Dynamical mean-field theory: (some) theory and (some) algorithm

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Dynamical mean-field theory (DMFT)

- Spectroscopic information is encoded in the one-particle Green’s function $G(z)$

- Quantum embedding ("Domain decomposition" in high dimensional space)

- Solve a (high dimensional) strongly correlated system by a series of (low dimensional) impurity problems

- “Glue” impurity problems together with single-particle type theories.

[Kananenka, Gull, Zgid, PRB 2015] …
DMFT applications: strongly correlated systems

- Electronic structure near a Mott transition

- Lattice dynamics and structural stability

- Optical properties

- Transport properties, and superconductors

- Heavy elements: Actinides and lanthanides

- ...
Algorithm

Hybridization fitting via semi-definite relaxation

[Majuto-Zaera, Zepeda-Núñez, Lindsey, Tubman, Whaley, L., 1907.07191]
How to solve the impurity problem?

\[(h^0)^{(p)}, \Delta^{(p)}(z) \rightarrow G_{\text{imp}}^{(p)}\]

- Open quantum system: \( z = i\omega \), Path integral. Quantum Monte Carlo (e.g. [Gull, Millis et al, RMP, 2011])

- Closed quantum system: Hamiltonian-based (e.g. [Zgid, Chan, J. Chem. Phys. 2011])
  - Allow broader range of impurity solvers (CI, CC, Selected CI..)
  - Hybridization fitting
Hamiltonian-based DMFT

- Hybridization fitting

\[
\Delta(i\omega) \approx h_{12}(i\omega - h_{22})^{-1}h_{21} = \sum_{\ell=1}^{N_p} \frac{V_{\ell} V_{\ell}^*}{i\omega - \lambda_{\ell}}
\]

- Replace a large (and possibly infinite) sized bath by a smaller one.

- Closed Hamiltonian system

\[
\hat{H}_{\text{imp}} = \hat{H}_{\text{cluster}} + \sum_{\ell=1}^{N_p} \lambda_{\ell} \hat{d}_{\ell}^\dagger \hat{d}_{\ell} + \sum_{\ell=1}^{N_p} \sum_{\alpha=1}^{N_c} (V_{\alpha,\ell} \hat{d}_{\ell}^\dagger \hat{a}_\alpha + \text{h.c.})
\]
Hybridization fitting

\[ \Delta(i\omega) \approx \sum_{\ell=1}^{N_p} \frac{V_\ell V_\ell^*}{i\omega - \lambda_\ell} \]

- Fitting problem: highly nonlinear optimization
- Surprisingly time consuming
- \( \Delta(z) \): Meromorphic matrix-valued function in \( \mathbb{C} \) with real poles
- Positive semi-definite (PSD) residue matrix
Semidefinite relaxation

\[ \Delta(i\omega) \approx \sum_{\ell=1}^{N_p} \frac{X_{\ell}}{i\omega - \lambda_{\ell}}, \quad X_{\ell} \geq 0 \]

- With a possibly smaller \( N_p \)
- Relaxed minimization problem

\[
\min_{\lambda_{\ell}, X_{\ell} \geq 0} J_{SDR}(\{\lambda_{\ell}, X_{\ell}\}_{\ell=1}^{N_p})
\]

\[
J_{SDR}(\{\lambda_{\ell}, X_{\ell}\}_{\ell=1}^{N_p}) = \frac{1}{N_\omega} \sqrt{\sum_{n=1}^{N_\omega} \left\| \Delta(i\omega_n) - \sum_{\ell=1}^{N_p} \frac{X_{\ell}}{i\omega_n - \lambda_{\ell}} \right\|_F^2}
\]
Semidefinite relaxation

- After obtaining the minimizers

\[ \hat{X}_\ell = U_\ell U_\ell^* = \sum_{q=1}^{N_c} U_{\ell,q} U_{\ell,q}^* \]

