 30 # number of iterations of full charge loop (1 = no charge self-consistency)
metal requires to compute EF at each iteration	 ntail = 300 # on imaginary axis, number of points in the tail of the logarithmic mesh cc = 2e-5 # the charge density precision to stop the LDA+DMFT run recomputeEF = 1 # Recompute EF in dmft2 step. If recomputeEF = 2, it tries to find an insulating gap.
will start with a good guess for sigma	GoodGuess = True # We have a good guess for self-energy, and the scheduler optimizes run for good existing self-energy # Impurity problem number 0 iparams0={"exe" : ["ctqmc" , "# Name of the executable"], "U" : [5.0 , "# Coulomb repulsion (F0)"], "J" : [0.8 , "# Coulomb repulsion (F0)"],
use rotationally invariant Coulomb U	 "CoulombF": [""Full", "# Can be set to 'Full"], "beta": [50, "# Inverse temperature"], "svd_Imax": [25, "# We will use SVD basis to expand G, with this cutoff"], "M": [10e6, "# Total number of Monte Carlo steps"], "Mlist": [[10e6]*5 + [20e6], "# Changing M"], "mode": ["SH", "# We will use self-energy sampling, and Hubbard I tail"],
number of MC steps will change. start with 10M, and later 20M	"nom": [200, "# Number of Matsubara frequency points sampled"],"tsample": [100, "# How often to record measurements"],"GlobalFlip": [1000000, "# How often to try a global flip"],"warmup": [3e5, "# Warmup number of QMC steps"],"nf0": [6.0, "# Nominal occupancy nd for double-counting"],}

> cp \$RESULTS/FeSe/DMFT/sig.inp .

copy a good guess for self-energy to achieve faster convergence.

Prepare & execute

> x kgen -f FeSe 1000

Do you want to shift k-mesh: I

- > run_lapw -NI
- > mkdir LDA; mv * LDA/
- > mkdir DMFT; cd DMFT
- > dmft_copy.py ../LDA

Optional: move LDA data to directory LDA and start new calculation in directory DMFT. Copy necessary data to new directory by *dmft_copy.py* script.

> echo "mpirun -np 2 -x OMP_NUM_THREADS=I" > mpi_prefix.dat

> run_dmft.py >& nohup.dat

Monitor the run

> less info.iterate	check the summary file
<pre>> less dmft_info.out</pre>	see what/how is executed
<pre>> less imp.0/nohup_imp.out.000</pre>	check what impurity is doing
> less FeSe.dayfile	execution log+convergence
> plot -g -ul:9,1:10 info.iterate	impurity occupancy convergence

Note: due to a flag "GoodGuess=True" in params file, the calculation started with converging electronic charge on the currently provided self-energy (sig.inp), and impurity solver is run only after the charge is converged.

> plot -x:10 -g -u1:3,1:5,1:7,1:9,1:11 imp.0/Sig.out.?.1 plot impurity self-energy

Checking the amount of force on atoms

We can switch on the calculation of force on atoms by changing a variable in *FeSe.in2*

currently FeSe.in2 line1 contains:

```
# FeSe.in2, line 1 :
TOT (TOT,FOR,QTL,EFG,FERMI)
Change to :
# FeSe.in2, line 1 :
FOR (TOT,FOR,QTL,EFG,FERMI)
```

After this change, *FeSe.scf* should contain a printout of the total force, for example:

	TOTAL FORC	E IN mRy/a.u. =	= F Fx	Fy	Fz	with/without	FOR in case.in2
:FOR001	: 1.ATOM	0.00000	0.00000	0.000000	0.00000	total forces	
:FOR002	2: 2.ATOM	19.938644	0.00000	0.000000	-1.226529	total forces	
	TOTAL FORC	E WITH RESPECT	TO GLOBAL CART	ESIAN COORDINATES:			
:FCA001	: 1.ATOM		0.0000	0.000000	0.00000	total forces	
:FCA002	2: 2.ATOM		0.0000	0.000000	-1.226529	total forces	
	TOTAL FORC	E WITH RESPECT	TO THE GLOBAL	COORDINATE SYSTEM:			
:FGL001	: 1.ATOM		0.000000000	0.000000000	0.000000000	total forces	
:FGL002	2: 2.ATOM		0.000000000	0.00000000	-1.226529	total forces	

This format is identical to the format in Wien2k, and it says that the Se atom has a force in z-direction of -1.22mRy/a.u. (for different RKmax, it might be somewhat different value). We can grep the force at each step by

grep ':FGL002' FeSe.scf

The DFT equilibrium value is **z=0.245** DMFT equilibrium value : **z=0.268** Experiment : **z=0.265** Note: force of a few mRy/a.u. is very small and says that the structure is already optimized. This is because zposition of Se z=0.27 in this structure file is very close to its equilibrium in DMFT (and experiment). The LDA equilibrium value is z=0.245, which would create very large force in DMFT.

If you wish to optimize the structure (which is not needed in this case), you just need to edit *FeSe.inm* file and change MSR1 in the first line to MSR1a, i.e.,

FeSe.inm, line 1: MSR1a 0.0 YES (BROYD/PRATT, extra charge (+1 for additional e), norm)

Postprocessing : maxent + dos

Maximum entropy steps are identical as in previous MnO example. The same maxent_params.dat file should be good.

- > mkdir maxent; cd maxent
- > cp ../sig.inp .

> cp \$RESULTS/MnO/DMFT/maxent/maxent_params.dat .

- > maxent_run.py sig.inp
- > mkdir ../onreal; cd ../onreal
- > dmft_copy.py ../
- >cp ../maxent/Sig.out sig.inp

edit the second line of FeSe.indmfl file and change the flag matsubara to 0

- > cp ../mpi_prefix.dat .
- > x lapw0 -f FeSe
- > x_dmft.py lapw l
- > x_dmft.py dmft l



> x_dmft.py lapw1 --band

edit the second line of **FeSe.indmfl** file and change **omega_min, omega_max** to -1 1. We use 200 frequency points

> x_dmft.py dmftp

> wakplot.py 0.5

should give the following spectra:



Thank you!

Why DOES I.P.A. EVER work?

 Due to Pauli exclusion principle, the fermions tend to avoid each other, and single particle methods take into account fermi statistics through use of the Slater determinants. (Small phase space for scattering)

2) Conservation laws are unchanged by the interaction and suggests continuity of the noninteracting picture.

Example: Bloch theorem for single-particle states leads to the concept of bands.

$$H(u_{\mathbf{k}}(\mathbf{r}+\mathbf{R})e^{i\mathbf{k}\mathbf{r}}) = \varepsilon_{\mathbf{k}}(u_{\mathbf{k}}(\mathbf{r}+\mathbf{R})e^{i\mathbf{k}\mathbf{r}})$$

3) Fermi liquid theory: there should be a one-to-one correspondence between the excitations of the non-interacting system and the low energy quasiparticle excitations if continuity is satisfied [no intervening phase transition, like metal-insulator transition]

4) The "Luttinger theorem" states that, so long as the actual interacting system can be considered to evolve continuously from a non-interacting system, the volume enclosed by the Fermi surface does not change.

5) Screening is strong in simple metals, so that effective onsite Coulomb repulsion is only of the order of a few eV in Cu or Fe. But is of the order of 10eV in transition metal oxides.