- Expansion

\[ \sum_{\ell=1}^{N_p} \frac{\hat{X}_\ell}{i \omega - \hat{\lambda}_\ell} = \sum_{\ell=1}^{N_p} \sum_{q=1}^{N_c} \frac{U_{\ell,q} U_{\ell,q}^*}{i \omega - \hat{\lambda}_\ell} = \sum_{r=1}^{N_{\text{eff}}} \frac{\tilde{V}_r \tilde{V}_r^*}{i \omega - \tilde{\epsilon}_r} \]
Optimization for the poles

- Nested optimization

\[ J_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}) = \min_{X_\ell \geq 0} J_{\text{SDR}}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p}) \]

- Derivative evaluation

\[ \frac{\partial}{\partial \lambda_k} J_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}) = \left. \frac{\partial}{\partial \lambda_k} \left( \sqrt{\sum_{j=1}^{N_\omega} \left\| \Delta(i\omega_j) - \sum_{\ell=1}^{N_p} \frac{X_\ell}{i\omega_j - \lambda_\ell} \right\|^2_F} \right) \right|_{X_\ell = X'_\ell} \]

\[ X'_\ell = \arg \min_{X_\ell \geq 0} J_{\text{SDR}}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p}) \]
Pseudocode

**Algorithm 1** Pseudo-code for the nested optimization routine.

**Require:** Initial guess \( \{\lambda_\ell\}_{\ell=1}^{N_p} \)

**Ensure:** \( \{\hat{\lambda}_\ell, \hat{X}_\ell\}_{\ell=1}^{N_p} \)

\[
\begin{align*}
\textbf{while} & \quad \| \nabla J_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}) \| > \epsilon_{\text{tol}} \textbf{ do} \\
& \quad X'_\ell \leftarrow \arg\min_{X_\ell \geq 0} J_{\text{SDR}}(\{\hat{\lambda}_\ell, X_\ell\}_{\ell=1}^{N_p}) \text{ by solving the SDR problem} \\
& \quad \lambda_\ell \leftarrow \text{BFGS}(\lambda_\ell, \nabla J_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p})) \\
\textbf{end while} \\
& \hat{\lambda}_\ell \leftarrow \lambda_\ell \\
& \hat{X}_\ell \leftarrow \arg\min_{X_\ell \geq 0} J_{\text{SDR}}(\{\hat{\lambda}_\ell, X_\ell\}_{\ell=1}^{N_p})
\end{align*}
\]
Results

• 2D Hubbard model.

• Impurity size (to be solved by the impurity solver)

• Impurity solver: adaptive sampling configuration algorithm (ASCI)

• Can use other impurity solvers as well, e.g. coordinate descent

[Tubman, Lee, Takeshita, Head-Gordon, Whaley, JCP, 2016]
[Mejuto-Zaera, Tubman, Whaley, 2017] [Wang, Li, Lu, 2019]
### Accuracy

- **Fitting error**

<table>
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<tr>
<th>$N_p$</th>
<th>$N_b^{\text{eff}}$</th>
<th>Error</th>
<th>$N_b^{\text{eff}}$</th>
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(a) Diagonal component of $\Delta$.

(b) Off-diagonal component of $\Delta$. 

Efficiency: Timings for fitting

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<th>$N_p$</th>
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BOBYQA (Bound Optimization BY Quadratic Approximation) [Powell,2009]
Derivative-free method.
Gradient based method easily trapped in local minima (empirical evidence)
Robustness: fit error vs. initial pole energies

\( N_p = 2 \)

\[
\begin{array}{|c|c|c|}
\hline
\text{Nr. of samples} & 1184 & 1195 & 1469 \\
\text{Rate of Success} & 83\% & 88\% (43\% + 45\%) & 55\% \\
\text{max}[\sigma(\epsilon_\ell)] & 6e-6 & 5e-4 & 2e-4 \\
\hline
\end{array}
\]

\( N_p = 3 \)

\( N_p = 4 \)

Initial poles randomly distributed in \([−10,10]^{⊗N_p}\).
Performance in DMFT

- 2D Hubbard, half filling, $\frac{U}{t} = 8$
- $2 \times 3, 2 \times 4, 1 \times 6$ cluster size, $N_p = 2, 3, 4, 6$
- Max fitting frequency $i(0,2)t$

\[ A(k, \omega) = -\frac{1}{\pi} \text{Im}(G_{\text{latt}}(k, \omega)) \]
Theory

Self energy sparsity
Well-posedness of the SCF loop (causality)
Luttinger-Ward formalism

[L., Lindsey, 1902.04796]
[L., Lindsey, Schneider, in preparation]
[L., Lindsey, 1809.02900, 1809.02901, PNAS 2018]
Example of quantum impurity problem

• Single-impurity Anderson model (SIAM)

\[ \hat{H} = \sum_\sigma \epsilon_f \hat{f}_\sigma^\dagger \hat{f}_\sigma + \sum_{\langle j,j' \rangle_\sigma} t_{jj'} \hat{c}_{j\sigma}^\dagger \hat{c}_{j'\sigma} + \sum_{j,\sigma} \left( V_j \hat{f}_\sigma^\dagger \hat{c}_{j\sigma} + V_j^* \hat{c}_{j\sigma}^\dagger \hat{f}_\sigma \right) + U \hat{f}_\uparrow^\dagger \hat{f}_\uparrow \hat{f}_\downarrow^\dagger \hat{f}_\downarrow \]

• Hamiltonian

• Self energy is a sparse matrix

\[ \Sigma(Z) = \begin{pmatrix} 0 & \cdots & \sigma(Z) & \cdots \\ \vdots & \ddots & \vdots & \ddots \\ \sigma(Z) & \cdots & 0 \end{pmatrix} \]
Sparisity of self energy for quantum impurity

Theorem ([L.-Lindsey, 1902.04796])

For the quantum impurity problem, the sparsity pattern of the self energy is only determined by the sparsity pattern of the interaction. W.L.O.G.

\[ \Sigma(z) = \begin{pmatrix} \Sigma_{\text{imp}}(z) & 0 \\ 0 & 0 \end{pmatrix}. \]

- Foundation of DMFT / CT-QMC etc
- “Folk theorem” since [Feynman-Vernon, 1963] at the latest with diagrammatic arguments
- Our proof is linear algebraic (non-perturbative)
- Extensible to classical impurity problems, fermionic and bosonic systems, zero and finite temperature, non-equilibrium systems, impurity problems with broken particle-number symmetry (related to superconducting systems)
Causality

Lehmann representation (retarded Green’s function)

\[
G_{ij}^R(z) = \sum_n \frac{f_{i,n} f_{j,n}^*}{z - (E_{N+1}^n - E_N^0) + i\eta} + \sum_n \frac{g_{i,n} g_{j,n}^*}{z + (E_{N-1}^n - E_N^0) + i\eta}
\]

\[
\epsilon_n = E_N^0 - E_{N-1}^n
\]

\[
\mu = E_{N+1}^n - E_N^0
\]

\[
\text{gap}
\]

\[
\text{Re}
\]
Well-posedness of the DMFT loop

Causal

1. Analytic in the upper half of the complex plane
2. Spectral density $-\text{Im} \left(G^R(z)\right)$ is PSD (implying that diagonal is non-negative).

- Whether $G^R, \Sigma^R, \Delta^R$ during the SCF loop remains causal.

- For imaginary frequency fitting:
  - Real poles
  - Residue matrix PSD

[Kotliar, Savrasov, Pálsson, Biroli, 2001]
[Biroli, Parcollet, Kotliar, 2003]
Causality

• Spectral density: Discrete PSD operator valued measure on \( \mathbb{R} \) (for some finite \( K \))

\[
\sum_{\ell=1}^{K} X_\ell \delta(\cdot - \lambda_\ell)
\]

• Stieltjes transform

\[
S := \left\{ f : \mathbb{C} \to \mathbb{C}^{N_b \times N_b} \left| f(z) = \sum_{\ell=1}^{K} \frac{X_\ell}{z - \lambda_\ell}, X_\ell \geq 0, \lambda_\ell \in \mathbb{R} \right. \right\}
\]

• Question: \( G(z), \Sigma(z), \Delta(z) \in S \)?
Well-posedness

**Theorem** ([L., Lindsey, Schneider, in preparation])

If $\Delta^{(p)} \in \mathcal{S}$, then in the next iteration $\Delta_{\text{new}}^{(p)} \in \mathcal{S}$.

\[
(h^0)^{(p)}, \Delta^{(p)}(z) \rightarrow G_{\text{imp}}^{(p)}
\]

\[
\Sigma^{(p)}(z) = [z - (h^0)^{(p)}] - [G_{\text{imp}}^{(p)}(z)]^{-1}
\]

\[
\Delta_{\text{new}}^{(p)}(z) = [z - (h^0)^{(p)}] - \Sigma^{(p)}(z) - [G_{\text{latt}}^{(p)}(z)]^{-1}
\]

\[
\Sigma(z) \approx \bigoplus_{p=1}^{P} \Sigma^{(p)}(z)
\]

\[
G_{\text{latt}}(z) = (z - h^0 - \Sigma(z))^{-1}
\]
Sketch of the proof

• Step 1. Lehmann representation

\[ \Delta^{(p)}(z) \in S \rightarrow G_{imp}^{(p)} \in S \]

• Step 2.

\[ G_{imp}^{(p)} \in S \rightarrow \left( G_{imp}^{(p)} \right)^{-1}, \Sigma(p) \in ? \]

Proposition

Let \( \cap_{k=1}^{K} \ker(X_k) = \{0\} \),

\[ f(z) = \sum_{k=1}^{K} \frac{X_k}{z - \varepsilon_k} \in S. \]

Then there exists a holomorphic \( g(z) := zM + C, M > 0, \) and \( g(z) - f^{-1}(z) \in S. \)
Sketch of the proof

\[
(G_{\text{imp}}^{(p)})_{11}^{-1} = z - C - f(z), \quad \Sigma^{(p)} = C' + f(z), \quad f(z) \in S
\]

• Step 3. Prove the Schur complement form

\[
\Delta_{\text{new}}^{(p)}(z) = h_{12} \left( [z - h^0 - \Sigma(z)]_{22} \right)^{-1} h_{21}
\]

Proposition

Let \( g(z) = zM + C - f(z) \), where \( f \in S, M > 0 \).
Suppose that \( \bigcap_{k=1}^K \ker (X_k) = \{0\} \). Then \( g^{-1} \in S \)

• Hence \( \Delta_{\text{new}}^{(p)} \in S \)
Continued fraction representation

- Continued fraction along the iteration

\[ \Delta_{\text{new}}^{(p)}(z) \approx D_0 \frac{1}{z - E_0 - D_1 \frac{1}{z - E_1 - D_2 \frac{1}{z - E_2 - \ldots}} \cdots D_2^*} \]

- Limit may not be in \( \mathcal{S} \)

- Matrix product states [Wolf et al, PRX 2015] (single-site ase)

- Cluster-DMFT: Matrix orthogonal polynomials (MOP) and block Lanczos method.

- Moment matching instead of broadening

[L., Lindsey, Schneider, in preparation]
Luttinger-Ward functional


\[ \Omega[G] = \Phi[G] - \text{Tr} \left( (G_0^{-1} - G^{-1}) G \right) + \text{Tr} \log(-G) \]

- Single line for DMFT

\[ \Phi[G] = \sum_{p=1}^{N_p} \Phi^{(p)}[G^{(p)}], \quad \Sigma[G] = \Sigma^{(p)}[G^{(p)}], \quad \Sigma^{(p)}[G^{(p)}] = \nabla \Phi^{(p)}[G^{(p)}]. \]
Does Luttinger-Ward functional exist?

Nonexistence of the Luttinger-Ward Functional and Misleading Convergence of Skeleton Diagrammatic Series for Hubbard-Like Models

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• $\Sigma[G]$ is not single valued.

• Also [Gunnarsson et al PRL 2017] [Vucicevic, Wentzell, Ferrero, Parcolelet, 2018]

• Still controversial
Does Luttinger-Ward functional mean anything?

Variational structure of Luttinger–Ward formalism and bold diagrammatic expansion for Euclidean lattice field theory

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Edited by George Papanicolaou, Stanford University, Stanford, CA, and approved January 24, 2018 (received for review November 29, 2017)

\begin{itemize}
  \item Euclidean lattice field model (Gibbs measure)
  \end{itemize}

\begin{equation}
Z = \int_{\mathbb{R}^N} e^{-\frac{1}{2} x^T A x - U(x)} \, dx
\end{equation}
Gibbs measure

\[ Z = \int_{\mathbb{R}^N} e^{-\frac{1}{2} x^T A x - U(x)} \, dx \]

- Quartic potential (mimicking Coulomb interaction)

\[ U(x) = \frac{1}{8} \sum_{i,j=1}^{N} v_{ij} x_i^2 x_j^2 \]

- Two-point correlator (Green’s function)

\[ G_{ij} = \frac{1}{Z} \int_{\mathbb{R}^N} x_i x_j e^{-\frac{1}{2} x^T A x - U(x)} \, dx \equiv \langle x_i x_j \rangle \]
Bold diagrams

- “thin-line” diagram: expand with $G_0$

- “thick-line” diagram ("bold" diagram): expand with $G$

\[ \begin{align*}
\text{[Diagram]} &= \text{[Diagram]} + \text{[Diagram]} + \text{[Diagram]} + \ldots
\end{align*} \]

- Partial resummation to infinite order
Green’s function insertion
Bold diagram simplifies self energy

- Just 2 terms for 2\textsuperscript{nd} order expansion!

- All bold diagrams are 2-particle irreducible (2PI)

- Luttinger-Ward formalism [Luttinger-Ward 1960]: perturbative argument
Constrained minimization

**Theorem (Variational structure)**

\[ \Omega[A] = \inf_{G \in S_{++}^N} \left( \frac{1}{2} \text{Tr}[AG] - \mathcal{F}[G] \right) \]

where

\[ \mathcal{F}[G] = \sup_{\rho \in \mathcal{G}^{-1}(G)} S[\rho] - \int U \rho dx \]

Levy-Lieb, density matrix functional theory
Luttinger-Ward functional

- Luttinger-Ward functional:

\[ \Phi[G] = 2\mathcal{F}[G] - \text{Tr}[\log G] - N\log(2\pi e) \]

Does it remove singularity in general?

**Theorem** (Continuous extension)

\[ \Phi[G] \text{ extends continuously to } \partial S_+^N \]
Bold diagrams, rigorous statement

Quartic interaction $\epsilon U(x)$

**Theorem** (bold diagrammatic expansion)

$$
\Phi[G] = \sum_{n=0}^{\infty} \epsilon^n \Phi^{(n)}[G], \quad \Sigma[G] = \sum_{n=1}^{\infty} \epsilon^n \Sigma^{(n)}[G]
$$

$\Sigma^{(n)}[G]$: n-th order bold diagram. Asymptotic expansion

$$
\Phi^{(n)} = \frac{1}{2n} \text{Tr}[G \Sigma^{(n)}[G]]
$$

Bold diagram explained!
Open questions / future works

- Overlapping elements: causality
- Long range interaction
- Combine with ab initio methods. Diagrammatic methods.
- Why DMFT could be a good theory [Metzner, Vollhardt, PRL 1989] \((d = \infty)\): \(\Sigma\) decays faster than \(G\) (rigorous proof)
- Whether one can / how to fix Luttinger-Ward for fermions
- Further increase the robustness of hybridization fitting
- Symmetry (can be included in the relaxation)
- Continued fraction representation / matrix orthogonal polynomials / Matrix product states
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DOE Early Career; CAMERA; DOD MURI

Thank you for your attention!